

## ***Interactive comment on “Aerosol particle size distribution in the stratosphere retrieved from SCIAMACHY limb measurements” by Elizaveta Malinina et al.***

### **Anonymous Referee #2**

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General comments on Malinina et al. (2017):

A lot of good work went into this paper, and it produces several interesting results. But the paper has a few deficiencies that appear to be major, although some may be the result of my misinterpretation. I hope that a combination of clarifying statements and perhaps reassessment of some key statements may resolve these issues.

Specific comments:

Sect. 1, 3rd paragraph:

I have a small concern with the wording of the final sentence: “Biomass burning” is cat-

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egorized as a “natural” (rather than “anthropogenic”) source of stratospheric aerosol precursors. But “biomass burning” is a general term that includes both “natural” and “anthropogenic” fires, so it straddles both categories. This sentence also would benefit from references (both for the proposal that anthropogenic activities contribute significantly to stratospheric aerosol precursors, and for the current consensus that they do not).

Sect. 1, 5th paragraph:

It might be worth mentioning at the end of this paragraph that lidar measurements provide profiles of back-scattering coefficient, which requires further interpretation to determine the extinction coefficient profile.

Sect. 2.1, 2nd paragraph:

Referring to the “horizontal direction” is probably clear enough, but referring to the across-track direction” might be clearer.

Sect. 2.2, 1st paragraph:

A reference for the statement that “space borne instruments commonly provide 2-3 degrees of freedom per altitude level” would be useful. Several space borne instruments exist that use various techniques to observe aerosols, so adding a reference would help to clarify which technique(s) are being described.

Sect. 2.3, 3rd and 4th paragraphs:

The algorithm used in this study is presented as “according to Rodgers (2000)”, and equation (4) does correspond to Rodgers (2000) equation (4.5), given the definition of  $\hat{x}$  as the deviation from the a priori value. Rodgers (2000) equation (4.5) corresponds to a linear problem, for which no iteration is necessary, but this paragraph describes the non-linearity of the present problem, which requires iteration.

The form that this iteration takes in the present work is expressed in equation (5),

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which replaces the a priori state vector with its most recent update at each step of the iteration. This appears to take the approach described by Rodgers (2000) in Sect. 5.6.2, which is entitled “A popular mistake:” It produces slow convergence to a “non-optimal, exact” solution, rather than faster convergence to the “optimal, maximum a posteriori” solution advocated by that author.

So it appears that the solution method used in this study uses the Rodgers (2000) formalism (notation, error analysis, etc.), but in fact the retrieval might be better described as weighted least squares, with the a priori information playing a negligible role (except as a starting point for the iterations). This may be justified, but the reference to Rodgers (2000) without further qualification is misleading, and the authors should explain and justify their methodology more clearly.

One more concern about this area of the text: In the 3rd paragraph, the method is described as constraining “the state vector variance” to “1% relative to the solution from the previous iteration.” But the 4th paragraph states that the iterations stop when “the maximum difference between the components of the solution vector does not exceed 1%” (among other criteria). This is confusing: Does the first statement mean that the maximum change in the state vector is 1% for each iteration? If so, then the second statement implies that the iterations would immediately stop, so perhaps it means that the average change cannot exceed 1% for each iteration? This should be clarified.

Sect. 2.3, 6th paragraph:

The text states that “The first guess parameter values are defined as  $R_{mod} = 0.11\mu m$  and  $\sigma = 1.37$ .” This follows the description given by von Savigny et al. (2015), Sect. 2.2, which selects a “mono-modal log-normal aerosol particle size distribution with a median radius of 0.11  $\mu m$  and a distribution with of  $\sigma = 1.37$  following the in situ balloon observations by Deshler (2008)”. And Deshler (2008) shows a bi-modal log-normal size distribution for which the fine mode is characterized by  $r_1 = 0.11\mu m$  and  $\sigma_1 = 1.37$  in Fig. 3c (as well as a relatively small coarse mode, described by  $r_2 = 0.38\mu m$  and

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$\sigma_2 = 1.07$ ).

The parameters  $r_1$  and  $r_2$  are described as median radii in Deshler (2008), and are used to define the size distribution mathematically in earlier references (e.g., equation (2) of Deshler et al., 2003). This usage defines  $r_1$  as equivalent to the parameter  $r_g$  defined in equation (1) of the present work, rather than  $R_{mod}$  (as defined in the text below that equation).

It therefore seems that either:

- a) The text in this paragraph is incorrect, and 0.11  $\mu m$  is used as  $r_g$  in equation (1) rather than  $R_{mod}$ , or
- b) The aerosol radius parameter defined by von Savigny et al. (based on the earlier work of Deshler et al.) is being misused in the current work.

This should be clarified in the text, since its implications extend to the entire set of cases described later in Table 1.

Sect. 2.3, 6th and 7th paragraphs:

The 6th paragraph states that “below 12 km and above 46 km the aerosol number density is set to zero.” But the 7th paragraph describes scaling that is done to estimate the aerosol outside the retrieval range. I take this to mean that a “scaled” aerosol profile is used from 12 km to the lowest retrieved altitude, and from the highest retrieved altitude to 46 km, and zero aerosol is assumed outside those ranges – is that correct?

Sect. 2.3, 7th paragraph:

This analysis is confined to the tropics. I don’t object to this limitation, but the explanation given is that “issues related to differences in observation and illumination conditions need to be dealt with” before other bands can be considered. Some more details about these issues would be useful.

Sect. 2.3, 8th paragraph:

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This paragraph states that the “effective spectral Lambertian albedo is concurrently retrieved based on the limb radiances at the same tangent heights where aerosol particle size retrieval is performed.” More should be said about how this is done. A frequently used technique (e.g., Sect. V-G of Rault and Loughman, 2013) infers the effective albedo from radiances at tangent heights above the aerosol layer, but that is not consistent with this text.

Sect. 3.1, 3rd paragraph:

The low sensitivity to particle number density ( $N$ ) is a surprising result. As I picture it, the limb radiance for optically thin paths is roughly proportional to the aerosol scattering coefficient in the tangent layer, which is itself proportional to  $N$  (for a given size distribution). But this text (as illustrated in Fig. 1) reports that the limb radiance changes by  $\approx 10\%$  when  $N$  is doubled, throughout the aerosol layer, at 1530 nm (where the line of sight extinction should be dominated by aerosol scattering, due to weak molecular scattering and the relative lack of gaseous absorption).

The relative insensitivity of the aerosol size distribution properties to  $N$  plays a key role in the method proposed, so I must be misunderstanding something important about this question. I hope the authors can add some text to explain this point further.

Sect. 3.2, 6th paragraph:

This paragraph ends by stating that “For all the scenarios modelled surface albedo was perturbed by 0.35.” Does this mean that the initially assumed surface albedo differs from the true albedo by 0.35 in all cases?

Sect. 3.2, 8th paragraph:

This paragraph describes the relative error in retrieved properties in the sensitivity study. Later (in the last paragraph of this section), some analysis is given describing the expected variability of  $N$ , based on earlier studies. Has any similar analysis been done to estimate the expected variability of  $R_{mod}$  and  $\sigma$ ? That information would

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help put the retrieval errors into context better, but of course the lack of a long-term, global data record of these parameters is part of the motivation for this work!

Sect. 4, 6th paragraph:

This paragraph notes changes in the particle distribution function shapes (a heavier “tail”, for example) as the Manam aerosol is dispersed. Are all of these distributions single-mode log-normal? And if so, how can these apparent variations in shape occur except through variations of  $\sigma$  with time? The last sentence of this paragraph says that  $\sigma$  does not change significantly.

Sect. 5, 2nd paragraph:

The Damadeo et al. (2013) reference that describes the V7 SAGE II data refers back to Thomason et al. (2008) to describe their method of computing effective radius. That latter reference mentions log-normal size distributions, but it isn’t totally clear that they assume single-mode log-normal distribution when computing the effective radius. This may be relevant as one compares the V7 SAGE II effective radius database to the values derived in this study.

Thomason et al. (2008) also mentions that the SAGE measurements lack sensitivity to aerosols with radius  $< 100$  nm (0.1  $\mu\text{m}$ ). This might also affect the results of the comparison presented in this section, since the median radius is near 0.1  $\mu\text{m}$  for many of the size distributions considered.

Sect. 5, 5th paragraph:

The final sentence summarizes the results of this comparison by calling 30% differences “a good agreement.” What standard was used to assess how good this level of agreement is? Does it flow clearly from the estimated error in the SAGE effective radius, in combination with the stated precision of the SCIAMACHY aerosol property error estimates?

Sect. 6:

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The effective radius comparison is summarized in the last sentence, noting that agreement improves from  $\approx 30\%$  at the bottom of the stratospheric aerosol layer to better than 10% near its top. Might this be related to the fact that the aerosol distribution (as derived by Deshler et al.) is frequently bi-modal at lower altitudes, before becoming a single-mode distribution (as assumed in this study) at higher altitudes?

This leads to a more general question: Were any studies done to define how this algorithm might perform if the true aerosol size distribution is not single-mode log-normal, but instead has some other (plausible) shape? Or if the true aerosol size distribution varies with altitude?

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