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Atmospheric Measurement Techniques Discussions

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

Anonymous Referee #1

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General Comments:

This paper presents a new algorithm for the retrieval of aerosol particle size distribution (PSD) from SCIAMACHY. As a result of a sensitivity study of the three mode parameters (particle number density N, mode radius Rmod, and standard deviation σ), the authors conclude that the sensitivity is higher for Rmod and for σ than for N, and in order to alleviate the problem of limited degree of freedom, they decide to retrieve the 2 first parameters and to assign the N parameter using ECSTRA, a climatology published in the literature. This paper is an important milestone since it concerns the first PSD retrieval from limb scattering measurements. The study has been conducted

carefully, but the order of magnitude considered for the various mode parameters may be not well suited, leading, in my opinion, to erroneous conclusions. Fortunately, I don't see any reason why it would invalidate the methodology used. Further details are given below. The authors are invited to revise the English language: sentences are sometimes very long and confusing, and their structures and the use of some word is incorrect (e.g. many confusions between "a" and "the"). It might be useful to let read the manuscript by a native speaker.

Specific comments:

Abstract:

p.1, ll. 8-9: It is not true that the aerosol particle number density is unambiguous: it depends on the kind of distribution function used, and on the assumed particle composition. Which approach (PSD or extinction) is optimal depends on the use (e.g. for modelling applications), and it has to be noted that the extinction is basically expressed by the integral of the product of N by the PSD, and since the integration process smooths out all small-scale deviations, the extinction is expected to be a more robust parameter than the PSD mode parameters.

1. Introduction:

p.2, ll. 8-9: It is not true that the aerosol particle number density is unambiguous: it depends on the kind of distribution function used, and on the assumed particle composition. Which approach (PSD or extinction) is optimal depends on the use (e.g. for modeling applications), and it has to be noted that the extinction is basically expressed by the integral of the product of N by the PSD, ans since the integration process smoothes out all small-scale deviations, the extinction is expected to be a more robust parameter than the PSD mode parameters.

p.2, l.23: For the sake of completeness, the authors might specify what is the main source of OCS.

p.2, ll. 27: Please refer to other sources.

p.2, l. 27: Biomass burning can be of anthropogenic origin. Also Asian anthropogenic sources transported to the stratosphere by the Asian summer monsoon system might be cited.

p.2, ll.31-32: I don't understand this "drawback", with respect to PSD retrieval: the PSD retrieval is obviously also determined by the aerosol composition, and the fact that assumptions are made on the PSD in the forward model for limb sounding induces a bias which is obviously a drawback for PSD retrieval.

2. Instrument and applied algorithm:

p.4, ll. 24-28: The formulation is misleading. The reasons to use a unimodal lognormal function are a lack of information content and the fact that this function looks reasonable and theoretically acceptable! Ideally, if enough information was available and the aerosol composition was known in detail, a multimodal function taking into account the microphysical properties of all kind of aerosol present would be better.

p.4, l. 29: 6 independent pieces of information are needed.

p.5, l.4, 6, etc.: "linear space" and "logarithmic space" are unclear. Please revise. ("in r", "in log(r)").

p.5, l.13: "spectral information" in unclear: Please specify.

p.5, l.28 : Please define the "weighting functions" of refer to another paper. "Jacobian": of which matrix ?

p.5, l.29: how is the a priori chosen ?

p.6, ll. 5-6: There is some contradiction between "state vector obtained at the previous iteration" and "constrained to 1% relative to the solution. . .". And why the 1% constrain ?

p.6, L.17: Please add "[set] arbitrarily", or specify.

p.6, l.25: Above 35 km, it is known that all H2SO4 is only present in gas phase. It is irrelevant to retrieve sulfate aerosol above this altitude.

p.6, l.30-32: Please specify the issues.

p.6, l.33: Please explain.

3. Sensitivity studies:

p.8, ll. 3-4, p.9, l.13: Yes it is true for the values chosen here, but another point is to know if these values are chosen in an adequate way. Results published in the literature show that, during the period considered here, N varies between about 10-3 cm-3 at $\sim$20 km and 10-4 cm-3 at $\sim$30 km [Kremser et al., 2016, op. cit.] and even more in other periods [e.g. Deshler et al., 2003, op. cit.]. Taking into account the characteristic dependence of N in altitude (linear in log(N)), this means that N is multiplied by 2 over a distance of about 3 km, which is comparable with SCIAMACHY's resolution. On the other hand, the variation of Rmod and σ is much more gradual in the vertical direction. The choice of the values used in the sensitivity study illustrated in Fig. 1 seems thus not coherent over the 3 parameters. Further, some of the values chosen for Rmod are extremely small (See Bingen et al, 2004, and Deshler et al., 2003, op. cit. for comparison), and I am not fully convinced they are really representative of the typical values found in the lower tropical stratosphere.

p.8, l.11: "Particle number density can be neglected": false ! "and considered to be constant": I don't agree, as explained above. However, I don't see why this would invalidate the methodology used here: it is perfectly acceptable, in my view, to select 2 of the 3 parameters and assign N as done here.

p.8, L. 13: "increase the uncertainty for the volcanic periods": this is certainly not the right way to do: the mean value remains the most probable. I believe this might be a reason, together with the assumptions of background aerosols made for the forward model, why SCIAMACHY's results show a systematic negative bias with respect to SAGE. (See Ch. 5). p.8, l.26: What do the authors mean by "geometric variations in tropics" ?

p.8, l.31: "This is an evidence. . .". What is an evidence ?

p.9, l.4: "settings": "characteristics" ?

p.9, l.5: What do the authors mean ? Shouldn't the choice of synthetic data be independent of the instrument configuration ?

p.9-21: Why do the authors distinguish 3 types, while Table 1 gives 5 scenarios ? About "volcanic (2N)" scenario, see rem. Above.

p.9, ll.12-34: the word "(un)perturbed" is abundantly used here and this is confusing. Amongst others, the "unperturbed scenario" is perturbed by a noise perturbation. Please use a more clear formulation. In this particular case, "nominal scenario" may help.

p.9, l.21 and figs 4-8: the distributions used here may be realistic at a given location (latitude and altitude), but keeping the vertical profile of PSD constant is not realistic at all ! Please qualify.

p.10, Table 10: The author might usefully add the values of rg and w. If my understanding is right, the values of rg for the 5 scenarios are: {17, 18, 11, 6.6, 6.6}.10-3 micron, what is irrealistically small, and in the size range of condensation nuclei, which seems not realistic to describe the cases mentioned here. As mentioned in Kremser et al., 2016, only median radii > 0.2 micron are observable for instruments like SAGE. SCIAMACHY's spectral range is not very different, and this instrument is not able to observe such small particles. Values of w are found accordingly very small, as {0.12, 0.09, 3.7, 1.24, 1.24}.10-3 micron.

p.10, l.18: If my calculation of w is correct, this is not true.

p.11, ll. 7-12: See above. I don't think that such conclusions can be drawn from the synthetic cases considered here, because I am afraid that they are not representative of the reality.

4. Results and discussion:

p.11, l. 19: Clouds are expected to be composed of very large particles. Thus, why this criteria ?

p.11, l.20: This criteria seems correct in the altitude range where clouds are expected, i.e. up to 2 km above the tropopause. But if fit is applied everywhere, it is likely to exclude important signatures of volcanic plumes (See e.g. Bourassa et al., J. Geophys. Res., 115, 2010).

p.11, ll.25- and further: It is very important to realize and to mention that monthly zonal means are not well suited to describe volcanic plumes: such plume fills a very limited part of the whole space and time interval, and the averaging "dilutes" the contribution of the volcanic aerosol load. Hence, it doesn't provide a realistic value of the instantaneous aerosol features, and is always biased (very) low. Averaging is only effective for steady situations.

p.12, l.11: the amount of SO2 was significantly smaller than the other cited eruptions, but was not small. See [Bingen et al., Remote Sensing Env. , 2017] for a recent volcanoe inventory. Again, the use of monthly means dilutes the effect of the plume and biases the contribution of the eruption low.

p.12, l.30: Are these distributions individual profiles, or monthly zonal means ? This shoud be explicitely mentioned, with a reminder of the warning here above.

p.12, l.33: N=1 cm-3: base on what ?

p.13, ll. 10-12: What do the authors mean ? For Manam, the values of the green curve at large r are lower than all the other ones ! It is worth to mention that Manam erupted again on 4 March 2006, injecting significant amounts of SO2 up to 17 km height (See volcanoe inventory in [Bingen et al., Remote Sensing Env., 2017]). This new eruption might have influenced the PSD found on 31 March 2006.

5. Comparison with SAGE II:

p.14, ll. 18-25: See comment above.

6. Conclusions:

p.15: Conclusions should be revised according to what is discussed above.

p.15, l.7: The authors should avoid the word “unique”, of precise “unique so far”.

p.15, l.9-10: “did not show any distinct behaviour” is meaningless.

p.15, l.13: Please add a mention on the systematic negative bias.

Technical corrections:

p.1, l. 7: Please revise the sentence.

p.1, l.12: “The aerosol particle density is kept constant.”

p.1, l.14: “Overall” instead of “Generally” ?

p.2, l.5: “colossal” : I am not sure this word is used in this context. p.2, l.12: “for modelling the processes”

p.2, l. 23: Please remove parentheses.

p.3, l.3: “Otherwise”.

p.3, l.4: ‘unambiguously”

p.3, l.9: “Stratospheric aerosol properties”

p.3, l.11: Sentence is not correct.

p.3, ll. 19-21: Sentence is not correct.

p.3, ll. 27-29: Please revise the sentence.

p.3, l.31: “ever” instead of “possible”.

p.4, l.3: “international”; otherwise the country should be mentioned.

p.4, ll. 6: Please revise the sentence.

p.4, ll. 12-16: Please revise the sentences.

p.4, l.21: “indicating” instead of “responsible” ?

p. 4, ll. 25-26: Please revise the sentence.

p.4, l. 31: Please revise the sentence.

p.5, ll.9-10: Unclear. Please revise the sentence.

p.5 and further: “a priori” instead of “apriori” !

p.6, l.11: “the root mean square between two subsequent iteration”: unclear.

p.7, l.10: “on a statistical approach”.

p.9, l. 7: Please revise the sentence.

p.10, l.7-8: Please correct the sentence; write “w” instead of “the latter parameter” !

p.10, l.11: “it is obvious”.

p.13, l.7: “its”

p.13, l.24: Please revise the sentence.

p.14, l.15: “compared to” instead of “opposite to”.

p.15, l.7: “an instrument”

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