

Referee Report on “Stratospheric aerosol characteristics from space-borne observations: extinction coefficient and Angstrom exponent, by Malinina et al.

General comments:

This paper reports the continuation of a previous work published by Malinina et al. (2018) presenting a new dataset of particle size information retrieved from SCIAMACHY. It proposes an extension of the dataset including the Angström coefficient and the aerosol extinction coefficient in the tropical zone (20°S-20°N), as well as a sensitivity study and error analysis of their dataset. Afterward, the authors explore the link between the Angström coefficient and the parameters characterizing the particle size distribution (PSD). Based the sensitivity study, the authors claim that limb viewing instruments are more accurate for PSD retrieval than occultation instruments.

If the approach chosen by the authors provides an interesting and valuable insight into the problem of aerosol size retrieval from spaceborne instruments, I cannot agree at all with their conclusions on the sensitivity study. This conclusion is based on a too limited and biased analysis, which doesn't take (sufficiently) into account critical elements such as the impact of the bias induced by the assumptions made on the PSD in the forward model, and the influence of the covered spectral range and of the use of multiple wavelengths on the information content.

Further, the authors propose a long discussion about the usability of the Angström coefficient to derive size information and about the ill-posedness of the problem. Beyond long developments that are in some cases not really new or relevant, many arguments used in this discussion are truncated or even wrong, and this study should benefit a lot from a more accurate reading of the literature on this subject.

If the authors want to compare the capabilities of occultation and limb viewing experiments, they need to revise thoroughly their sensitivity study to take into account all aspects of the inversion problem, including what was published in the past on this topic. As presented here, most of the conclusions of the authors, including the conclusion that limb-viewing instruments are more accurate than occultation instruments cannot be drawn, and are thus basically wrong.

Detailed comments:

Abstract:

- L. 5-6, p.1, “These uncertainties can be mitigated...”: I am not sure that the mitigation is very efficient, because the assumptions made on the PSD for the forward model obviously precede the PSD retrieval, thus influences the PSD retrieval.
- L. 6-8, p.1: I don't agree with this statement. See discussion later on the body of the manuscript.

- L. 12-14, p.1: This is not a correct and complete estimate of the error on the extinction and Angstrom coefficient. This statement has to be revised. See comments on Section 5.1.
- L. 16-17, p.1: Since OSIRIS is based on the same measuring technique (limb viewing geometry) and makes use of very similar assumptions on the PSD as SCIAMACHY in the forward model, the results of this comparison have to be considered very cautiously. This should be mentioned by the users. See also remark on Section 5.3.
- L. 19-21, p.1: Also, the Angström coefficient depends on the considered spectral range. This should also be mentioned.

1. Introduction:

- L. 4-5, p.2: I don't understand this sentence: the role of stratospheric aerosols on what? There is an abundant literature about stratospheric aerosols, and stratospheric aerosols are the scope of a SPARC (Stratospheric Processes and their Role in Climate) activity called SSiRC (Stratospheric Sulfur and their Role in Climate, See Kremser et al., 2016). The authors mention many works addressing the role of aerosols in climate, in the specific case of Asian summer monsoon, in geoengineering, and many other aspects. The aerosol evolution and role in the radiative forcing of the atmosphere was also the subject of several publications (e.g. Bingen et al., Remote Sens. Env., 2017; Brühl et al., Atm. Phys. Chem., 2018). On the other hand, it is true that the current period is characterized by a particularly low amount of satellite experiments with profiling capabilities. If this is what the authors want to (rightly) emphasize, they should reword their sentence. Otherwise, they should remove this sentence.
- L. 18, p.3: GOMOS processed with the AerGOM algorithm provides the extinction coefficient in the range 300-750 nm (Vanhellemont, et al., 2016, op. cit.).
- L. 18-20, p.3: The conversion of the backscatter coefficient to extinction is not a straightforward process since it requires the knowledge of the lidar ratio, a time-, space- and aerosol composition-dependent parameter. Errors on this parameter can thus induced a large variability and a large uncertainty on the derived extinction. This fact should be mentioned in the present discussion. Several publications address this problem, e.g. Rogers et al., Atm. Meas. Tech, 7, 4317-4340, 2014.

2. Instruments and data:

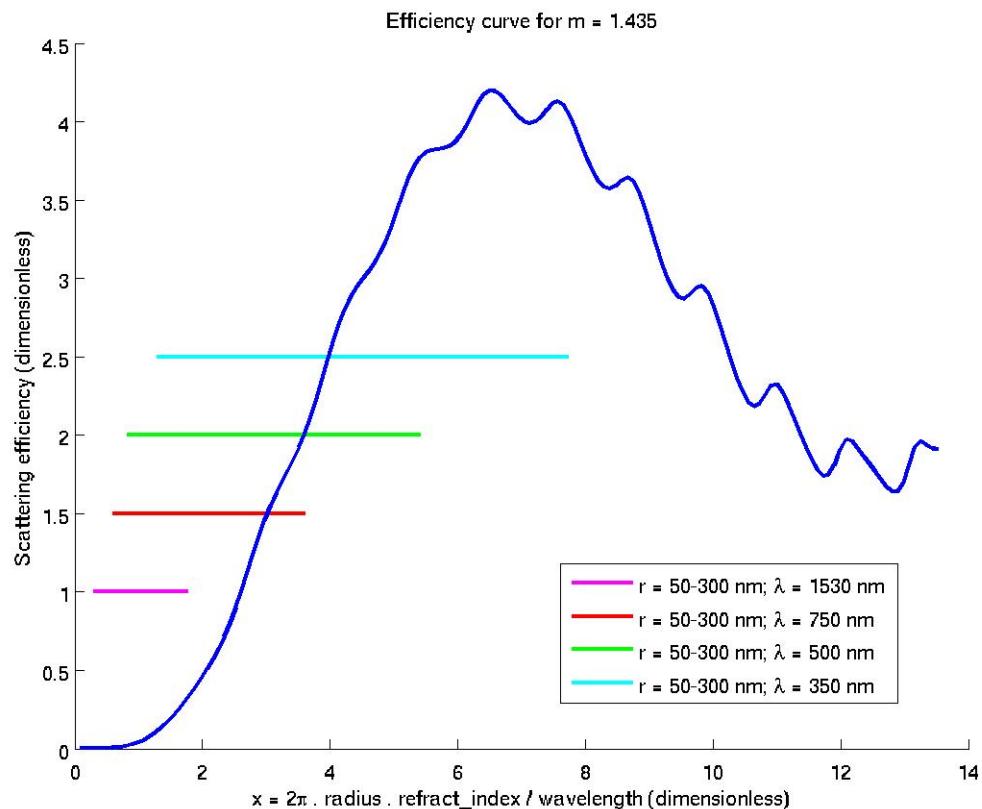
- L. 15-16, p.4: how was the fixed particle number density determined from ECSTR A? ECSTR A is a climatology of stratospheric extinction based on a parameter describing the overall volcanic state of the atmosphere, but does not provide size parameters. Please describe your methodology.
- L. 20, p.5: It should be mentioned that the approach proposed by Thomason et al, 2008 concerns the non-volcanic case.

3. Sensitivity of measurements to aerosol parameters

- L. 26-27, p.5: “unimodal” and “lognormal” are two independent concepts. The authors should add something like: “, here with a lognormal function:”
- L. 28, p.5: It might be useful to mention units, especially for the particle number density.
- L. 6-7, p.6: I don’t understand this statement. Several degrees of freedom are required to retrieve the extinction coefficient at several wavelengths. It may look like extinction retrieval requires less degree of freedom than PSD retrieval, but, as explained at the end of the section, this is due to the fact that a very significant information content is hidden in the model used, more particularly in terms of PSD assumed in the forward model, including a distribution function and the related mode parameters. Consequently, the authors should qualify this statement, make the link with the important reminder at the end of the section, and at least precise if they are only considering the case of limb viewing instruments for which additional information on the PSD is provided in the forward model.
- L. 8-9, p.6: The formulation of the extinction coefficient is not correct. It has to be written as the integration of the PSD weighted by the extinction cross-section (See, for instance, d’Almeida et al., Atmospheric Aerosols, Deepak Publishing, 1991). Eq. (3) amounts to considering that the extinction cross-section can be approximated by its value for a fixed particle radius r_{med} and a wavelength λ , and put out of the integral, what in general is not correct. The extinction cross-section is a parameter describing the extinction of light with wavelength λ by a single particle characterized by its radius and index of refraction. Hence, it doesn’t depend on aerosol mode parameters. It is very important to clarify how β_{aer} is computed because it determines how to interpret the error assessment for the extinction in §4.1.
- L. 6-8, p.7: This is confusing. Are the authors talking about the occultation case (Eq. (5))?
- L. 11-13, p.7: Since the solar scattering angle influences the phase function which depends critically on the PSD (the phase function being a weighted integration of the PSD), this issue has to be carefully investigated.
- Eq. (7) and (8): The parameter K is different in both equations and has actually different dimensions in both cases. Hence, a different notation should be used, for instance, K_{obs} and K_{scat} .
- L. 23, p.7: There is a factor π missing in the expression of K , related to the particle cross-section πr^2 .
- L. 26, p.7: “Showed it”: What did they showed?
- L. 25-27, p.7: I am not sure that these details are useful: it seems that Thomason’s formulation is different although it is the same, admittedly based on another choice of variable (volume of aerosol per volume of air, instead of particle radius). The authors might consider just mentioning that Thomason and Poole use a similar formulation instead of emphasizing the differences, in order to avoid confusion.
- L. 1, p.8: There seems to be an error in K ’s dimensions in the case of OSIRIS. If Eq. (7) is applied, K ’s units should be expressed in W (with I in W/m^2 and $n(r)$ in

number of particles per volume unit). Even if applying the expression derived by Rieger et al. (2014), a radiance factor appears in the expression. Consequently, K should include the contribution of the energy flux, and not be dimensionless.

- L.14, p7-L. 11, p.8: Overall, this discussion is a bit confusing: It seems that the aim is to show that in all cases, the inversion problem can be formulated using a similar expression, either using Eq. (7) for the occultation case, or Eq. (8) for the limb case. But immediately afterward, it is explained why this model is considered as much too simple in the OSIRIS case, and how it is not suited and will not be used for sensitivity studies in the case of SCIAMACHY. What is then the utility of this discussion?
- Eq. (9), p.8: This separation between “aerosol” and “Rayleigh” signals supposes that there is a clear distinction between both, and the authors most probably infer this Rayleigh signal from ancillary data of air density, temperature and pressure. The reality is much more complex, since very thin aerosol particles are also Rayleigh particles and their contribution to scattering cannot be discriminated from the molecular Rayleigh compound. This is especially the cases for “thin aerosol” cases considered by the authors. The only way to separate both contributions is to rely on the meteorological data that might be inaccurate with respect to the local condition encountered by the spaceborne instruments.
- L. 5-11, p.8: This model only takes into account the Rayleigh and aerosol compounds. How do the other contribution (trace gases) interfere in this analysis ?
- L. 12-13, p.8: After the previous discussion, we know that Eq. (8) will not be used, but not which model/expression/equation was actually used to quantify the sensitivity. This should be clarified.
- L. 17, p.8-L. 14, p.9: This sensitivity study is limited to the sensitivity of individual extinction channels, and this for two wavelengths in the infrared, including the 1530 nm-wavelengths, which is much higher than the particle size range the authors consider as relevant (50 -300 nm). The behaviour of the sensitivity curve S and its quantitative assessment is absolutely insufficient to assess the performances of (existing) limb viewing instruments versus occultation instruments, for several reasons:
 - If it is true that the size distribution influence twice the expression of the radiance (Eq. 6, through the scattering coefficient and the phase function), the error made by assigning inaccurate values of the aerosol mode parameters affects equally twice this parameter.



- The value of the wavelength influences significantly the scattering efficiency, as illustrated on the figure above. In particular, if the wavelength is very large compared to the particle radius, aerosol particles behave as Rayleigh particles, scattering becomes independent of the particle size (scattering $\sim \lambda^{-4}$) and the size parameter cannot be discriminated. The figure shows the dependence of the scattering efficiency as a function of the parameter $x = 2\pi m / \lambda$ (see e.g. van de Hulst, Light Scattering by Small Particles, Dover Publications, 1957), where the refraction index m is representative for a 75% H_2SO_4 -25% H_2O aerosol composition and the spectral range 350-1530 nm. It also shows, for particle size range 50-300 nm used on Figure 1 of the paper, the range of x parameter covered for the two wavelength considered by the authors ($\lambda = 750$ and 1530 nm, in red and magenta) and two other wavelengths representative for the spectral range covered by the most occultation instruments observing in the UV-visible-near IR range ($\lambda = 350$ and 500 nm, in cyan and green). It is obvious that the dynamic range corresponding to 1530 nm is particularly reduced (and similar to the Rayleigh regime). The 750 nm channel provides more variability in the scattering efficiency curve than the one at 1530 nm, but with a large overlap with the dynamic range of this first channel. On the other hand, wavelengths of 500 nm and 350 nm provide a much larger dynamic range covering almost all possible values of the scattering efficiency between 0 and more than 4 (van de Hulst, op. cit., 1957).

- The authors don't take at all the critical aspect that particle size retrieval can be retrieved by the combination of extinction values at several values. In this respect, it is clear from the figure above that the dynamic range covered by the wavelengths spread over the UV-visible-near IR range like for instruments such as SAGE II and III, POAM III, or GOMOS provides a much larger information content than the combination 750-1530 nm. This aspect is of crucial importance in the case of real aerosol particle population where the diversity of particle sizes blurs the scattering response of individual particles, especially when several aerosol modes are present simultaneously (i.e. with a large mode width of the equivalent lognormal size distribution). The larger the spectral range covered in the Mie regime, the higher the information content.

As a conclusion of this discussion, even if the calculation of the modelled sensitivity is correct, it is not sufficient to assess the performances of a technique and it doesn't provide definitive arguments to conclude about the comparison of the overall capabilities of (existing) limb viewing versus occultation instruments.

- L. 8-9, p.9: 0.1 μm is not a magic limit for the sensitivity of occultation instruments, but it corresponds to some upper limit of the Rayleigh scattering regime. It depends thus on the spectral range covered by the instrument.
- Figure 2, p.9: Both figures compare the limb and occultation geometries in a spectral range limited to wavelengths values where the occultation geometry is particularly insensitive, and that doesn't correspond to the spectral range covered by most occultation instruments (See above). It is actually visible that the sensitivity in the occultation case is increasing toward the smallest wavelengths. Consequently, this comparison is biased and it doesn't reflect the true sensitivity of real sensors used for aerosol remote sounding from space.
- L. 19-25, p.10: Taking into account the various elements cited above, I cannot agree at all with these conclusions.

4. Error assessment

- L. 15-17, p.11: The chosen scenarios cover quite nicely aerosol particle populations with radii up to about 250-300 nm. One should still bear in mind that volcanic aerosols, in view of existing estimates from satellite and balloon-borne datasets, may reach significantly higher values during particular (volcanic) periods the SCIAMACHY lifetime.
- L. 15-22, p.11: If I understand well, the errors (calculated as the "median relative error") reflects the variability obtained from an ensemble of simulations where Gaussian noise was added to the synthetic radiance. On the other hand, the synthetic scenarios are constructed using a fixed size distribution (i.e. a fixed choice of r_{med} and σ) and only N is supposed to decrease exponentially as described in L. 9, p.11. The realistic character of this profile is thus very relative. Consequently, the quantity investigated here is very different from the extinction uncertainty, that include experimental, instrumental,

modelling and other retrieval errors into account. The use of the term “error” is thus particularly misleading, and should be replaced by something more adapted.

- L. 1-4, p.12: See remark on L. 8-9, p6.
- Eq. (10), p.13: See remark on L. 8-9, p6.
- L. 22, p.13-end of p.14: Same remark as for L. 15-22, p.11.

5. Comparison of the measurements results

- L. 10-11, p.15: The difference in the measurement technique is not an issue at all ! It is the essence of validation efforts to use different datasets, including datasets based on different measurements techniques, to assess the strengths and weaknesses of the measurements to be investigated. This statement is thus inappropriate and should be removed. Arguments developed here may be pertinent to discuss the origin of possible weaknesses and strengths, but not as some kind of a priori disclaimer to relativise the adequateness or validity of a comparison exercise.
- L. 12-13, p. 15: As explained above, the sensitivity analysis proposed here is biased and insufficient, mainly because it doesn't take into account the whole spectral range taken covered by the occultation instrument, here SAGE II. Hence, even if it has been shown, indeed, that SAGE II is less sensitive to thin particles than to particles in the range $\sim 0.25-0.40 \mu\text{m}$ (See for instance Bingen et al., Ann. Geophys., 2003 for a comparison with balloonborne measurements) so that r_{eff} is indeed expected to be biased high in the case of SAGE II, a more rigorous analysis taking into account all aspects of measurements and retrieval (including the impact of the different assumptions and approximations made in the limb viewing case) is needed to draw definitive conclusions about comparisons between SCIAMACHY and SAGE II.
- L. 28, p.15: “the standard error on the mean relative difference” might be more clear.
- L.28-29, p.15: Both parameters provide different information, and the one presented should be the most appropriate to assess the quality of the agreement between datasets ! If the standard deviation is so large that it makes the figure very busy, it means that the quality of the profile-to-profile comparison is very poor, and this should also be shown in some way !
- L. 33-34, p.15: I don't see why a similar behaviour is expected for the mean relative difference of the extinction at 1020 nm and r_{eff} : the extinction at 525 nm, which shows a quite different behaviour on Figure 5, plays an equally important role in the computation of SAGE II's effective radius (Thomason et al., 2008, op. cit.).
- L. 34, p.15: What is the usefulness of the reference to Sect. 2.3 ? There is nothing more about r_{eff} in Sect. 2.3 than what is said here. A reference to some paper where the derivation of r_{eff} is presented would be more useful.
- L. 4-10, p.16: This figure only shows that there is a bias of about 8% between the two ways used to derive the extinction coefficient from the PSD. This is very different from providing any assessment of the error on the extinction

coefficient. Further, the uncertainty on the PSD has, to my knowledge, not been correctly characterized: available “errors” (Malinina et al., op. cit., 2018) are derived in a similar way as the extinction “error” in the present paper and similarly express a variability with respect of an ensemble of more or less realistic synthetic cases (See remark on L. 15-22, p.11). Hence, I am not very sure Figure 6 adds more information on the extinction uncertainty, other than emphasizing some incoherence in the processing chain producing Ext, the PSD and the Angström exponent, most probably related to the successive assumptions and approximations made.

- L. 16, p. 16: What do the authors mean ? The green curve corresponding to the Ext_{1020} in Figure 5 and the blue curve corresponding to $\alpha_{525/1020}$ have completely different shapes !
- L. 2, p. 17: What do the authors mean by “the bias in this comparison” ? I guess they just mean something like “this behaviour” ?
- L. 5-7, p. 17: I am not sure that adding complexity to the problem by extending the latitudinal range will help answering this question. A better way is probably to carefully examine the impact of every assumption and approximation made in the SCIAMACHY retrieval, and its possible altitude dependent character. Using real and totally independent measurements like balloonborne OPC measurement profiles to reconstruct a forward model and every step of the retrieval might help understanding the remaining issues.
- L. 15, p.19: I don’t agree with that. As explained above, 8% is the difference that was used between the two methods used here, but it does not represent the uncertainty on the recalculated Ext_{750} . What can be concluded from this calculation is that the additional uncertainty on Ext_{750} due to the recalculation is at least 8%.
- L. 1-4, p.20: It is very likely that, beyond the same measurement technique and the same use of spectral information, the similarities between the retrieval schemes, including the assumptions and approximations made, greatly contribute to the similar performances on both datasets. Hence, both SCIAMACHY and OSIRIS datasets might present similar biases, which are impossible to discern from the comparison SCIAMACHY-OSIRIS but potentially (very) significant. In this way, the comparison might be (very) “unfair” with respect to SAGE II and lead to the possible wrong conclusion that SCIAMACHY and OSIRIS are likely to be more accurate than SAGE II. It is very important to add such a discussion point here to exclude wrong conclusions on the respective degree of reliability of SCIAMACHY, OSIRIS, and SAGE II.

6. Discussion

- L. 16, p.21-L. 3, p.22: I don’t understand the rationale used here. We are dealing here with an underconstrained problem (as stated by the authors in L. 15-16, p.21) with 3 unknown: some radius and some spread characterizing the shape of the PSD, and a measure of the number of particles characterizing its amplitude. The way used to correctly constrain the problem was to fix the particle number for Malinina et al., and to fix the spread for Rieger et al. These

are just two possible choices (amongst other possible ones), and combining the results of both approaches will not bring any additional information at all ! Further, using $\{R_{\text{mod}}, w\}$ or $\{r_{\text{med}}, \sigma\}$ are just two equivalent ways to model the same thing, as nicely illustrated by the two panels in Figure 11.

- L. 1-17, p.23: If I understand well, the authors found that there are an infinite number of solution for a PSD giving a the same spectral dependence of the extinction as one value of the Angström coefficient. Since 3 parameters are used in the PSD and only one for the Angström coefficient to describe the same kind of information (actually the spectral dependence of the measured signal), this result is quite trivial and I don't see the added value of such a long discussion. It would be much more interesting to explain how the authors intend to solve the problem in the case of SCIAMACHY. The way to deal with this problem was already addressed in the past (Echle et al., J. Geophys. Res., 103, 19993-19211, 1998; Fussen et al., Atmosph. Env., 35, 5067-5078, 2001), so that, if the authors are willing to present their own method applied to SCIAMACHY, they should at least refer to these works in the discussion.
- L. 15-18, p.23: This part of the discussion are absolutely truncated. The authors are here finding that more than one Angström coefficient, or equivalently, more than 2 extinction channels, should help deriving the PSD. There is absolutely nothing new in this, and telling that "all known space-borne instruments" provides only one Angström coefficient amounts to saying that all known space-borne instruments provides extinction at, at most, two wavelengths, what is obviously wrong. In particular, as mentioned above, extinction coefficients from occultation instruments are used over several wavelengths covering a large spectral range, what helps solving the ill-posedness of the problem (see e.g. Fussen et al., Atmosph. Env., 35, 5067-5078, 2001; Bingen et al., Ann. Geophys., 21, 797-804, 2002). Actually, this disappointed findings shows again that the comparison made here between occultation and limb viewing instruments is very incomplete, and the statement that limb instruments have a better potential or are more sensitive to the aerosol size is wrong or, at least, very premature.
- L. 18-23 p.23: After an eruption, the PSD evolves according to the microphysical processes taking place, and using a Mie model, the evolving PSD determines unequivocally the extinction behaviour, and hence the Angström coefficient through Eq. (10). The inability to derive unambiguously the PSD from the Angström coefficient obviously doesn't implies the other way around. The authors seem to claim that the behaviour of the Angström coefficient is unpredictable and depends on the detail of their formalism ("with σ remaining unchanged"), but this is obviously wrong and the point is that the exploration of Figure 11 just doesn't provide sufficient information to foresee the behaviour of the Angström coefficient after an eruption.

7. Conclusion

- Based on all what was explained above, I disagree on most of the conclusions given here. The reasons for this have been given before and don't require any

repetition. I would just mention that the “most correct conclusion” in my opinion (L. 18-19, p.24: “it is impossible to derive any reliable information (...)”) is still incorrect: the information provided by the Angström coefficient is incomplete, but is reliable if the extinction measurements used for its calculation are reliable.

Technical corrections:

- L. 11, p.2: “periods of heavy aerosol loading” ?
- L. 34, p.2: Incorrect use of the parentheses; reference “Bingen et al” could be moved after “Known existing datasets” to make the text more fluent: “Known existing PSD data sets (Bingen et al. (2004)) were obtained (...) to 2005 (Yue et al., 1989; Thomason et al (2008) etc.).
- L.30, p.3: “errors in the extinctions” or “errors in Ext”.
- L. 24, p.5: “following a lognormal distribution”.
- L. 7, p.6: “by lack of information”?
- L. 6, p.7: Incorrect sentence.
- L. 15, p.9: duplicated “the”.
- L.3, p.10: “reasonable”.
- Caption Figure 3: “The solid lines show the extinction calculated from PSD...” ?
- L. 7, p.12: “latitudinal”.
- L.8, p.13: “a subject for further studies” ?
- L. 28, p.15: The authors might add the colour used for $Ext_{750}(\alpha_{525/1020})$ to be complete.