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Discussions

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

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We thank the reviewers for the time they spent thoroughly reading the manuscript and constructively commenting on the paper. We answered the reviewers' questions and gave the explanations, which were needed. To distinguish the referees' comments from the author's responses, the comments are shown in italicized font and the responses are highlighted in blue.

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 categorized 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).

The paragraph and formulations have been revised.

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.

The information about the backscatter coefficient has been added.

Sect. 2.1, 2nd paragraph:
Referring to the "horizontal direction" is probably clear enough, but referring to the across-track direction" might be clearer.

Wording "horizontal direction" has been changed to "horizontal cross-track direction".

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.

The text has been modified and the references have been added.

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), 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.

We corrected the paragraph, so the Rodgers' approach is emphasized. We revised the text and stated, that our retrieval is "the weighted regularized inversion similar to 0-order Tikhonov method". We also added a justification of the choice of this method instead of the Rodgers' optimal estimation.
We agree with the reviewer, that the formulations in the paragraph about the changes of the a priori information was misleading. We have improved it, and hope, that in the revised manuscript it is clear, that the solution x_{n+1} can reach any possible value, and the variance contributes to the cost function and is not a hard constrain.

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 um 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 $\sigma_2 = 1.07$).

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 um 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.

The first guess parameters were set arbitrarily, so the parameters are $R_{mod} = 0.11 \mu m$ and $\sigma = 1.37$ (r_g=0.12). The text have been revised to avoid a possible confusion. Here we do not cite neither von Savigny et al. (2015), nor Deshler (2008), so their works have not been misused. In our algorithm the first guess parameters do not play any significant role, and our internal test have shown, that changes in the first guess parameters (e.g. R_{mod}=0.13 μm or R_{mod}=0.15 μm) influence the retrieved results by less than 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?

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The reviewer's understanding is correct. The text has been slightly modified to avoid possible confusion.

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.

The issues have been specified.

Sect. 2.3, 8th paragraph:

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.

As we show it in Fig. 3 and Sect. 3.2 at 35 km aerosol influence at the radiances is rather small, thus the information from this tangent altitude contributes mainly to the albedo retrieval, and other tangent altitudes are employed for the stability reason. This explanation has been added to the text.

Sect. 3.1, 3rd paragraph:

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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.

There was some misunderstanding about the influence of N on the intensities. In the manuscript we meant, that changes in N by 200% have the same influence on the intensities as 13% change in R_{mod} or 10% change in σ. This place in the manuscript has been revised to avoid possible confusion. As can be found in the text books on radiative transfer (e.g. Chandrasekhar,1960) for measurements of the scattered light intensity $I \sim \sigma_s N p$, where $p = f(R_{mod}, \sigma)$ is the scattering phase function and $\sigma_s = f(R_{mod}, \sigma)$ is aerosol extinction cross section. The dependency of p on R_{mod} and σ is stronger, than linear. For that reason R_{mod} and σ have stronger contribution to I, than N.

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?

We have reformulated the text of the manuscript and added in the algorithm description the initial guess for albedo (0.5), and in the Sect. 3.2 "true" modelled value (0.15).

We also have added information about the retrieved values of albedo for the synthetic retrievals.

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 σ? That information would 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!

Unfortunately, we do not have any realistic assumptions for the changes in R_{mod} and σ, there are just very rough approximations taken from Deshler (2008). These approximations were assumed to be constant the whole profile. Since with this study we intend to test the algorithm sensitivity, we don't think, that the chosen scenarios of R_{mod} and σ will bias the error assessments. The clarification has been provided in the revised manuscript.

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 σ with time? The last sentence of this paragraph says that σ does not change significantly.

For the whole study we assume unimodal log-normal distribution. By the "distribution width" in the last sentence of the paragraph we meant absolute distribution width (w),

defined in Eq. (2) of the manuscript, and not σ. Parameter σ can change, though, as we mention in the Sect. 2, σ is relative parameter and is not well suitable to describe the real width. The variations in the shape for Manam eruption occur through variation of both, R_{mod} and σ, although this variations occur in a fashion, that w is not changing drastically.

To reduce the possible confusion we have revised the manuscript and added "w" in the places, where we discuss the absolute distribution width.

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 um). This might also affect the results of the comparison presented in this section, since the median radius is near 0.1 um for many of the size distributions considered.

That is true, Thomason et al. (2008) do not mention the the shape of the distribution of stratospheric aerosols, although in Sect. 4.2 Damadeo et al. (2013) the single mode log-normal distribution is assumed for the retrieval process. Similarly, in the earlier studies on SAGE II like Thomason et al. (1997) there is a link to Yue et al., (1986), where the unimodal distribution is also specifically mentioned. For that reason we think, that comparison is consistent. SCIAMACHY and SAGE II overlap period was volcanically quiscent, so following Yue (1999) the distribution will be very well described as unimodal.

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The low sensitivity of SAGE II might have affected the comparison, and we have added this statement in the revised manuscript as one of the possible reasons for the differences.

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?

Based on the reported errors, we think, that difference within 30% with SAGE II is a good result. Also, as reviewer mentions in previous comment, in Thomason et al. (2008) the low sensitivity to the particles with radius $<$0.1 μm is emphasized. Period from 2002 till 2005 is considered to be volcanically quiscent, with a large amount of smaller particles, thus SAGE II data can contain larger uncertainty, than for the volcanically active period. For that reason, we think that the results are quite good. Important to remember, there are not so many collocations with SAGE II, so the result could be biased and to give a more precise comparison.
We added the above mentioned reasons to the revised manuscript.

Sect. 6:
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 algo-

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rithm 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?

The different shape in the lower and upper altitudes should not have influenced the comparison, because both SCIAMACHY and SAGE II assume unimodal log-normal particle size distribution, and the overlap period is considered to be volcanically quiscent, when the coarse mode is quite weak.
We have not conducted any studies in the algorithm performance on the bimodal distribution, but even Deshler et al. (2003) use unimodal distribution in some cases (not just at the upper altitudes). In multiple publications on the aerosol retrievals from space borne instruments unimodal distribution is considered to be quite representative. We will consider to provide the tests suggested by reviewer in the future studies.

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