

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

Felix Wrana et al.

felix.wrana@uni-greifswald.de

Received and published: 2 December 2020

Comment: This paper describes an approach to retrieve the median radius and mode width of a particle size distribution supposed to be lognormal. The method is based on the use of a look-up-table of extinction ratios computed from extinction channels chosen adequately. The method is applied on measurements from the SAGE III experiment on the International Space Station (ISS). This study many similarities with the approach used by Echle et al. (J. Geophys. Res., 103, 19193, 1998), and it might be useful to cite this work which addressed aspects and issues relevant in the present context. The paper is written in a clear way and overall, the methodology and data

C1

analysis are carried out with the necessary care. The topic of this study is very relevant, but some important imprecisions should be first addressed and clarified before publication.

Reply: We thank the reviewer for his/her constructive, thoughtful and very helpful comments. We tried to answer every question in an appropriate way and implemented almost all of your suggestions. To illustrate some points we attached some figures, which below we will simply call “Figure 1” etc., while if we talk about the figures in the manuscript we will point that out specifically.

Comment: L. 20, p.1-L. 32, p.2: For the completeness, the growing importance of large fires in the feeding of the stratospheric aerosol layer should also be mentioned.

Reply: Thanks for the suggestion, we included it.

Comment: L. 30, p. 2: “while sedimentation . . . aerosol layer”: this sentence should be rephrased: sedimentation limits the averaged size, not the size of individual particles.

Reply: This sentence has been changed. It now says: “The formed aerosols grow through coagulation and further condensation, while sedimentation limits the averaged aerosol size in the aerosol layer.”

Comment: L. 30, p. 2: “Evaporation . . . temperatures”: Since the last aforementioned atmospheric layer is the troposphere, the authors should mention again that they are now considering the stratospheric altitudes.

Reply: We changed it to: “In the stratosphere, evaporation due to rising temperatures with height generally determines the upper boundary of the aerosol layer”

Comment: L. 66-68, p.3 : For the sake of completeness, the authors should also cite works by Bauman et al., (e.g. Bauman et al., J. Geophys. Res., 108, D13, 4382, 2003), and Bingen et al. (e.g. Bingen et al., Ann. Geophys., 21, 797-804, 2002).

Reply: Thank you for these suggestions. We included citations of both works.

C2

Comment: L. 116, p.5: "As a result": It is the other way around: Making the assumption that aerosol particles are spherical, the Mie theory can be used.

Reply: Yes, you are right, our wording was unclear here. We reformulated it in an unambiguous and accurate way.

Comment: L. 122-123: Is there any compelling reasons to make this hypothesis (e.g. possible convergence of an iterative process to unrealistic solutions with a mode width out of range), or is it just a matter of defining a realistic range for the LUT ?

Reply: Thank you for your question. Yes, it is in part a matter of defining a physically realistic range for the values covered by the lookup table (LUT). But mainly it is a necessity (as briefly discussed in L. 165-166) due to the behavior of the dependence of the extinction coefficients from Mie calculations on the width of the size distribution. This is illustrated in Figure 1 of the attachment of this reply. In this figure a section of the set of curves used as a LUT in the retrieval is shown. Here, it is extended by curves for larger mode width values, namely 2.1, 2.2, 2.3, 2.5, 2.7 and 3.0 to enable a more intuitive understanding of the problem mentioned in the paper. With sigma values larger than 2.0 there is an overlap of the extinction ratio curves in some areas of the field. In principle this means for the retrieval that there are two possible solutions for the median radius and mode width in these places. This is unfortunate and a limitation of the retrieval method. Firstly however, as can also be seen in Figure 1, these higher mode width values are associated with unrealistically small median radii (around 10 nm and smaller). Secondly such high mode width values are very rarely found in other works. For example, in Figure 2 a histogram of the mode width values for all in situ measurements available from the Wyoming optical particle counter (OPC) measurements, in which a monomodal log normal size distribution was found as the best fit is shown. In this data set 7.6% of the mode width values exceed 2.3, and only 2.97% exceed 2.5. This is not nothing, and has to be kept in mind when analyzing the SAGE III/ISS data set, but in our opinion does not call into question the validity of the data set generated in general. Thirdly, for the SAGE III/ISS data set analyzed so far, only

C3

very few measurements fall into the space where overlap exists, if for example 2.5 was deemed the maximum realistic value, much less so for 2.3, which is illustrated by Figures 3, 4, 5 and 6. In each of these exemplary figures all aerosol extinction ratios from the measurements of SAGE III/ISS from one sunrise or sunset event are shown. Each black dot with error bars represents the measurements at the three wavelengths at one specific tangent altitude. We made no specific selection of measurement events, and are showing the first event as well as the 500th, the 1000th and the 10000th event observed by the instrument.

Of course, the results produced by the retrieval method described in this manuscript have to be interpreted in the context of the assumptions made, as it is the case with all retrievals. Still this particular assumption had to be made to be able to retrieve the respective data. But you pointed out an important issue and we made it more clear in the manuscript.

As a separate issue, regarding your question in parentheses, we want to point out, that here may be a misunderstanding in regard to how the retrieval of the median radius and mode width values in the context of the lookup-table was carried out. The retrieval process is not iterative. For each measurement taken by SAGE III/ISS for a single altitude the retrieval method produces a median radius and a mode width by interpolation between the values of the lookup-table exactly at the position of the measurement data point. The "error bars" play no role in the retrieval itself, only in the identification and exclusion of noisy data. This means, that almost no matter where inside the field of the LUT the data point lands, the retrieval gives a unique result.

Comment: Figure 2: It is not clear, from the caption, what is the meaning of the numbers annotated in the figures ("50nm", "60nm", etc.). It is also not clear to which point these values refer. The use of arrows could help specifying the link between the values and corresponding dots (if this is the link the authors mean).

Reply: Thank you for pointing this out. We added the following description to the

C4

caption of the figure: "The dots mark r_{med} values with increments of 10 nm. The respective r_{med} values of five dots are shown in the color corresponding to their mode width."

Comment: L. 54, p. 2 and L. 131, p.6: The acronym "PSD" should be defined in L. 54, p. 2.

Reply: We implemented it.

Comment: L. 133, p. 6: n and k are determined by the assumption made in L. 114-119, p.5. This should be clarified. It might also be useful to mention, here or in the paragraph on L. 154-157, p.7, that these indexes of refractions are wavelength-dependent.

Reply: Thank you, we pointed that out in L. 133 and also implemented your second point.

Comment: L. 146-153, p. 7: In §2, it is mentioned that the extinction at 1543.92 nm is based on a 30-nm bandwidth channel. While the relative uncertainty for this channel is twice the uncertainty at 1021.20 nm (See Table 1), the uncertainty on the wavelength is much higher than in the case of the other channels, including 1021.20 nm for which the spectral resolution is probably about 2 nm (as mentioned in §2). Did the authors perform a sensitivity study to evaluate what the impact of these increased uncertainties (on both the extinction and wavelength) is, how it affects the retrieval of the PSD mode parameters, and to which extend the choice of the 1543.92 nm channel is better than the one of 1021.20 nm (if it is).

Reply: Yes, the averaged uncertainties of the extinction coefficients of the 1543.92 nm channel are higher than of the 1021.2 nm channel. However, 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

C5

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 7 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.

Regarding your second point on the uncertainty of the wavelength (if we understand your comment correctly): The uncertainty in the location of the central wavelength is very small compared to the width of the channel. We compared the extinction coefficient from Mie calculations for a fixed assumed monomodal size distribution (median radius=130 nm, $\sigma=1.6$, number density= 1 cm^{-3}) when assuming a monochromatic window exactly at 1543.92 nm with the extinction coefficient resulting from a mean value of each extinction coefficient if the Mie calculation is carried out at each wavelength across the 30 nm wide wavelength window in 0.5 nm steps. The resulting difference between both extinction coefficients was 0.015%, which supports our statement.

Comment: L. 163-164, p. 7: How does this interpolation occur ? Following the example give in Fig. 3, the solution for each tangent altitude is likely to cover a large range of σ -values. Did the author perform some regression ?

Reply: See last part of our reply to comment on L. 122-123. The interpolation was performed only for each single data point, the "error bars" play no role in the retrieval itself. Therefore, there is only one solution at each altitude.

Comment: L. 165-169, p. 7: I don't see why it is necessary to exclude solutions with large values of σ . In all cases, a range of solutions is likely to provide the set of extinction ranges, taking into account the uncertainty of the different extinction

C6

channels. Moreover, a large value of the mode width could be useful as an indication that the assumption made on the aerosol composition is not appropriate (e.g. due to the presence of clouds). In the past and in other frameworks, the exclusion of “unrealistic values” led to overlook unexpected but critical physico-chemical processes, as important as the discovery of the ozone hole. This should make the authors cautious while rejecting values.

Reply: We discuss this above in our reply to the comment on L.122-123. You raise a very good point. We are aware that values should not be excluded without good reason. It is true that not every possible combination of size distribution parameters can be found with our retrieval method. The points we make in our reply to the comment on L.122-123 hopefully explain why this assumption had to be made anyway and that it works very well at least for the conditions that can be found in the SAGE III/ISS data set so far. Each retrieval data set has to be interpreted in the context of its assumptions and this is also the case here.

Comment: L. 170, p.7-l. 184, p. 8: This argumentation is not true because it considers the response of a single particle, and not of a population of particles with a possibly large value of mode width. Hence, it does not take into account the fact that the combination of responses from all individual particles with slightly different radii “blurs” the extinction efficiency signal, in particular in the case of thin particles with respect to the wavelength. In this case, the extinction curves may be much less distinct, and the retrieval of the mode parameters, much less reliable. It should be noted that restricting the allowed range of mode width values may alleviate artificially the problem, leading again to overlook possible solutions.

Reply: We are not completely sure if we understand this comment correctly. Yes, in Figure 4 of the manuscript we only look at single particles to illustrate a general point about the impact of aerosols of different size on the extinction efficiency at different wavelengths, which is part of the calculation of the extinction coefficients for the LUT, as we discuss in L.136-138. But these lookup-tables themselves are based on extinction

C7

coefficients, which are calculated from size distributions, not single aerosol particles.

We can see that our figure and its corresponding text can be confusing in its current position in the flow of the manuscript and moved it to L.139, directly after equation 2 and also tried to word it more clearly. This way there should be a clearer separation from the lookup-tables, which are based on size distributions instead of single aerosol particles.

Comment: L. 193, p. 9: How do the authors choose the extinction channel and why ? This should be specified. Would it be meaningful to consider all of them to reduce the uncertainty?

Reply: The extinction channel at 756 nm was used to retrieve the number densities, because it has the lowest average measurement uncertainties of the three. However, the choice does not matter very much, since the difference between the results using different ones of the three wavelengths is very low, on average considerably below 1%. Thank you for pointing this out, we added this information to the manuscript.

Comment: L. 210-213, p. 10: Also the uncertainty on the wavelength might play a role, see comment on L. 146-153, p.7.

Reply: As discussed in our reply to the comment on L. 146-153 the effect of the uncertainty in the position of the center wavelength of the 1543.92 nm channel on the extinction coefficient is very small.

Comment: L. 239-243, p. 11: See comment on L. 165-169, p. 7, and L. 170, p.7-l. 184, p. 8.

Reply: We are not fully sure, which points regarding the other comments on L.165-169 and L.170-184 this comment is pointing to. Hopefully we have covered the topic in our replies to the respective comments.

Comment: L. 304, p. 15: Please indicate the geolocation of Calbuco to ease the analysis of the figure.

C8

Reply: Thank you for this suggestion. We added the information.

Comment: L. 324-325, p.16: The comparison does not only depend on the accuracy of the mode parameter retrieval, but also upon the extent to which the extinction spectral dependence for the actual aerosol population is well described by the Ångström law.

Reply: We included this in our sentence as follows: "This is a measure of the accuracy with which 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 (see Figure 3), as well as to what extent the Ångström law correctly describes the spectral dependence of the aerosol extinction."

Comment: L. 337-338, p. 17: This statement has to be qualified and reformulated: indeed, no assumption is required to retrieve the aerosol extinction, but conversely, the authors did use an assumption on the particle size (i.e. lognormal function) to derive expressions of the different mode parameters.

Reply: Perhaps the sentence is misleading. It is certainly correct that our size parameter retrieval requires assumptions (mono-modal log-normal PSD), which are probably quite strong assumptions. However, the retrieval of aerosol extinction coefficients from the solar occultation measurements does not require any a priori assumptions on the PSD. For aerosol retrievals from, e.g., limb-scatter measurements an aerosol PSD has to be assumed for extinction coefficient profile retrievals.

Comment: L. 339, p.17: The authors should specify they consider solar occultation in the present case.

Reply: Thanks for pointing this out. Stellar occultation certainly has a better geographical coverage. We replaced "satellite occultation measurements" by "satellite solar occultation measurements".

Comment: L. 346-348, p.17: I am not sure I understand this statement: if the mea-

C9

surements are not independent, off-diagonal terms of the covariance matrix have to be additionally taken into account, and the risk of systematic error may be higher.

Reply: The reviewer is correct, that the covariance terms have to be taken into account in the Gauss' error propagation law if the extinction measurements at different wavelengths are not independent. However, one of the partial derivatives the covariances are multiplied with, is negative. For that reason the total variance term will decrease if the covariances are considered.

Comment: Technical corrections L. 250, p.11: "were compared". L. 261, p.12: Did the authors check that the excluded values are not likely to be due to high aerosol load after a volcanic eruption (e.g., from their

Reply: Thank you for pointing out the typing error. Regarding the second part of the comment (a part of this comment unfortunately seems to be missing): Yes, we looked at the data that is removed by this cloud filter. It mostly removed tropospheric data without interfering with the signals which are visible after the Ambae (April and July 2018), Raikoke and Ulawun (both June 2019) eruption.

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

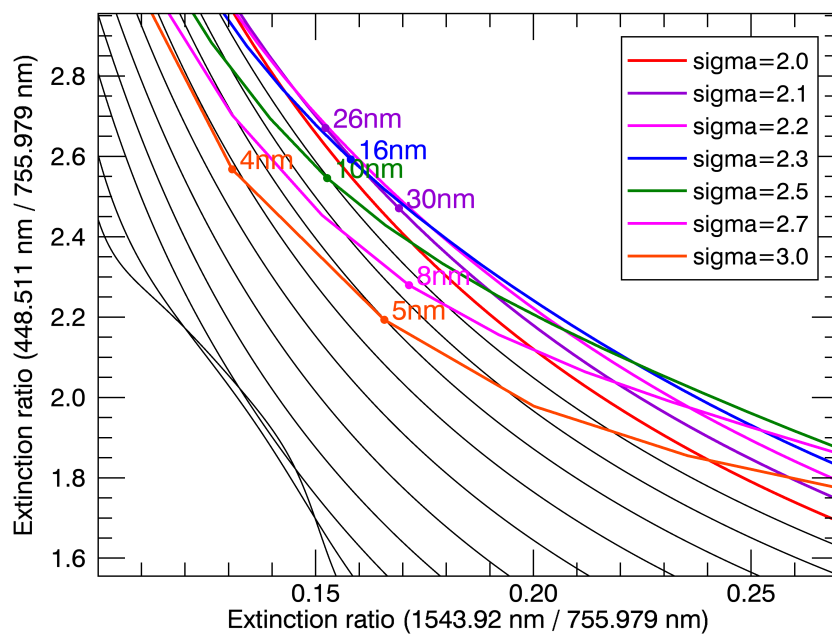


Fig. 1. Look-up-table with higher mode width values. Curves with sigma values lower than 2.0 are colored black.

C11

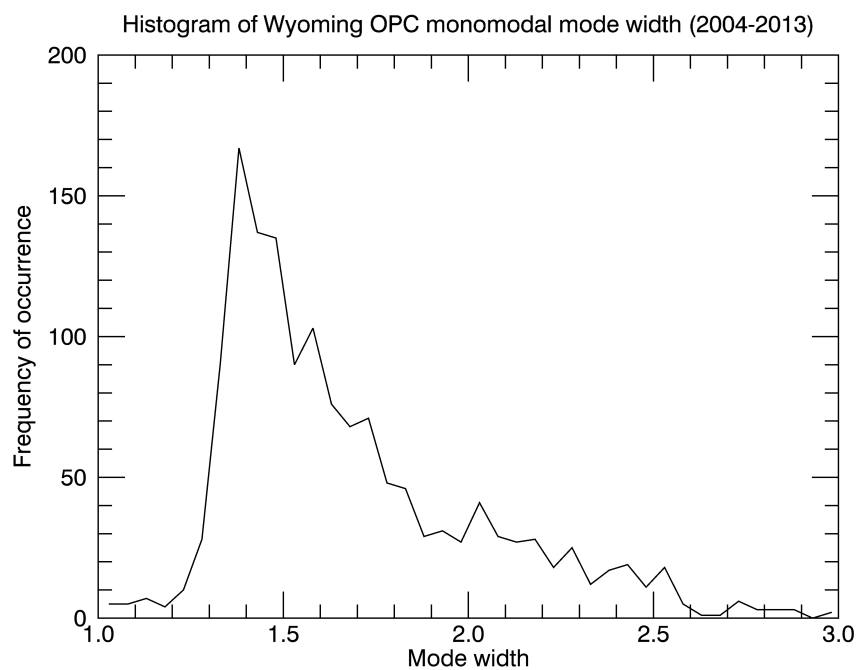


Fig. 2. Histogram of mode width from Wyoming OPC measurements for all available size distributions fitted to be monomodal. This includes data from 2004 to 2013.

C12

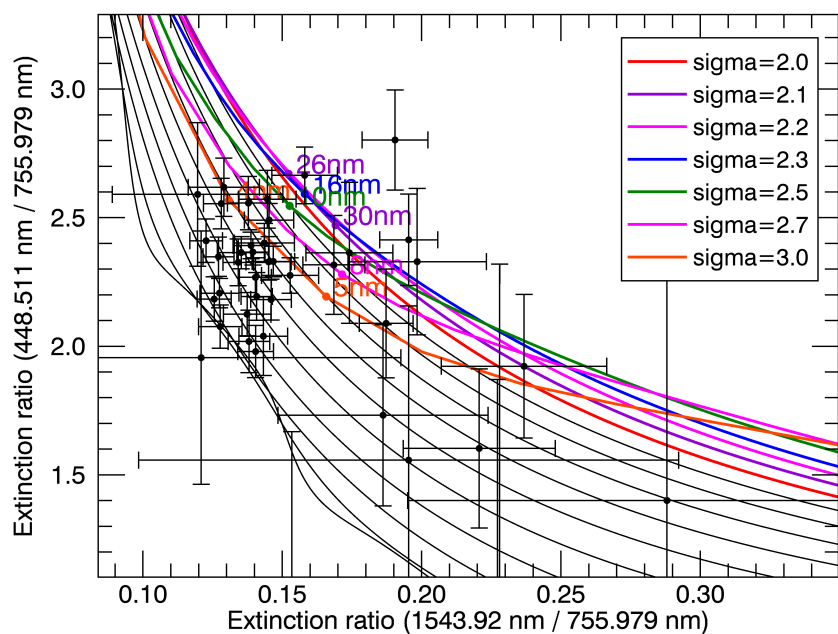


Fig. 3. Look-up-table with higher mode width values with data from an exemplary solar occultation event observed by SAGE III/ISS. The first event observed by SAGE III/ISS is shown.

C13

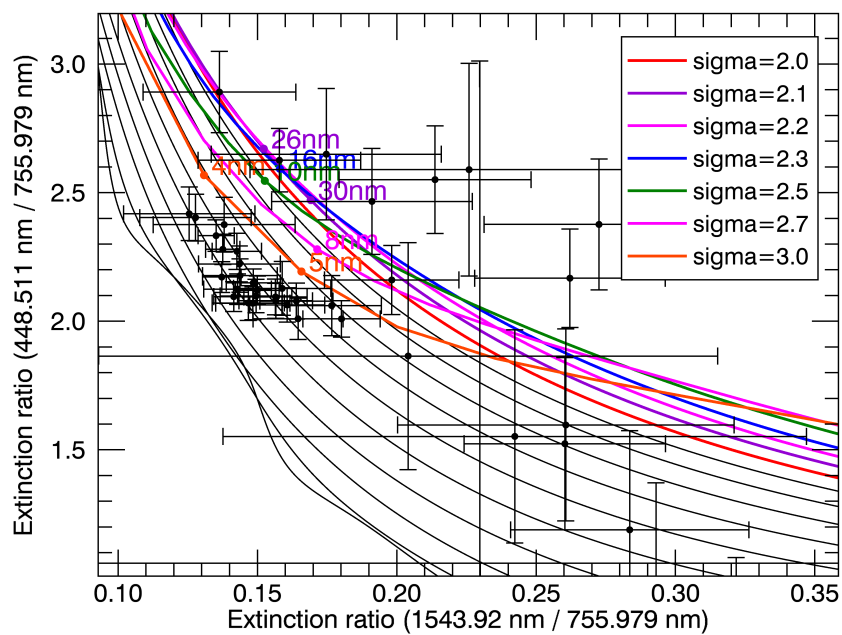


Fig. 4. Look-up-table with higher mode width values with data from an exemplary solar occultation event observed by SAGE III/ISS. The 500th event observed by SAGE III/ISS is shown.

C14

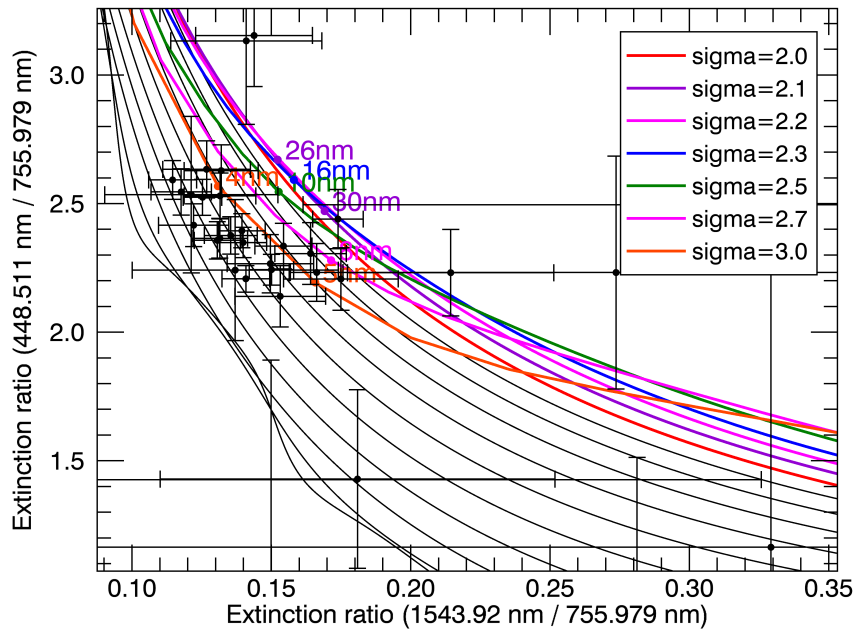


Fig. 5. Look-up-table with higher mode width values with data from an exemplary solar occultation event observed by SAGE III/ISS. The 1000th event observed by SAGE III/ISS is shown.

C15

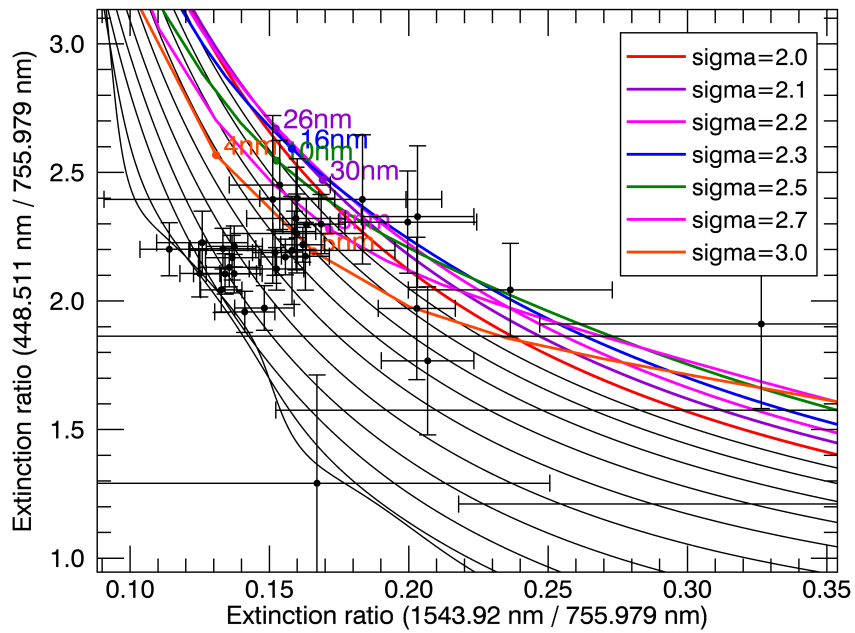


Fig. 6. Look-up-table with higher mode width values with data from an exemplary solar occultation event observed by SAGE III/ISS. The 10000th event observed by SAGE III/ISS is shown.

C16

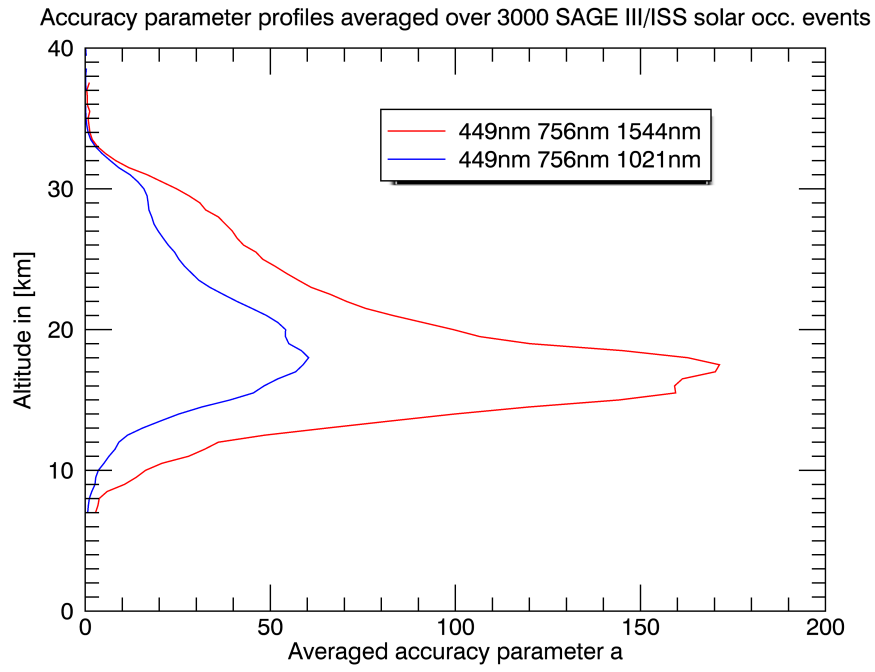


Fig. 7. Accuracy parameter profiles for the wavelength combinations 449nm/756nm/1544nm (red line) and 449nm/756nm/1021nm (blue line) averaged over 3000 SAGE III/ISS solar occultation events.