

Interactive comment on “Issues related to the retrieval of stratospheric aerosol particle size information based on optical measurements” by Christian von Savigny and Christoph Hoffmann

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Reply to comments by reviewer 1

Comment: The paper contains some useful information. But overall content of the paper is weak and would be of limited interest to the researchers in the field. Hence I do not recommend the paper for publication in its present form. In the following I highlight my main concerns. If these concerns are adequately addressed the paper may become suitable for publication.

Reply: We thank the reviewer for his/her comments, and we tried to address all the points raised in an appropriate way.

Comment: The paper discusses errors in the retrieval of aerosol particle “size” from optical measurements. This is justified in the abstract by saying that the “size” AND “size distribution” are fundamental properties of the aerosol, implying that they are two distinct quantities. Obviously, they are not. The word “size” is an ambiguous term for aerosols whose radii can vary by orders of magnitude. It is not until later in the paper one finds that by size they mean “median radius”. But why chose this quantity instead of the effective radius, a commonly used parameter in the aerosol community, defined as the area-weighted radius.

Reply: We appreciate the reviewer’s comment and fully agree that “size” is an ambiguous term. We have adjusted the manuscript such that is clear from the beginning what “size” refers to. We now also include results showing how retrievals of the effective radius behave for occultation and lidar geometries if the coarse mode fraction is varied, following the reviewer’s suggestion.

We would like to point out that we do not claim (and did not do that in the paper) that the median radius is an appropriate measure of aerosol size. The reason we use it is that it has been used in previous studies, e.g., on stratospheric aerosol particle size retrievals from the SAGE II solar occultation instruments. The main motivation of the paper was to investigate a potential reason for the finding, that aerosol particle size retrievals from solar occultation measurements seem to yield systematically larger values than retrievals from other measurements. This is essentially independent of the specific measure of aerosol size used (e.g., median, mode or effective radius). We included statements in the manuscript clarifying that we do not claim that some of the assumptions made are fully correct.

Comment: This is not just a matter of personal preference. Many investigators have found that effective radius is a robust measure of aerosol size, since it is less sensitive to the assumed particle size distribution than other parameters, such as median or modal radius. Indeed there is no scientific consensus on the median radius of aerosol particles, since microphysical models of aerosols indicate that bulk of the aerosol particles in the stratosphere (possibly more than 90 instruments, including in situ optical particle counters, are insensitive. Though these particles are important for the formation of larger particles their effect on solar radiation, and hence on climate is minimal. So, it is not clear in what sense the median radius is a “fundamental” property of aerosols.

Reply: We are well aware of the issue that optical measurements are insensitive to the smallest particles and this is exactly an important aspect of the study. The reviewer’s comment touches on some fundamental issues, which cannot be resolved with optical measurements. In order to retrieve particle size information (whatever assumptions are made) we need to assume a particle size distribution. This is also the case, if we retrieve the effective radius. If we assume a mono-modal log-normal distribution, then effective radius is related to the median radius and the distribution width via an analytical relationship. In other words, there is not a really fundamental difference between retrieving the effective radius and the median radius (assuming a width parameter).

However, we do follow the reviewer’s suggestion and now also test the effects of a changing coarse mode fraction on the effective radius and think this is a very good idea.

We also want to point out that we do not claim that the median radius is a key fundamental property of the aerosol. The particle size distribution is a funda-

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mental property in our opinion. However, we do not know what the actual size distribution is, and we will probably never know it exactly for any given case. We are well aware that the log-normal particle size distribution is a model, which will very likely differ from the actual size distribution in any given case. However, this is not a key point of the study. The main point of the paper is to investigate, how aerosol size retrievals based on a mono-modal size distribution behave, if the actual size distribution is bi-modal. We even state explicitly on page 7 of the paper: "We would like to point out that we do not claim that the actual particle size distribution of stratospheric aerosols is a bi-modal log-normal distribution."

Comment: The choice of the median radius to define aerosol "size" then leads to the paper's key conclusion that the spectral dependence of aerosol extinction cannot be used to retrieve it with high accuracy. But I am not aware of anyone who has claimed otherwise. While the spectral dependence of aerosol extinction, often condensed into Angstrom Exponent (AE), is a useful size parameter in its one right, using this information one can estimate one of the two parameters of a unimodal lognormal distribution, the modal radius (same as median radius for this distribution) or the width, by prescribing the other parameter a priori. However, it is absurd to claim any scientific validity to either parameter. The primary purpose of doing this is to estimate the effective radius under the assumption that it can be estimated robustly in spite of the inherent ambiguity in the retrieval process. The paper would have been a decent paper if the authors had chosen to focus on this issue.

Reply: We agree with the reviewer that the use of "aerosol size" is not very precise and that the median radius is not a good descriptor of aerosol size. But the appropriateness of the median radius as a descriptor of the aerosol size is not the main point of the paper. We carried out these synthetic retrievals of the median radius, because (a) this has been done in previous studies, e.g., based

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on solar occultation measurements, and (b) we wanted to investigate a potential reason for the essentially systematic high bias in median radii retrieved from these occultation measurements.

We now also test how retrievals of the effective radius from two-colour lidar and occultation measurements depend on the coarse mode fraction. Furthermore, we show the Angstrom exponents as a function of coarse mode fraction.

Comment: The authors, however, do discuss errors in the retrieval of other size related parameters that are commonly used by the aerosol community, such as surface area density(SAD). So this part of the paper is more relevant. But not adequate attention has been paid in discussing the message of the figures 4-6. For example all these figures show a monotonic relationship between particle coarse mode fraction (CMF) and the size related parameters retrieved from extinction AE. Though quantitatively they do not agree with a similar parameter estimated from another distribution, whose parameters are somewhat arbitrarily chosen, it is hard to put lot of significance to this disagreement. One doesn't know, for example what the results would have looked like had they kept the modal radius fixed and had retrieved the width, as is commonly done by the SAGE group. Also the assumption that the modal radius and width of coarse mode particle distribution doesn't vary as the CMF changes is very likely inaccurate. Finally, it would have been very useful if the authors had plotted their calculated relationship between CMF and AE. Since there is a very long history of AE measurements from SAGE, it would have provided some perspective on how often CMF greater than 1

Reply: We thank the reviewer for these useful comments. We realize that the chosen parameters may not be representative. We now test a few more cases to investigate, how dependent the retrieval results are on the a priori assumptions.

In addition, we also show the dependence of the AE on CMF, as suggested by the reviewer.

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Comment: Finally, what is most notable from plots 4-6 is the lack of monotonic relationship between CMF and size parameters retrieved from Lidar color ratio. This indicates that LIDAR color ratio doesn't contain useful information about aerosols size, irrespective of how it is defined. This should have been quite apparent had they plotted the relationship between CMF and Lidar color ratio, so there would be no need to do actual retrieval to make the point. Though this wouldn't be a surprise to the various LIDAR groups, this conclusion is important enough to other readers to be highlighted in the abstract.

Reply: Looking at Fig. 3, there is not a non-monotonic dependence between CMF and the median radius, but a monotonic one. We would like to point out that the non-monotonic relationship between volume density/surface area density and CMF results from the non-monotonic relationship between number density and CMF (Fig. 4), not from the relationship between median radius and CMF. The median radius increases monotonically with CMF in the CMF-range considered. The same is true for the effective radius (not shown in the paper). It is therefore not correct to state that the lidar color ratio contains no useful information on aerosol size.

We included a more detailed discussion of these aspects in the paper and hope this appropriately addresses the reviewer's concerns.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-342, 2019.

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