

## ***Interactive comment on “Retrieval of stratospheric aerosol size distribution parameters using SAGE-III/ISS extinction measurements at three wavelengths” by Felix Wrana et al.***

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Comment: Summary: Variability of the natural stratospheric aerosol (SA) layer properties relevant to climate and chemistry remains an important field of active research. Wrana et al., present an approach to remotely monitor two properties of the SA particle size distribution using solar occultation measurements like those furnished by the SAGE III/ISS mission. The article reads well, clearly presents the problem and their approach to a solution. It certainly is among the first to apply such an approach to the new SAGE III/ISS data set. However, it is not clear in what way this work is different in principle from previous publications that have retrieved mono-modal lognormal size dis-

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tribution properties from multi-wavelength aerosol extinction coefficient measurements, such as Wang et al., 1994 (doi: 10.1029/JD094iD06p08435) or Bingen et al., 2004 (doi:10.1029/2003JD003518). The article is worthy of publication once the truly ‘novel’ portions are clearly defined and substantiated.

Reply: Thank you for your helpful comments. We tried to answer every comment in an appropriate way. Other methods to determine information on stratospheric aerosol PSD were certainly applied in the past (e.g. the papers by Bingen, which are cited in our paper) and they are of course also important. The novel aspect of our approach is:

- The use of more than one extinction ratio provides more robust estimates of median radius AND width of the assumed mono-modal log-normal PSD. This is possible due to the broader spectral range of SAGE III/ISS (up to 1544 nm) as opposed to SAGE II (up to 1021 nm).
- Retrieving only, e.g. median radius and fixing the width does not allow for a unique solution. As recently pointed out by Malinina et al. (2019), one extinction ratio (or one Angstrom-exponent) may lead to distributions with very different mean radii. This is not the case with the retrieval method presented in the manuscript, where unique solutions are found for almost all SAGE III/ISS measurements, except for some combinations of extinction ratios where 2 solutions are possible if very large mode width values together with particularly small median radius values are allowed.

Comment: 1. Article should include reference to Wang et al., (doi: 10.1029/JD094iD06p08435) who used multi-wavelength SAGE II aerosol extinction to retrieve SA parameters using single-mode lognormal & modified gamma representations.

Reply: Thanks for the suggestion, we included citation of this work.

Comment: 2. Abstract, first sentence: It is not clear to me what is ‘novel’ about this approach in view of previously published work.

Reply: See our response to the general comment above. We added a brief statement to the abstract and the conclusion to make the novel aspects of the described approach

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more evident.

Comment: 3. The assumption of composition is understandable in view of the stated research goal to support the investigation of the impact of volcanic eruptions on climate and atmospheric chemistry. However, analysis should be done regarding errors in composition, specifically biomass burning events that have occurred during the first three years of SAGE III/ISS operations. It would be interesting to see when the 'validity-check' with the Angstrom exponent fails. Maybe it is an indicator of a situation of improper composition assumptions.

Reply: The reviewer raises a very important point here. Indeed, the optical properties, namely the refractive indices, of aerosols coming from biomass burning events will probably differ from the ones we used assuming a composition of only sulfuric acid and water. Sadly, a detailed discussion of this topic would probably go beyond the scope of this study, as in literature a wide range of both the real (roughly between 1.34 and 1.9) and imaginary part ( $\sim 0.0082$  to  $0.468$ ) of the refractive indices of aerosol from biomass burning can be found (Bluvshtein et al., 2017, doi:10.1002/2016JD026230 ; Poudel et al., 2017, doi:10.3390/atmos8110228 ; Sarpong et al., 2020, doi:10.3390/atmos11010062). The composition and therefore the refractive index can vary based on the burning fuel, e.g. the type of wood, and other factors like burning conditions. In addition, values are usually only provided for very specific wavelengths, as opposed to the broad wavelength spectrum we would need for error calculations. Lastly these studies usually provide tropospheric values, which may not be enough to make a statement about stratospheric conditions, since at least mixing with background sulfate aerosols will probably play a role.

However, since we perturbed the refractive indices in our error calculations, we included information on the error of the median radius and mode width that resulted from a perturbation of the refractive indices by a certain value, depending on wavelength, in the manuscript. This can help to at least broadly evaluate the effect on median radius and mode width, should certain values of the complex refractive index be assumed.

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Also, following your suggestion, in Figure 1 in the attachment we show the results of the comparison of the Angstrom exponents in the same way as explained in the manuscript, but averaged for October 2017 and between  $30^{\circ}\text{N}$  and  $60^{\circ}\text{N}$  only. In this time and latitude window the effects of the North American Wildfires of 2017 are clearly visible in the aerosol parameters (such as median radius and effective radius). But the test showed roughly the same result as when we averaged over the whole data set from June 2017 to December 2019 (as in the manuscript). This confirms our thinking, that the test comparing the Angstrom exponents should not be a helpful tool to identify cases where the assumptions about the aerosol composition fails. This is because, it only tests if a comparable spectral dependence of the aerosol extinction can be obtained from the extinction coefficients provided in the SAGE III/ISS data set and from Mie calculations with the size distribution parameters which were retrieved from those extinction coefficients. Even if the refractive indices used for the lookup-table and therefore the retrieval of the particle size distribution parameters would not correctly represent the optical properties of the actual aerosol population, the output of the retrieval would be a size distribution which can reproduce the same extinction ratios or Angstrom exponents, apart from the errors due to the interpolation process itself. This is why we described the test as a validity check for how accurately "[...] the retrieval algorithm assigns median radius and mode width values to the data points resulting from the extinction ratios of the SAGE III/ISS measurements corresponding to the position within the set of curves [...]" in the manuscript.

Comment: 4. Given that the focus is volcanic eruptions, the authors should examine the case of bimodal size distributions or cases that are more representative of time following an eruption. The conditions of June 2017 were fairly unperturbed with respect to stratospheric aerosols.

Reply: Unfortunately, it is not really possible to retrieve parameters of a bi-modal PSD from the SAGE measurements, because the number of free parameters is too large. If two mono-modal log-normal modes are assumed one would need to retrieve median

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radius, width and number density for both modes, i.e. 6 parameters. The conditions of June 2017 were chosen on purpose because they best represent background conditions of the months that are available in the SAGE III/ISS data set at this point in time. While this paper is an introduction of the retrieval method itself, in a follow-up paper different volcanic eruptions during the SAGE III/ISS operation and the evolution of the stratospheric aerosol size distribution in their aftermath are supposed to be discussed.

Comment: 5. The authors have a sound approach to choosing wavelengths for the retrieval, paying attention to the quality of the SAGE III/ISS data. However, the relative uncertainties shown in Table 1 are twice as large at 1543nm compared to those at 1021nm. There should be a discussion of how the ‘increased information content’ available at 1543nm vs. 1021nm out-weighs the increased uncertainty.

Reply: Thank you for this suggestion, we agree and added a discussion about that topic in the manuscript. While the averaged uncertainties of the extinction coefficients of the 1543.92 nm channel are higher than of the 1021.2 nm channel, the “distance” between the individual curves (with a specific mode width value) of the lookup-table, which can be seen in the left panel of Figure 2 in the manuscript, is larger for the 1543.92 nm channel. Only together this distance between the curves and the measurement uncertainty (represented by the error bars in Figure 3 in the manuscript) determine how precise the retrieval is, or how big the error of the retrieved parameters is. This is why the “accuracy parameter” which we defined in L. 180 was introduced, which takes account of both factors. In Figure 2 in the attachment of this reply we averaged this accuracy parameter at each altitude of the SAGE III/ISS solar occultation data set over 3000 sunrise and sunset events while using the 1021.2 nm channel (blue line) or the 1543.92 nm channel (red line). As the figure shows, despite larger extinction coefficient measurement uncertainties, the 1543.92 nm channel is suited much better for the retrieval because of how far apart the curves of the LUT are.

Comment: 6. Line 148: “: : while avoiding potential problems: : :”

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Reply: Included.

Comment: 7. In the left panel of Fig. 2 and Fig. 3, how do you reconcile the multiple solutions near the coordinate (0.13, 2)? The narrowest distribution oscillates across several slightly wider distributions.

Reply: Sadly, there is not much we can do about it. Even though it would be better, if this ambiguity did not exist, the very small percentage of data that falls into this narrow area of multiple solutions can be flagged and excluded from analysis.

Comment: 8. Lines 165-169 mention limiting the width to  $\sigma < 2$  to cover cases in general and cites previous work showing values not exceeding 1.9. However, given the limitations of the Mie kernels, it is not clear how the previous work would not have a similar limitation as this current work. That is, does the previous work invoke a similar assumption/limitation on the distribution width?

Reply: The few previous studies that retrieved both the median radius and mode width without assuming one or the other found different ways to deal with the issue of information content of measurements and number of free parameters in the analytical expression of the size distribution. We use the extended spectral coverage of the SAGE III/ISS measurements in our method to increase the information content in our lookup-table. To illustrate this, in Figure 3 in the attachment we show the lookup-table that is the basis of our retrieval with the data points from an exemplary sunrise event observed by SAGE III/ISS. For comparison, in Figure 4 we show the same image but using the 1021.2 nm channel instead of the 1543.92 nm channel. The scales of the axes are chosen so that the error bars are roughly of the same size relative to the image, to facilitate a comparison of the lookup-table relative to the errors of the extinction ratios. It can be seen, that the broader wavelength spectrum of the SAGE III/ISS instrument makes the retrieval method presented in this manuscript feasible and robust, as opposed to e.g. SAGE II (max. wavelength 1020 nm). This may be why this kind of method was not used before and why this kind of limitation was not discussed in other

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works.

Comment: 9. Line 235: “which” instead of “wich”

Reply: Thank you for pointing out the typing error. We fixed it.

Comment: 10. Line 338: “aerosol” instead of “aerol”

Reply: Fixed.

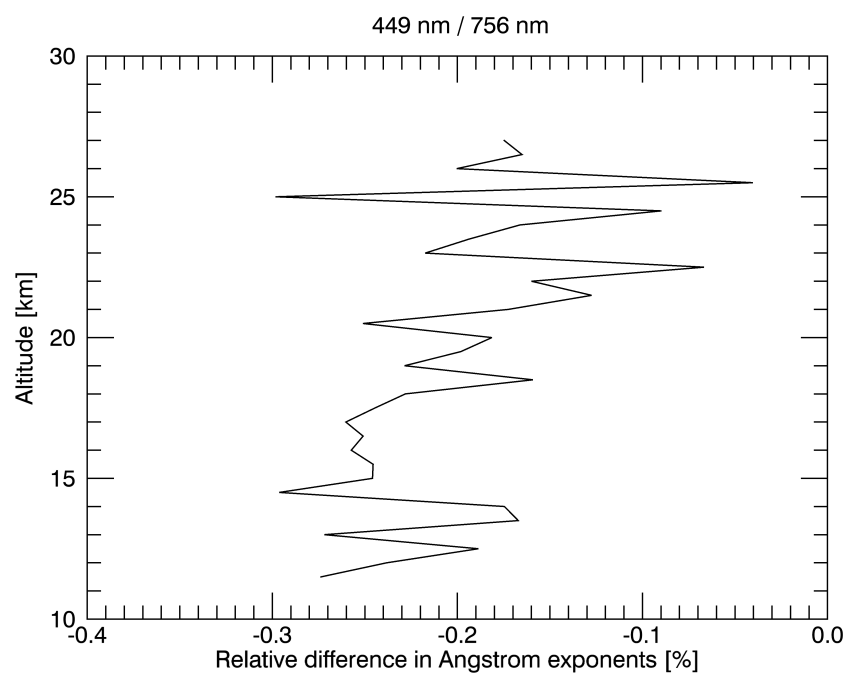
Comment: 11. Line 350 mentions “both validation methods suggest”, but it is not clear to me what method other than the Angstrom exponent computation is used for ‘validation’.

Reply: Yes, you are right, our wording was wrong here. The sentence was referring to the reasonable total errors of the size distribution parameters as well as the comparison of the Angstrom exponents. We changed it accordingly.

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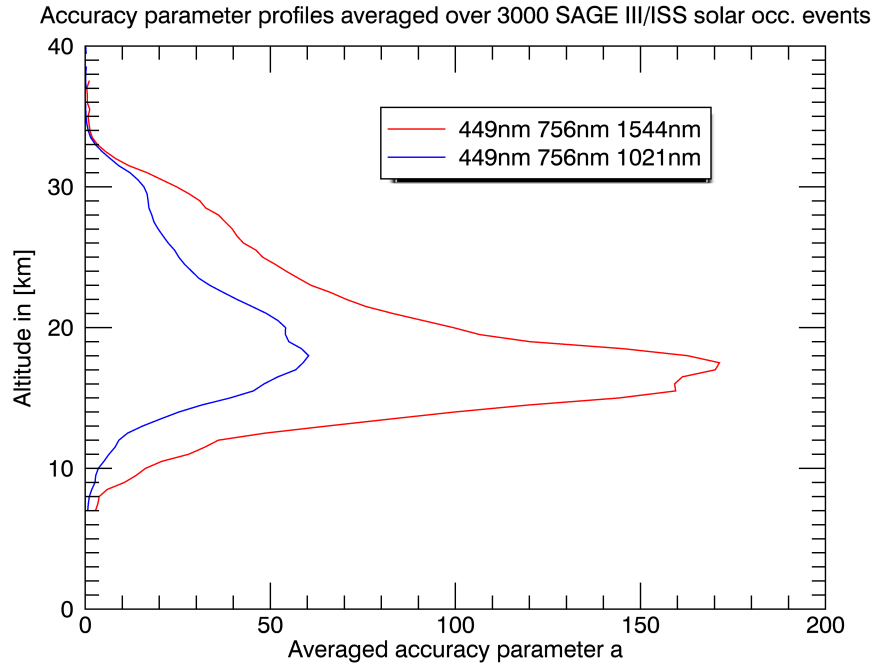
Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2020-277, 2020.

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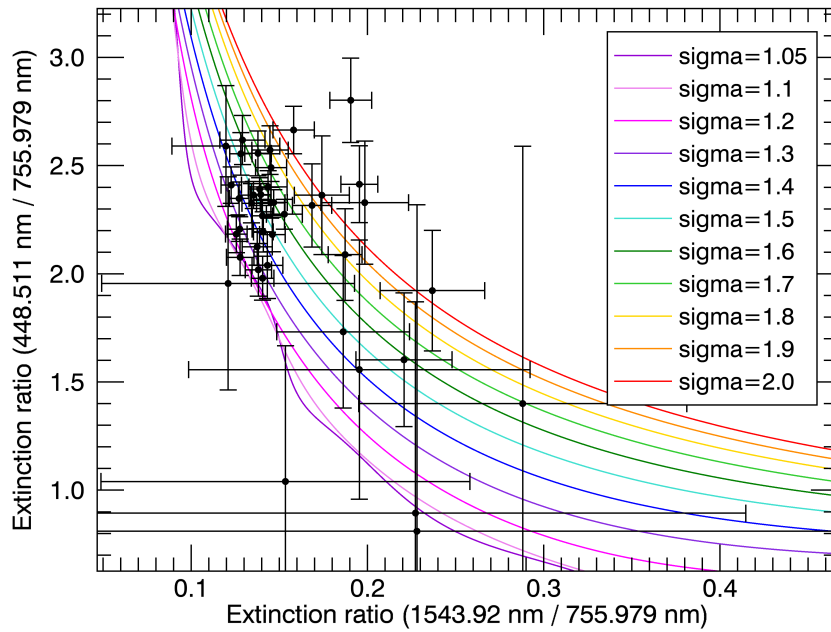
**Fig. 1.** Relative difference between Angstrom exp. from SAGE III/ISS and from Mie calculations with retrieval results averaged for october 2017 between 30°N and 60°N (Canadian Wildfires).

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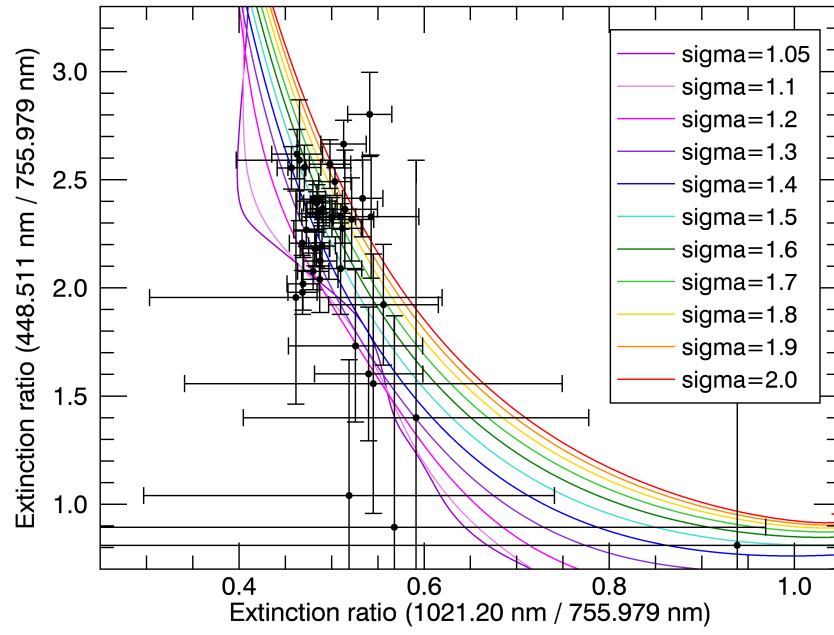
**Fig. 2.** Accuracy parameter profiles for the wavelength combinations 449nm/756nm/1544nm(red line) and 449nm/756nm/1021nm (blue line) averaged over 3000 SAGE III/ISS solar occul-tation events.

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**Fig. 3.** Lookup-table using the wavelength combination 449nm/756nm/1544nm including mea-surement data of an exemplary solar occultation event from SAGE III/ISS including error bars for comparison with Figure 4.

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**Fig. 4.** Lookup-table using the wavelength combination 449nm/756nm/1021nm including measurement data of an exemplary solar occultation event from SAGE III/ISS including error bars for comparison with Figure 3.