Author's response to the Referees comments on the manuscript "Stratospheric aerosol characteristics from space-borne observations: extinction coefficient and Ångström exponent" by Elizaveta Malinina et al.

We thank the reviewers for the time they spent thoroughly reading the manuscript and commenting on the paper. We hope we have answered the reviewers' questions and improved the explanations where 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.

1 Anonymous Referee #1

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.

After the thorough reading of the reviewer's comments we realized that there was a major misunderstanding of the PSD retrieval algorithms from the limb measurements. If we understand the reviewer correctly, she/he is talking about the PSD approximation in the forward model for the aerosol extinction retrievals. However, for the PSD retrievals this assumptions are irrelevant, since there is no such intermediate step as extinction retrieval. After re-reading the manuscript, we realized that the text was misleading, so it was corrected accordingly. The misunderstanding on the algorithm also led to the misunderstanding of the Ångström exponent discussion and the sensitivity studies. With respect to the last one we agree, that the chosen wavelength interval was too short. In the revised manuscript we included shorter wavelengths. More details can be found in the "Detailed comments" section.

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.*

While working on the reviewer's comments we realized that reviewer seems to misunderstand the retrieval algorithms used by limb scatter instruments. For limb scatter instruments the radiances are used directly to retrieve PSD products and, thus, unlike occultation instruments, do not require extinction retrieval as an intermediate state. Thereby, the uncertainties are mitigated if PSD information is retrieved.

- *L. 6-8, p.1: I don't agree with this statement. See discussion later on the body of the manuscript.*

Following the discussion in the body of the text the sentence has been revised.

- *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.*

Please, refer to this section for the detailed explanation. Here, we replaced the term "error" by "parameter error" to be more precise.

- *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.*

Please, refer to the discussion to this remark.

- *L. 19-21, p.1: Also, the Angström coefficient depends on the considered spectral range. This should also be mentioned.*

The sentence has been revised. We added the information that our studies are valid for an Ångström exponent for a single wavelength pair.

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.*

The sentence has been revised.

- *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.).*

The information has been added in the text.

- *L. 18-20, p.3: The conversion of the basckatter 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.*

The uncertainties in Ext from CALIOP have been mentioned in the revised manuscript.

2. *Instruments and data:*

- *L. 15-16, p.4: how was the fixed particle number density determined from ECSTRA ? ECSTRA 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.*

The original text in the manuscript was inaccurate. N was chosen in accordance with background ECSTRA climatology. Thus, assuming unimodal lognormal distribution with r_{med}=0.11 μm and σ=1.37 N was recalculated from ECSTRA Ext values. The text of the manuscript was changed accordingly.

- *L. 20, p.5: It should be mentioned that the approach proposed by Thomason et al, 2008 concerns the non-volcanic case.*

In the paper by Thomason et al. (2008) multiple ways of surface area density calculations are presented. They include operational formula, as well as two methods for non-volcanic cases. For this reason text of our manuscript has not been changed.

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:"*

The text of the paragraph has been changed in accordance with the comments of both reviewers.

- *L. 28, p.5: It might be useful to mention units, especially for the particle number density.*

We disagree with the reviewer on this point. Since r and r_{med} can be presented in any units of length (L), N, which has a dimension of L^{-3}, can represented by any units of length as well. The range of SCIAMACHY N values, which was used in the study, is presented in Sect. 4.1.

- *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.*

We agree with reviewer that for the retrieval of Ext at multiple wavelengths, several independent pieces of information are needed. However, in that particular sentence of the manuscript we were talking about Ext coefficient at one wavelength. The sentence is there just to introduce the Eq. (3) and the link between the PSD and Ext. We have changed the sentence to make it more clear.

- *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.*

We disagree with the reviewer in that matter. The formulation of Ext is correct, in case the unimodal lognormal distribution is assumed and Mie scattering theory is used. In Mie theory, which is used for the aerosol parametrisations, the cross-sections are calculated using the integration over the given PSD.
It is true, that the usual aerosol extinction formulation is

$$Ext_\lambda = \int_0^\infty Q(r,\lambda)\pi r^2 \frac{dN}{dr} dr = \int_0^\infty \hat{\beta}_{aer}(\lambda)\frac{dN}{dr} dr,$$

where Q is the aerosol extinction efficiency, N particle number density and $\hat{\beta}_{aer}$ is aerosol extinction cross-section for the particle with radius r. However, when the integration is done over the whole particle size range with the assumption of unimodal lognormal distribution, the above described expression is transferred to

$$Ext_\lambda = \beta_{aer}(r_{med}, \sigma, \lambda)N.$$

The refractive indices for our research were calculated using OPAC database (Hess et al., 1998), which was explicitly mentioned in the cited Malinina et al. (2018) and Rieger et al. (2018) papers. However, we added the information on the assumptions on Ext calculations to the sentence marked by the reviewer.

- *L. 6-8, p.7: This is confusing. Are the authors talking about the occultation case (Eq. (5))?*

As it was written in the original text we are talking about I_{dir}, which is an occultation case. We tried to make the sentence more clear.

- *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.*

The issue has been carefully investigated in the cited papers. The text has been changed accordingly.

- *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}.*

The parameters have been changed to K_{dir} and K_{dif}.

- *L. 23, p.7: There is a factor π missing in the expression of K, related to the particle cross-section πr^2.*

According to information on the page 19 (end of the first paragraph) in the cited Twomey (1977) book, the formula is correct.

- *L. 26, p.7: "Showed it": What did they showed?*

In that sentence we meant that Thomason and Poole (1993) showed K. We revised the sentence to make it less confusing.

- *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.*

In our opinion it is important to highlight that our sensitivity studies as well as kernel assessment by Rieger et al., (2014) are not directly comparable to the study of Thomason and Poole (1993).

- *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.*

There is no error (please, see Eq. (7) and its derivation in Appendix A in Rieger et al., (2014)). Rieger et al. (2014) were not working with the radiance, but with measurement vector defined as $y = ln(\frac{I_{aer}+I_{Ray}}{I_{Ray}})$. That resulted in the dimensionless K.

- *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?*

The aim is to show which studies have been done before and why we do not follow this particular way. Additionally, it should be noted, that K is not related to the inversion problem formulation.

- *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.*

In our study we can distinguish between the aerosol and Rayleigh signal, because we use the model data. This fact is explicitly highlighted two sentences later. The reviewer is absolutely right assuming, that we calculate the Rayleigh signal from ancillary meteorological data. Since the meteorological data contains certain uncertainties, we introduce the "low sensitivity area" and justify it's threshold while discussing the results.

- *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?*

For the original study, where only 750 nm and 1530 nm were taken into consideration, the influence of the trace gases was negligible, because those wavelengths are outside of absorption lines. For the new study which includes 386 and 525 nm sensitivity, ozone climatology was taken into consideration. This information is included into the revised manuscript.

- *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.*

We could not follow what reviewer meant in this particular comment. The formula for S was introduced 3 lines above. Additionally, in the highlighted sentence we name and provide the reference to the model we used in our studies. To make it more clear we deleted the paragraph separation before this sentence.

- *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* λ^{-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 (λ= 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 (λ= 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.

If we understand the comment of the reviewer correctly, there was a misunderstanding about the algorithm. As we specifically mentioned in Malinina et al.,(2018) we do not use aerosol extinction coefficient to retrieve PSD parameters. Instead, as we wrote in Sect. 2.1, limb radiances were used directly to obtain R_{mod} and σ. Although the logarithms of the occultation radiances are proportional to aerosol extinction coefficient, for limb radiances the dependency is more complex. This fact was additionally highlighted in the Sect. 3.1 and shown with the Eqs. (5) and (6). Thus, reviewer's comment on the increased error by the wrong assignment of the aerosol mode is irrelevant for the limb radiances.
We agree with the reviewer that the spectral range presented in the original manuscript might be incomplete for the conclusions we've made. Thus, to make the study deeper we added in the

revised manuscript the sensitivity assessment at λ=386 nm and λ=525 nm. These particular wavelengths were chosen because they were employed by SAGE II instrument. Summary of our new study is following: λ=525 nm does not provide much of additional information about the smaller particles for the occultation measurements, but λ=386 nm does. Thus, in order to obtain information on smaller particles by the occultation instruments the wavelengths about 386 nm should be taken into consideration. For more details, please refer to the revised manuscript.
The reviewer assumed that we did not consider the combination of the multiple wavelengths (radiances or extinctions) to retrieve the PSD information. However, even if a combination of the wavelengths is used, e.g. to obtain r_{eff} by SAGE II (λ=525 nm and λ=1020 nm), it does not increase the sensitivity to the small particles as both of the wavelengths are insensitive to them.
At the end, we want to highlight, that this study was not performed to compare the overall capabilities of the limb and occultation instruments, but to show with a different approach the 0.1 μm sensitivity limit presented by Thomason and Pool (1993) and compare this sensitivity to the ones from limb instruments. For this purpose model studies are sufficient.

- *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.*

We agree with the reviewer that 0.1 μm is not a magic number. It is a result of the use of the combination of the radiances at λ=525 nm and λ=1020 nm by SAGE II. But it should be highlighted that for now SAGE II is the only occultation instrument providing the PSD information in the peer-reviewed publications. Thus, we do not see any contradictions.

- *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.*

We agree with the reviewer that the Fig. 2 was relatively biased with respect to the occultation measurements. As we wrote in the answer to the above mentioned comment, we added λ=386 nm and λ=525 nm, the wavelengths used by SAGE II.

- *L. 19-25, p.10: Taking into account the various elements cited above, I cannot agree at all with these conclusions.*

We revised the conclusions based on the analysis of the sensitivity of the radiances at λ=386 nm and λ=525 nm.

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.*

The only known to us datasets, which were published in the peer-reviewed journals, cover the period from 2002 to 2012 and provide PSD information, are the ones from SAGE II and OPCs. The official SAGE II provides effective radius r_{eff}, which does not have unique translation into r_{med} or R_{mod} and, thus, is not easy to interpret the particle sizes based on that value. The SAGE II dataset published in Bingen et al. (2004) covers the period before 2002. OPCs provide r_{med}, σ and N, however, the PSD is considered to be bimodal lognormal. Indeed, r_{med} for the coarse mode can increase 0.25 μm. Though, in our estimation PSD is considered to be unimodal lognormal, and in that case r_{med} is somewhere in between the fine and coarse mode fitted by OPCs. Thus, the mode radii of about 0.20 μm seem to be reasonable for the considered period. If the reviewer knows any other published PSD datasets, which are covering the period from 2002 to 2012, we kindly ask to provide the reference.

- *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.*

The reviewer understood our approach to the error assessment correctly. However, in the manuscript we assess so called parameter error (see Rodgers (2000), von Clarman et al., in preparation) rather than the full uncertainty. We revised the text to make this term more clear.

- *L. 1-4, p.12: See remark on L. 8-9, p6.*

Please, see the answer to L. 8-9, p6.

- *Eq. (10), p.13: See remark on L. 8-9, p6.*

Please, see the answer to L. 8-9, p6.

- *L. 22, p.13-end of p.14: Same remark as for L. 15-22, p.11.*

See the answer to the comment L. 15-22, p.11. We revised the text to make the term "parameter error" more clear.

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.*

We disagree with the reviewer in that matter. The difference in the measurement technique is an issue. This issue is related to the difference in the radiative transfer and as a result different sensitivity of SCIAMACHY and SAGE II to the small particles. Thus, it was known for the comparison presented in Malinina et al. (2018) that there will be differences between r_{eff}. Here, we tried to present more appropriate comparison between the instruments. We believe, that presenting comparison of extinction coefficients from SAGE II (accurate value) and aerosol extinction coefficients recalculated from SCIAMACHY PSD product is more suitable than the r_{eff} comparison. We did not try to provide the reader the impression that the comparison is irrelevant, in turn we tried to do it scientifically more correct based on the information we have.

- *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 0.25-0.40 µm (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 rigourous 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.*

We got the impression, that the reviewer misunderstood the PSD retrieval algorithm from SCIAMACHY. Again, we do not use the aerosol extinction coefficients, but the radiances. Thus assumptions on the PSD in the limb viewing case are irrelevant for that studies. Further assumptions were already addressed in Malinina et al. (2018). With respect to the sensitivity, please refer to the answers to Sect. 3 and the revised Sect. 3.

- *L. 28, p.15: "the standard error on the mean relative difference" might be more clear.*

The reviewer might have misread the sentence. In the manuscript it is "the standard error OF the mean.", which is a statistical term.

- *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 !*

Standard error of the mean is calculated as standard deviation divided by square root from the amount of observations. In the beginning of the the section we provide the amount of collocations, so the standard deviation can easily be recalculated if a reader needs this particular value. We do not show standard deviation, because at the altitude range from 24 to 31 km the relative differences are quite close to each other and the lines even cross sometimes. Thus, plotting standard deviation will make the plot unreadable.

- *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.).*

In the paper by Thomason et al. (2008) there is no formula for r_{eff} derivation provided, it is just noticed that Ext_{1020} and Ext_{525} are used to derive PSD information. In the cited in Thomason et al. (2008) paper by Chu et al. (1989) there is also no formula given. Additionally it should be noted that for surface area density it was shown that Ext_{1020} plays bigger role than Ext_{525}. Thus, we are not sure that Ext_{525} is equally important. However, even if it is the case, we expect r_{eff} to show similar behavior to the components it is calculated from, except they are not absolutely anticorrelated. And this is not the case for Ext_{525}. For that reason, we do not think that the similarities in r_{eff} and Ext_{1020} is something unexpected.

- *L. 34, p.15: What is the usefulness of the reference to Sect. 2.3 ? There is nothing more about* r_{eff} *is 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.*

In the answer to the previous comment we noted, that we could not find the exact formula for r_{eff}. In Sect. 2.3 we cite Damadeo et al. (2013) and Thomason et al. (2008), who notice that both, Ext_{1020} and Ext_{525} are used. If the reviewer knows any other paper or source of the information on how r_{eff} is calculated, we will be happy to include it into the next revision of the manuscript.

- *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.*

In this passage we haven't tried to assess errors. We exactly wanted to show the differences in recalculation of Ext_{750} with Ångström exponent in comparison to the "true" value. When comparing the measurements from different instruments providing aerosol extinction at different wavelengths, Ångström exponent is used to recalculate the extinction coefficient (e.g. von Savigny, 2015; Rieger et al., 2018). Thus, we think it is important to show numerically the incertanies in this recalculation. With respect to errors, as we noted in the above mentioned comment, we assessed the parameter error. The same as was done in Malinina et al. (2018).

- *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* $a_{525/1020}$ *have completely different shapes !*

We meant that $\alpha_{525/1020}$ is almost mirrored from Ext_{1020}. However we agree with the reviewer, that the phrasing was not correct and we revised the section.

- *L. 2, p. 17: What do the authors mean by "the bias in this comparison" ? I guess they just mean something like "this behaviour" ?*

The sentence has been changed to "The differences observed in this comparison are expected".

- *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.*

We agree with the reviewer that comparison of SCIAMACHY dataset with OPCs will be beneficial. However, the longest series of OPCs measurements is presented in Laramie, which is located in the mid-latitudes. During the SCIAMACHY lifetime there were 3 tropical OPC campaigns, however to provide an extensive comparison the Laramie dataset should be taken into consideration. Thus, to compare SCIAMACHY and OPC profiles, the existing algorithm should be applied to the mid-latitudes. Additionally, that will increase the amount of collocations with SAGE, which will allow to provide statistically significant comparisons.

- *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%.*

Please, refer to the answer to the L. 4-10 p.16.

- *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.*

Yes, both SCIAMACHY and OSIRIS use the same measurement techniques and the same wavelength information. However, the algorithms are quite different. Firstly, OSIRIS fits r_{med} and N with the assumption of σ=1.6 from the measured radiances and from that the extinction coefficients and Ångström exponent are calculated (please, see Sect. 2.2). For SCIAMACHY, N is assumed and R_{mod} and σ are fitted directly from the radiances and from those parameters aerosol extinction coefficients are calculated. So even the general way is the same (fix one of the PSD parameters $->$ get two others from the radiances $->$ recalculate extinction and Ångström exponent), the assumptions are absolutely different. Secondly, SCIAMACHY and OSIRIS have different geometries. Taking these differences into the consideration, we think that the observed differences in Ångström exponent are quite remarkable. It should be also noted, that in the highlighted by the reviewer text we emphasize the remarkable SCIAMACHY-OSIRIS consistency because of the instruments similarities and do not try to reduce SAGE II reliability.

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_{mod}, w or r_{med}, σ are just two equivalent ways to model the same thing, as nicely illustrated by the two panels in Figure 11.*

We agree with the reviewer that the presented ways to constrain the problem are not the only possible. This was presented in the manuscript in l. 2 p. 22. We also agree that combining the approaches will not add any additional information and the use of the pairs R_{mod}, w and r_{med}, σ is the equivalent way to present the same Ångström exponent. However, in the manuscript it was not stated that combination of the methods will add information and it was highlighted that both panels of Fig. 11 present the same. That's why we do not know how to address this comment.

- *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.*

It seems to be that there is a misunderstanding. The purpose of the highlighted paragraph is to show that single value of Ångström exponent can not be used as a measure of the particle size. This was shown by presenting $\alpha_{750/1530}$ as a function of R_{mod} and w and r_{med} and σ. Even though the result seems to be trivial and was briefly introduced by Ångström himself in (Angström, 1929), this is often forgotten. In the scientific community, Ångström exponent at single wavelength is quite often used to discuss about changes in the particle size. It might not be as widespread in the literature, but is quite often used at conferences and at the discussion level. For that reason, we wanted to highlight this fact again and illustrate, why this belief is wrong. As we stated already, for the SCIAMACHY retrieval algorithm there is no problem of the extinction and Ångsröm exponent spectral dependency, because unlike for the occultation measurements, we retrieve PSD parameters directly from the limb radiances. This statement was explicitly highlighted in Malinina et al. (2018) and mentioned in section 2 of this manuscript. Thus, the suggested by the reviewer references are irrelevant for this particular paragraph. We added the particular purpose of the paragraph into the manuscript text.

- *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.*

Here we meant, that usually in peer-reviewed publications and in the conference presentations the Ångström exponents at single wavelength are used directly to judge about the change in PSD, which is fundamentally wrong. As reviewer rightly notices, that can be done if multiple Ånstgröm exponents are provided. In particular that approach is helpful for the occultation instruments. However, to obtain the information on PSD from multiple Ångström exponents certain retrieval should be done. We have changed the formulations in the paragraph to make the purpose and the message more obvious.

- *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 unequivocably 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.*

The above marked paragraph is a summary of the whole section 6. In section 6 two main topics were addressed. The first addressed topic was the tropical climatology of $\alpha_{750/1530}$, which includes the discussion of the Ångström exponent changes after the volcanic eruption based on Figs. 9 and 10. While analyzing those Figs. we have noticed, that the distribution of $\alpha_{750/1530}$ after the volcanic eruption is very similar to the one of w. That led to the discussion of the second topic, i.e., the dependence of $\alpha_{750/1530}$ on the PSD parameters. Those topics are related, however they are independent in their own way. In the manuscript we never suggested to use Fig. 11 to predict the behaviour of the Ångstöm exponent after volcanic eruption. However, we understand that the formulations used in that paragraph might mislead the reader. For that reason, the paragraph was revised to resolve the issue.

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.*

We attempted to address all of the reviewer's comments. The conclusions for those comments have been changed accordingly.
The phrase the reviewer calls here as incorrect was taken outside of the context. We never meant that Ångström exponent provides unreliable information, we meant that "it is impossible to derive any reliable information on the changes in the aerosol size based solely on the Ångström exponent for one wavelength pair". The last was proven in Sect. 6.

Technical corrections:

- *L. 11, p.2: "periods of heavy aerosol loading" ?*

The marked by the reviewer phrase has been changed to "the periods with enhanced 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.).*

The sentence has been revised.

- *L.30, p.3: "errors in the extinctions" or "errors in Ext".*

The phrase has been revised.

- *L. 24, p.5: "following a lognormal distribution".*

Since there is neither grammatical, nor stylistic mistake in that phrase, we kept it unchanged.

- *L. 7, p.6: "by lack of information"?*

The suggested by the reviewer change will distort the meaning of the sentence. Because of that, it was left unchanged.

- *L. 6, p.7: Incorrect sentence.*

The sentence has been revised.

- *LL. 15, p.9: duplicated "the".*

The phrase has been revised.

- *L.3, p.10: "reasonable".*

We could not follow, what was wrong with the word "reasonable". The spelling was correct, so we kept the sentence unchanged.

- *Caption Figure 3: "The solid lines show the extinction calculated from PSD..." ?*

The sentence has been changed to "The solid lines show the Ext_{750} profiles calculated from PSD product.".

- *L. 7, p.12: "latitudinal".*

The sentence is correct. We meant altitudinal.

- *L.8, p.13: "a subject for further studies" ?*

The phrase has been revised.

- *L. 28, p.15: The authors might add the colour used for $Ext_{750}(\alpha_{525/1020})$ to be complete.*

The color has been added.

2 Anonymous Referee #2

General comments on Malinina et al. (2018):
This paper provides a useful framework for discussion of the information content of limb scattering vs. occultation measurements. The abstract states that "limb instruments have better potential for the PSD retrieval." This statement appears to be true, primarily because (as shown in the text) the scattered radiances are more sensitive to smaller particles than the transmitted radiation measured by occultation instruments. However, this conclusion is somewhat weakened by the last sentence of the abstract, which (correctly) reports that the retrieved quantity (Angstrom coefficient for a single pair of wavelengths) could correspond to an infinite number of combinations of the PSD parameters.

Based on this and next comments of the reviewer we realized, that there might be a misinterpretation of the general concept of our research. In our method, we used the retrieved PSD parameters: R_{mod} and σ (the retrieval method was presented in Malinina et al. (2018)); and then from the retrieved PSD parameters extinction coefficients and Ångström exponents were recalculated with Mie theory. We understand that this misinterpretation was caused by our formulations, so the text of the revised manuscript has been changed accordingly.

Two basic problems arise that deserve greater consideration.
1. The analysis presented throughout this paper assumes that the PSD has a single-mode log-normal shape. So the analysis shows that a set of median radius + mode width pairs could produce the same Angstrom coefficient, but confining all analysis to the single-mode log-normal possibility understates the ambiguity that actually exists: Many other types of PSD functions (bi-modal, gamma distribution, innumerable other functional shapes) exist that could also produce a given Angstrom coefficient.

We agree with the reviewer, that there is an ambiguity in the assumption of the PSD shape only by one type, and in the reality the shapes might change from one event to another. However, for our PSD parameters retrieval (see Malinina et al. (2018)), the uni-modal log-normal distribution is assumed. The study presented here is based on this particular product and, thus, is related to this particular assumption, which for now haven't been proven to be either better, or worse than the other ones, suggested by the reviewer. For that reason, we find it unnecessary to conduct studies with the other assumptions, as completely new retrieval algorithms need to be developed for this purpose, and there is not enough information to retrieve, e.g., bi-modal distribution. We changed the text of the manuscript to highlight, that our conclusions are based on our particular product and particular assumption.

2. Some work is cited that uses extinction measurements to infer PSD information, simply by assuming a type of PSD and selecting one of the many sets of PSD parameters that are *consistent with the observations + the spectral variation derived from Mie theory (Yue, 1999, for example). In many respects, the same method is used here for limb scattering, and it is not clearly quantified how the approach leads to better results when limb scattering measurements are used (granting that this is likely to be true because, once again, the scattering measurements are more sensitive to smaller particles).*

As mentioned in the first reply to the reviewer, there is a misinterpretation of the study concept. We do not use the method, which is usually applied for occultation measurements. In the previous paper (Malinina et al., 2018), which was cited in the manuscript, we state, that SCIAMACHY PSD retrieval does not use the retrieval of the aerosol extinction coefficient as an intermediate state. This particular retrieval algorithm retrieves R_{mod} and σ from the limb radiances directly. For this reason, we consider this reviewer's comment as inapplicable. The misinterpretation was caused by poor wording of the original manuscript. To make the text of the revised manuscript more clear, we added a more detailed description of the algorithm, and highlighted that the study is based on the PSD product.

Specific Comments:
Abstract:
The term "remarkable events" seems to be used as a synonym for "volcanic eruptions that perturb the stratosphere" (here and in several subsequent places). This should be clarified, perhaps by replacing that phrase with something more specific (such as "volcanic perturbations?")

By the remarkable events here and further not only the volcanic eruptions were meant, but also biomass burning events (e.g. Black Saturday in 2009) or any maintenance works or degradation of the instruments. We clarified that in the revised manuscript.

Sect. 1, 2nd paragraph:
The fact that the aerosols are assumed to be spherical should be explicitly stated here.

The assumption on the aerosol form has been added to the text.

Sect. 1, 3rd paragraph:
The second sentence ("Known existing ... (Damadeo et al., 2013)" is awkwardly worded.

The text has been revised in accordance with the reviewer's comment.

Sect. 2.1, 1st paragraph:
The solar/lunar occultation mode of the SCIAMACHY instrument is mentioned here – are those observations usable for this study? If the measurements have sufficient quality, they would certainly add to the limited number of coincident occultation + limb scattering measurements.

SCIAMACHY solar and lunar occultation measurements were done around 60° latitude in both hemispheres. Since currently the PSD product is limited only by the tropical region (20° N –20° S), there is no possibility to include those in the study. Additionally, yet there is no PSD product from SCIAMACHY occultation measurements. However, in the future the synergistic use of the both modes will be tested.

Sect. 3.1, 1st paragraph:
As discussed earlier, the failure to consider the consequences of any PSD other than single mode log-normal is a significant limitation of this study (just as it is a significant limitation of most

limb scattering aerosol retrieval work to date). Even a single example testing this approach for another type of PSD (realistic, but not single mode log-normal) would add significant value to this study, by providing some indication of how much the results presented depend on that restriction.

We chose to use uni-modal log-normal distribution because that is the one, which is assumed for our PSD retrieval algorithm. Additionally, uni-modal log-normal distribution was assumed for such instruments as SAGE II and OSIRIS. The suggested by the reviewer study is interesting as itself, but is outside of the scope of this paper.

Sect. 3.2, 5th paragraph:
It would be helpful to provide some guidance about the sigma value that corresponds to $w = 0.01$ microns for a few examples, since this is an unconventional way to describe the PSD.

The σ values corresponding to some combinations of R_{mod} and w were provided. Additionally, in Sect. 3.1 the formula of w calculation is presented (see Eq. (2)).

Sect. 3.2, 7th paragraph:
The basis for the perturbation analysis presented is confusing: Apparently the temperature and pressure are treated as variables that can be perturbed independently (without regard for hydrostatic balance, for example)?

Although the reviewer is right, that the temperature and pressure are not independent, for this particular study it does not play a major role. For SCIAMACHY PSD product, ECMWF operational analysis data for the specific date, time and location of each SCIAMACHY limb measurement were used. The same data was used in this study. This product has about 10% uncertainty for each of the variables. The other possible datasets (e.g. MERRA) have a similar uncertainty. Thus, it was decided to change pressure and temperature independently. This explanation was added in the manuscript.

Figure 9:
This figure illustrates some limitations on the conclusions that can be drawn from this analysis due to the constraints that have been imposed on the PSD during this analysis: What does it mean to perform a retrieval based on a single PSD, then analyze the variation of Angstrom coefficient with altitude (which should be zero, if a single PSD truly characterizes the stratospheric aerosol)? Or am I misinterpreting something in the methodology?

Because of the poor wording in the original manuscript, the reviewer misinterpreted the methodology of the study. The PSD parameters were first retrieved and then Ext and Ångström exponent was recalculated from it. Since the PSD varies, Ångström exponent varies as well. We added more clear explanations to the text regarding the way Ångstöm exponents were obtained.

References:
Yue, G. K.: A new approach to retrieval of aerosol size distributions and integral properties from SAGE II aerosol extinction spectra, J. Geophys. Res., 104, 27 491–27 506, 1999.

References:
Malinina, E., Rozanov, A., Rozanov, V., Liebing, P., Bovensmann, H., and Burrows, J. P.: Aerosol particle size distribution in the stratosphere retrieved from SCIAMACHY limb measurements, Atmos. Meas. Tech., 11, 2085-2100, https://doi.org/10.5194/amt-11-2085-2018,

2018.

Stratospheric aerosol characteristics from space-borne observations: extinction coefficient and Ångström exponent

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Abstract. Stratospheric aerosols are of a great importance to the scientific community, predominantly because of their role in climate, but also because accurate knowledge of aerosol characteristics is relevant for trace gases retrievals from remote sensing instruments. There are several data sets published which provide aerosol extinction coefficients in the stratosphere. However, for the instruments measuring in the limb viewing geometry, the use of this parameter is associated with uncertainties resulting from the need to assume an aerosol particle size distribution (PSD) within the retrieval process. These uncertainties can be mitigated if PSD information is retrieved. While occultation instruments provide more accurate information on the aerosol extinction coefficient, in this study, it was shown that limb instruments ~~have better potential for the PSD retrieval, especially during the background aerosol loading periods~~are more sensitive to the smaller particles in visible-near-infrared spectral range. However, the sensitivity of occultation instruments improves if the UV part of the wavelength spectrum is considered. A data set containing PSD information was recently retrieved from SCIAMACHY limb measurements and provides two parameters of the ~~log-normal~~ unimodal lognormal PSD for the SCIAMACHY operational period (2002-2012). In this study, the data set is expanded by aerosol extinction coefficients and Ångström exponents calculated from the retrieved PSD parameters. ~~Errors in the~~ Parameter errors for the recalculated Ångström exponents and aerosol extinction coefficients are assessed using synthetic retrievals. For the extinction coefficient the resulting ~~accuracy~~ parameter error is within ±25%, and for the Ångström exponent, it is better than 10%. The recalculated from PSD parameters SCIAMACHY aerosol extinction coefficients are compared to those from SAGE II. The differences between the instruments vary from 0 to 25% depending on the wavelength. Ångström exponent comparison with SAGE II shows differences between 10% at 31 km and 40% at 18 km. Comparisons with SAGE II, however, suffer from the low amount of collocated profiles. Furthermore, the Ångström exponents obtained from the limb viewing instrument OSIRIS are used for the comparison. This comparison shows an average difference within 7%. The time series of these differences do not show signatures of any remarkable events ~~.~~ (e.g. volcanic eruptions or biomass burning events). Besides, the temporal behavior of the Ångström exponent in the tropics is analyzed using the SCIAMACHY data set. It is shown, that there is no ~~simple~~ trivial relation between the Ångström exponent value at a single wavelength pair and the PSD because the same value of Ångström exponent can be obtained from an infinite number of combinations of the PSD parameters.

1 Introduction

According to the Fifth Assessment Report of IPCC (2013) (Intergovernmental Panel on Climate Change) clouds and atmospheric aerosols contribute the largest uncertainty to the estimates and interpretations of the Earth's changing energy budget. While there is a substantial number of publications and initiatives related to the ~~role of the tropospheric aerosols~~ tropospheric aerosols and their role in climate (e.g. Popp et al., 2016), the ~~role of~~ information on stratospheric aerosols is ~~currently not well addressed~~still sparse. Stratospheric aerosols influence climate through two major mechanisms. First, they scatter solar radiation and, during strong aerosol loading conditions, absorb the thermal infrared radiation upwelling from the troposphere; thus, changing the radiative budget of the Earth, and resulting in tropospheric cooling as well as stratospheric warming. As mentioned by Thomason and Peter (2006), the radiative effects of stratospheric aerosols are negligible during volcanically quiescent periods. However, after even small eruptions the influence of stratospheric aerosols on the climate becomes significant (Solomon et al., 2011; Fyfe et al., 2013). Furthermore, stratospheric aerosols play a key role in the stratospheric ozone depletion, which was reported to strengthen during the periods with the enhanced aerosol loading ~~periods~~ (Solomon, 1999; Ivy et al., 2017).

Accurate knowledge on the stratospheric aerosol loading is necessary for researchers in different fields. The atmospheric modelling community is particularly interested in this type of information, because climate models require knowledge about stratospheric aerosol to define the initial conditions and/or to assess the accuracy of their performance (Solomon et al., 2011; Fyfe et al., 2013; Brühl et al., 2015; Bingen et al., 2017). Other important applications of stratospheric aerosol data are the investigation of the effects of geoengineering (IPCC, 2013; Kremser et al., 2016) and use of stratospheric aerosol information to improve the retrieval of the stratospheric trace gases, e.g. water vapour (Rozanov et al., 2011) and ozone (Arosio et al., 2018; Zawada et al., 2018), from remote sensing instruments. Most commonly, stratospheric aerosols are characterized either by their extinction coefficient (Ext), or by one or several particle size distribution (PSD) parameters (e.g. median (r_{med}), effective (r_{eff}) or mode (R_{mod}) radius, distribution width parameter (σ), aerosol particle number density (N)). While for the instruments using the solar, lunar or stellar occultation measuring technique, Ext retrieval is quite straightforward; for limb viewing instruments the retrieved Ext is dependent on the assumed PSD parameters (more detailed in Sect. 3.2). Another complicating factor of Ext is its wavelength dependency, which is also determined by the PSD. To retrieve all parameters defining a commonly assumed ~~uni-modal log-normal PSD~~unimodal lognormal size distribution of the spherical particles, at least three independent pieces of information at each altitude level are needed. However, this requirement is usually not satisfied for space-borne measurements and some assumptions have to be made (Thomason et al., 2008; Rault and Loughman, 2013; Rieger et al., 2014; Malinina et al., 2018). Some information about PSD can be obtained from the Ångström exponent (Ångström, 1929), which describes the wavelength dependency of Ext, although this parameter, if only one wavelength pair is used, can not be unambiguously transformed into the PSD parameters.

While there are long-term data sets of the aerosol PSD parameters from Optical Particle Counters (OPCs) (Deshler et al., 2003; Deshler, 2008), for the space borne remote sensing instruments they are much more limited. ~~Known existing~~ For example, two data sets were obtained from SAGE (Stratospheric Aerosol and Gas Experiment) II, an occultation instrument which operated from 1984 to 2005 (Yue et al., 1989), (the . These data sets were described in Bingen et al. (2004); Thomason et al. (2008); Damadeo et al. (2013)), as well as . In the last publication the aerosol PSD product from SAGE III on Meteor-3M platform (2001-2005) (Damadeo et al., 2013)was also briefly mentioned. In February 2017 the successor SAGE III mission on board of ISS (International Space Station) began its operation. However, the data product description and validation results have not been published by the time of writing. Another recent aerosol PSD data set including the R_{mod} and σ (distribution width parameter) was obtained from SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY) limb data (Malinina et al., 2018). SCIAMACHY was one of the instruments operating on the Envisat satellite from 2002 till 2012 (Burrows et al., 1995; Bovensmann et al., 1999). More detailed information about the instrument can be found in Sect. 2.1. The data product v6.0 (Rieger et al., 2014) from OSIRIS (Optical Spectrograph and InfraRed Imager System) instrument on board on Odin satellite (Llewellyn et al., 2004) contains the Ångström exponent ($\alpha_{750/1530}$) from 2001 till 2012 (this instrument is described in Sect. 2.2 in more details). In addition, the theoretical basis for the retrieval of PSD parameters and Ångström exponent was presented by Rault and Loughman (2013) for the OMPS (Ozone Mapping Profiler Suite) instrument, launched in 2011 (Jaross et al., 2014) and currently operational. However, no application to the real data were reported so far. For Ext there are more existing data sets. Not only the above mentioned instruments have one or multiple Ext products (e.g. there are multiple algorithms for SAGE II (Damadeo et al., 2013, and references therein), SCIAMACHY (Ovigneur et al., 2011; Taha et al., 2011; Ernst, 2013; Dörner, 2015; von Savigny et al., 2015; Rieger et al., 2018), OSIRIS (Bourassa et al., 2012; Rieger et al., 2019) and OMPS (Loughman et al., 2018; Chen et al., 2018)), but other instruments employing limb or solar/lunar/stellar occultation measurement techniques also provide Ext at different wavelengths. For example, GOMOS (Global Ozone Monitoring by Occultation of Stars), operated with SCIAMACHY on Envisat, provides several Ext at 525 nm (Vanhellemont et al., 2016)in the range from 350 to 750 nm (Vanhellemont et al., 2016; Robert et al., 2016). In addition, the space-based lidar CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization Lidar) provides measurements of the aerosol backscatter coefficient, which is then converted to Ext. The Though the conversion of the backscatter coefficient to Ext is not straightforward and contains often high uncertainties related to e.g. lidar ratio, the Ext profiles from CALIOP have the highest vertical resolution among the space borne instruments , but comparably sparse horizontal sampling (Vernier et al., 2011).

Since there are several continuous Ext data sets which cover a wide time range, there are multiple comparison and merging possibilities for the evaluation of the long-term global behavior of stratospheric aerosols. SAGE II is considered to be one of the most reliable instruments in the era of occultation measurements, as it provided high-quality data for over 20 years, including the period during and after the Mount Pinatubo eruption, the strongest volcanic eruption of the last decades. For that reason, this instrument is often used for merging and comparison activities (e.g. Thomason and Peter, 2006; Thomason, 2012; Ernst, 2013; Rieger et al., 2015; Kovilakam and Deshler, 2015; von Savigny et al., 2015; Kremser et al., 2016; Rieger et al., 2018; Thomason et al., 2018).

In this manuscript we focus on the comparison of the Ångtröm exponents derived from SCIAMACHY, OSIRIS and to some extent from SAGE II measurements. Furthermore, we present an evaluation of the errors in the parameter errors in Ext and Ångtröm exponents, derived from SCIAMACHY PSD product. The manuscript has the following structure: Sect. 2 describes instruments and data used in the study, Sect. 3 presents an assessment of the limb and occultation instruments sensitivity to aerosol particles of different sizes. Sect. 4 includes the error assessment of the derived Ext at different wavelengths, as well as the errors of the Ångström exponents calculated from the derived Ext. In Sect. 5 comparison of the Ångström exponents from SCIAMACHY, OSIRIS and SAGE II is presented. The behavior of the Ångström exponents after the volcanic eruptions and the dependency of this parameter on the PSD parameters are discussed in Sect. 6.

2 Instruments and data

2.1 SCIAMACHY

SCIAMACHY was one of the instruments on the European Environmental satellite (Envisat), launched into the sun-synchronous orbit at 800 km altitude in March 2002 and operated till the loss of the contact in April 2012. SCIAMACHY made measurements in nadir, limb and solar/lunar occultation modes in 8 spectral channels, covering the spectral interval from 214 to 2386 nm with spectral resolution from 0.2 to 1.5 nm depending on the wavelength, and provided daily solar irradiance measurements. More detailed information can be found in Burrows et al. (1995); Bovensmann et al. (1999); Gottwald and Bovensmann (2011).

In this study we focus on the measurements performed in the limb viewing geometry. In this measurement mode the instrument scanned the atmosphere tangentially to the Earth's surface in the altitude range from about 3 km below the horizon, i.e., when the Earth's surface is still within the field of view of the instrument, up to about 100 km with a vertical step of 3.3 km and vertical resolution of 2.6 km. From SCIAMACHY limb observations two aerosol products were retrieved. One of them contains Ext at 750 nm (latest version is ~~presented~~ described by Rieger et al. (2018))~~as well as~~. The product used in this study, provides two parameters of the ~~aerosol PSD (~~unimodal lognormal aerosol PSD, namely, R_{mod} and σ ~~) were retrieved~~ (Malinina et al., 2018). The ~~PSD product was obtained by performing the retrieval with a fixed~~ aerosol number density (N) profile~~taken from ECSTRA climatology (Fussen and Bingen, 1999), and using the~~ , defining the third parameter of the PSD, was fixed throughout the retrieval. This profile was chosen in accordance with ECSTRA climatology for the background aerosol (Fussen and Bingen, 1999). In the retrieval process limb radiances normalized to the solar irradiance and averaged over ~~7~~ seven wavelength intervals (λ_1=750±2 nm, λ_2=807±2 nm, λ_3=870±2 nm, λ_4=1090±2 nm, λ_5=1235±20 nm, λ_6=1300±6 nm, λ_7=1530±30 nm) ~~. The retrieval was performed~~ were used directly without prior Ext retrieval. The the obtained PSD parameters profiles cover the altitude range from about 18 to 35 km. Spectral albedo was retrieved simultaneously with the PSD parameters, but only completely cloud free ~~profiles were used in the retrieval~~scenes were considered so far. More detailed information about the algorithm and the errors associated with a fixed N profile can be found in Malinina et al. (2018).

2.2 OSIRIS

OSIRIS is a limb-viewing instrument on board the Swedish satellite Odin, having a sun-synchronous orbit at around 600 km altitude. The mission started its operation in February 2001 and continues working at the time of writing. OSIRIS consists of two instruments, an optical spectrograph (OS) and infrared imager (IRI). OS makes measurements of scattered solar light in the spectral interval from 214 to 810 nm with 1 nm spectral resolution. Similarly to SCIAMACHY it observes the atmosphere tangentially to the Earth's surface in the altitude range from around 7 to 65 km with a vertical sampling of 2 km and vertical resolution of 1 km. IRI has a different measurement technique: it consists of three vertical photodiode arrays with 128 pixels each and filters 1260, 1270 and 1530 nm. Each pixel measures a line of sight at a particular altitude, and thus with each exposure the entire vertical profile covering around 100 km is created. Further information on the technical specifications of OSIRIS is presented in Llewellyn et al. (2004).

Based on the measured informationmeasurements by the OS, Ext profiles at 750 nm as well as were retrieved in the products v5.7 and 7.0, (Bourassa et al., 2012; Rieger et al., 2019). Additionally, the product v6.0 contains retrieved Ångström exponent ($\alpha_{750/1530}$)were retrieved. To obtain $\alpha_{750/1530}$ the information at one wavelength of the optical spectrograph (750 nm) as well as measurements at 1530 nm from the infrared imager were used. As a reference spectrum the measurement at the higher tangent altitude was applied. In the retrieval, r_{med} and N were fitted assuming fixed σ=1.6, and the obtained values were used to calculate the Ångström exponent and Ext at 750 nm. Due to a lack of the absolute calibration for the infrared imager, only albedo at 750 nm was retrieved. A detailed description of the algorithm and the products is given by Rieger et al. (2014).

2.3 SAGE II

SAGE II was a solar occultation instrument on the Earth Radiation Budget Satellite (ERBS). It operated from October 1984 to August 2005 in an orbit with 57° inclination at about 600 km altitude (Barkstrom and Smith, 1986). SAGE II was a Sun photometer with seven silicon photodiods with filters at 386, 448, 525, 600, 935 and 1020 nm wavelengths. During each sunrise and sunset encountered by the satellite the instrument measured solar radiance attenuated by the Earth's atmosphere. The measurements were provided from the cloud top to about 60 km with the vertical resolution of about 0.5 km. However, the spacial coverage of the measurements is quite sparse, as there is one sunrise and one sunset event per orbit. This results in 30 profiles per day, unlike SCIAMACHY and OSIRIS, which provide about 1400 profiles per day each. More technical information on SAGE II can be found in McCormick (1987).

For this study we used v7.0 of the SAGE II product, which is described in detail by Damadeo et al. (2013). In this version Ext profiles at 1020, 525, 452, and 386 nm are provided. For their retrieval, first, the slant-path transmission profiles were calculated at each wavelength, then using the spectroscopy data slant-path optical depth profiles were obtained for each of the retrieved species. With an "onion-peeling" technique the optical depth profiles were inverted to obtain Ext profiles. Later, based on the Ext at 525 nm and 1020 nm, r_{eff} as well as surface area density were obtained (Thomason et al., 2008) (Damadeo et al., 2013; Thomason et al., 2008).

3 Sensitivity of measurements to aerosol parameters

3.1 Aerosol parametrisation

Stratospheric aerosols are commonly represented by spherical droplets containing 75% H_2SO_4 and 25% of H_2O with particle sizes distributed log-normally (Thomason and Peter, 2006). Though in some studies using lognormally (e.g. Thomason and Peter, 2006)

. It should be noted, that different shapes of the aerosol size distribution were also considered. For example, Chen et al. (2018) used gamma-distribution for the updated OMPS Ext product. However, yet there is no evidence that any of those shape assumptions is a better or worse physical disription of aerosol. Historically, starting with Junge and Manson (1961) a lognormal distribution with one or two modes was used, e.g. for the in-situ instruments bimodal lognormal PSD is employed (e.g. Deshler et al., 2003; Deshler, 2008); however, for the space borne remote sensing instruments a unimodal lognormal distribution is most commonly considered (Damadeo et al., 2013; Rieger et al., 2014; von Savigny et al., 2015; Malinina et al., 2018):

$$\frac{dn}{dr} = \frac{N}{\sqrt{2\pi}\ln(\sigma)r}\exp\left(-\frac{(\ln(r_{med}) - \ln(r))^2}{2\ln^2(\sigma)}\right), \tag{1}$$

where N is the aerosol particle number density, r_{med} is the median radius and $\ln(\sigma)$ is the standard deviation of the $\frac{dn}{d\ln(r)}$ function. In some studies mode radius is used instead of the median radius (r_{med}) for the aerosol parametrisation. The former is defined as $R_{mod} = r_{med}/\exp(\ln^2(\sigma))$. In addition, Malinina et al. (2018) used the standard deviation of dn/dr function, which is referred to as the absolute distribution width:

$$w = \sqrt{r_{med}^2\exp(\ln^2(\sigma))(\exp(\ln^2(\sigma)) - 1)}, \tag{2}$$

because this parameter is easier for visual interpretation than σ, which is most commonly used in the aerosol parametrizations. In this study, similarly to Malinina et al. (2018), σ will be used when describing the retrieval settings, while w will be used in the results discussion.

As mentioned in Sect. 1, PSD parameters uniquely describe a ~~log-normal~~ lognormal distribution of the aerosol particle sizes, although often due to a lack of the information Ext at single wavelength is retrieved. ~~Aerosol~~ For the assumption of unimodal lognormal distribution, aerosol extinction coefficient at the wavelength λ is defined as

$$Ext_\lambda = \beta_{aer}(r_{med}, \sigma, \lambda...)N, \tag{3}$$

where β_{aer} is ~~the~~ calculated in accordance with the Mie scattering theory aerosol extinction cross section, which is dependent on the aerosol PSD (e.g. Liou, 2002). Some limited information about PSD is given by the Ångström coefficient or Ångström exponent, α, which was used in the empirical relation introduced by Ångström (1929):

$$\frac{Ext_{\lambda_1}}{Ext_{\lambda_2}} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha}. \tag{4}$$

However, the usage of α is associated with certain issues. In his work Ångström (1929) noted that the diameter of the particles calculated from α shows only an approximate coincidence with the average aerosol diameter directly measured. Furthermore he states, that the changes in the size of the particles do not necessarily lead to the changes in α. Another complication is related to the fact, that α value is spectral dependent and thus changes based on the wavelength pair used for its calculation (e.g. Rieger et al., 2014). ~~It is important to recall, that for limb instruments the retrieved values of Ext depend on the assumed PSD parameters. More detailed information about the influence of the errors in PSD parameters on the retrieved Ext can be found in Rieger et al. (2018); Loughman et al. (2018).~~

3.2 Measurement sensitivity

As mentioned before, occultation and limb scatter instruments employ different measurement approaches, resulting in different sensitivity to the aerosol parameters. When discussing occultation measurements in this study, we assume the measurements by solar occultation instruments. Such instruments register the solar radiation transmitted through the atmosphere during sunrise and sunset events as seen from the satellite. In contrast, limb scatter instruments measure profiles of the solar radiation scattered by the atmosphere. As it is presented in e.g. Rozanov et al. (2001), the radiative transfer equation is solved for occultation and limb observations in very different ways. The direct radiance as registered by an occultation instrument is described by:

$$I_{dir}(\Omega) = I_0 \exp\left(- \int\limits_0^s k(\hat{s}) d\hat{s} \right), \tag{5}$$

where I_0 is the incident solar flux, Ω is an angle, defining the radiance propagation direction, s is the full path length through the atmosphere along the solar beam, and k is the extinction coefficient.

The diffuse radiance, which is observed by a limb scatter instrument is given by:

$$I_{sc}(\Omega) = \int\limits_0^{s_{LOS}} \frac{1}{4\pi} \Bigg(\int\limits_{\Omega} \big(\eta_R p_R(\Omega,\Omega') + \eta_a p_a(\Omega,\Omega')\big) I_{sc}(\Omega') d\Omega' + \\ + \big(\eta_R p_R(\Omega,\Omega_0) + \eta_a p_a(\Omega,\Omega_0)\big) I_0 \exp\Big(- \int\limits_0^{s_{Sun}} k(\hat{s}) d\hat{s} \Big) \Bigg) e^{-\tau(s)} ds. \tag{6}$$

Here, s_{LOS} stands for the full path along the line of sight of the instrument, η_R and p_R for the Rayleigh scattering coefficient and the phase function, η_a and p_a for the scattering coefficient and the phase function of the aerosol, Ω_0 for the solar beam propagation direction, s_{Sun} for the full path along the solar beam and τ for the optical depth along the light-of-sight.

~~Comparing Eqs~~Analysing Eq. (5) ~~and (6)~~ the one can see, that it is quite straightforward to derive k from I_{dir}. In the wavelength intervals without any other absorber features k represents a sum of the aerosol extinction coefficient and Rayleigh scattering coefficient, i.e. $Ext{+}\eta_R$. In contrast, to obtain ~~I_{dif}~~ I_{sc} using Eq. 6 an iterative approach is needed. Furthermore, ~~I_{dif}~~ I_{sc} depends on the product of p_a and η_a. In turn, both, p_a and η_a are determined by the aerosol PSD parameters. Thus, in most of the Ext retrieval algorithms which rely on limb measurements an assumption on the PSD parameters is used, and those are kept fixed during the retrieval process. In addition, p_a is a function of the scattering angle. The issue related to the dependency of p_a and the limb radiances on the solar scattering angle (SSA) is well known, and was ~~discussed~~ carefully investigated by Ernst (2013); Rieger et al. (2014, 2018); Loughman et al. (2018). However, this limitation is not relevant to the current study, as the for used limb datasets PSD is retrieved directly from the measured radiances rather than from pre-retrieved Ext.

To understand how the differences in the measurement techniques influence the instrument sensitivity to aerosols, extended analysis is provided below. In some previous studies (e.g Twomey, 1977; Thomason and Poole, 1993; Rieger et al., 2014) the analysis of so-called kernels was used to show the contribution of the particles of different sizes to the observed radiance.

According to Twomey (1977) the measured intensity of the scattered light can be presented as:

$$I(\lambda) = \int_0^\infty K_{sc}(\lambda, r) n(r) dr, \quad (7)$$

where r is a radius of the particle and ~~K~~ K_{sc} is a kernel. For the measurements of the transmitted light the following equation is appropriate (Twomey, 1977):

$$\ln(I(\lambda)/I_0(\lambda)) = \int_0^\infty K_{dir}(\lambda, r) n(r) dr. \quad (8)$$

Whereas for the measurements of scattered solar light ~~K~~ K_{sc} does not have an analytic representation, for the occultation measurements ~~it~~ kernel K_{dir} is given by $r^2 Q_e(r/\lambda)$, where Q_e is the Mie extinction efficiency. Besides, the right sides of the Eqs. (7) and (8) have the same form, although they refer to different left sides. Indeed, for the scattered light measurements the left side is represented by $I(\lambda)$, while for the transmission the left side is $\ln(I(\lambda)/I_0(\lambda))$, which according to the Eq. (5) is $\tau = -\int_0^s k(\hat{s}) d\hat{s}$. Thomason and Poole (1993) derived ~~K~~ K_{dir} for the extinction measurements ~~and showed it for~~ from SAGE II. In their research ~~K~~ K_{dir} had units of m^{-1}, since they were assessing it per unit volume of air. For the limb measurements, ~~K~~ K_{sc} was derived for the single-scatter radiance by Rieger et al. (2014)~~, and the resulting K~~. In their work, Rieger et al. (2014) did not use Eq. (7) directly, but assessed the kernels for the measurement vector $y = ln(\frac{I_{aer}+I_R}{I_R})$, where I_{aer} and I_R are aerosol and Rayleigh radiance contributions. Thus, the resulting K_{sc} was dimensionless. It should be also noted, that for their study Rieger et al. (2014) preferred to calculate ~~K~~ K_{sc} for OSIRIS numerically, stating, that the derived formula contains too many approximations. ~~Thus, K~~ Thereby, kernels can be used in the assessment of the sensitivity of the instruments employing the same measurement technique, but is not suitable for the interinstrumental comparisons. As one objective of our study is the comparison of the sensitivities of the limb and occultation measurements to the particles of the different sizes, we will not follow the approach of Twomey (1977). Instead, we define the dimensionless measurement sensitivity, S, to the aerosol particles of a certain size as a change in the intensity of the observed radiation with respect to the aerosol free conditions. Thus, S is defined by:

$$S(\lambda, r_i) = \frac{I(\lambda) - I_R(\lambda)}{I_R(\lambda)}, \quad (9)$$

where $I(\lambda)$ is the radiance including both, Rayleigh and aerosol signals, and I_R is the Rayleigh signal. When S=0, the radiance has no contribution from the aerosol extinction or scattering. With increasing S increases the aerosol contribution to the measured radiance.

The quantitative assessment of S is made by modelling the intensities with the radiative transfer model SCIATRAN Rozanov et al. (2014) for limb measurements (on the example of SCIAMACHY limb geometries) and for occultation measurements. The intensities were modelled for the distributions with R_{mod} varying from 0.04 to 0.30 μm with the step (Δr) of 0.01 μm. ~~For~~ In accordance with the chosen Δr, for each distribution, σ has been chosen such that w is equal to 0.01 μm (~~this corresponds to the chosen Δr).~~ e.g. R_{mod}=0.10 μm, σ=1.10; R_{mod}=0.15 μm, σ=1.07; R_{mod}=0.20 μm σ=1.05). The same background N ~~was considered to be the same~~ profile was used for all simulations.

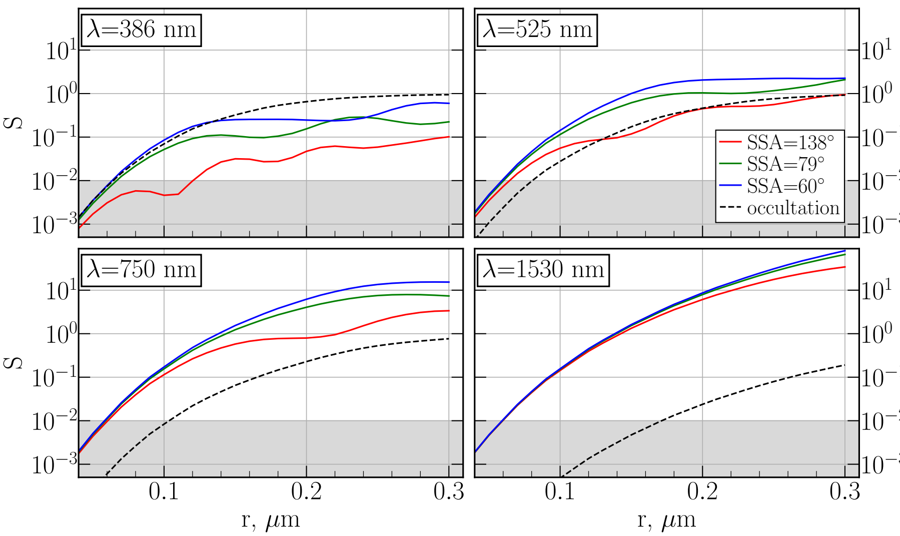

Figure 1. Modelled sensitivity, S, at 21.7 km for range of particles radii (r). The simulations were performed for different limb geometries with different solar scattering angles, SSA, and for one occultation geometry.

For the above described simulations S at λ=386 nm, λ=525 nm, λ=750 nm and λ=1530 nm are presented in Fig.1 for occultation (dashed black line) and for limb measurements (colored solid lines). As limb radiances depend on SSA, the simulations were done for three different observational geometries: SSA=60° (blue line), SSA=79° (green line) and for SSA=138° (red line). The angles from 60 to 140° represent the SSA range for SCIAMACHY measurements in the tropical region. The calculations were done using an ozone climatology for the month and location of the SCIAMACHY measurements. The grey shaded area shows S<0.01. We believe, that this empirical value stays for a typical uncertainty of the measurement-retrieval system, caused by the uncertainties in the radiative transfer modelling. A justification for this is provided further in this section.

As it is seen from Fig. 1, S for limb radiances at ~~both wavelengths~~ λ=750 nm and λ=1530 nm are obviously higher than those for the occultation measurements. For λ=525 nm, the curves representing SSA=60° and SSA=79° lay also above the occultation one, while the red curve (SSA=138°) crosses the occultation line in multiple points being generally comparable by the magnitude. Moreover, for these wavelengths all the SSAs the limb curves enter the shaded grey area at around 0.06 μm, while the occultation curves enter this area at about 0.08 μm at λ=525 nm, at 0.11 μm at λ=750 nm and at ~~0.20~~0.16 μm at λ=1530 nm. These results agree with those presented by Thomason and Poole (1993) for SAGE II measurements, and illustrate the well-known statement that the occultation measurements in visible-near-infrared spectral range are insensitive to the particles with the radii smaller than 0.1 μm (see e.g. Thomason et al., 2008; Kremser et al., 2016). ~~Better~~ Considering this spectral interval, better sensitivity to the smaller particles is observed for the limb measurements, which is explained by the fact that the aerosol PSD contributes in very different ways into the observed radiances for limb and occultation measurements. While I_{dir} depends inverse exponentially on k (or Ext), ~~I_{dif}~~ I_{sc} is, to a first order, proportional to the product of p and η. Both,

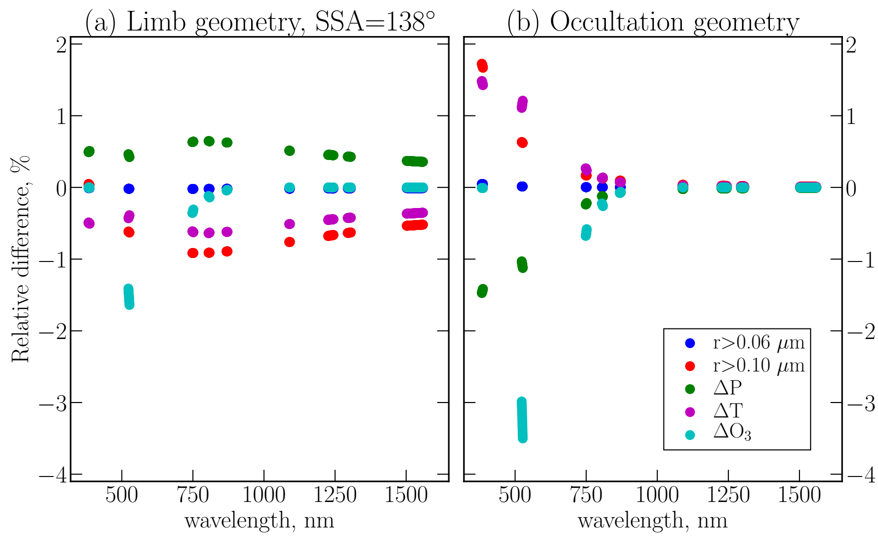

Figure 2. Relative changes in the radiance at 21.7 km for limb (panel (a)) and occultation (panel (b)) geometry. The responses in radiance due to 10% changes in temperature, atmospheric pressure and 5% changes in ozone concentration are presented by magenta, green and cyan dots, respectively. The cut off in PSD at 0.06 μm is depicted with blue dots, and the cut off at 0.10 μm is presented with red dots.

p and η, depend on the PSD parameters~~, and as~~. As a result, in the considered spectral region limb radiances tend to be more sensitive to aerosol particles of the smaller size, and thus provide more accurate PSD parameters during the the "background" aerosol loading conditions.

It should be noted that at shorter wavelengths the situation is different. Thus, for presented in Fig. 1 λ=386 nm the limb curves lay for all geometries below the occultation curve. Additionally, SSA=60°, SSA=79° and occultation lines cross the low-sensitivity area at around r=0.06μm, while the red line crosses the grey area at r=0.12 μm. Such a change of the sensitivity for the short wavelengths is determined by the fact, that for limb measurements the Rayleigh signal in this spectral range is dominating (e.g. Bourassa et al., 2012). At the same time, for the occultation measurements inclusion of this wavelength in the PSD retrieval might be useful for the volcanically quiescent periods.

To justify the choice of the sensitivity threshold, we provide an example of the relative differences in the ~~the~~ modelled radiance due to changes in different factors for both, limb (SSA=138°) and occultation, geometries assuming randomly picked PSD with R_{mod}=0.08 μm and σ=1.6. These relative differences at 21.7 km for different wavelengths are presented in Fig. 2, with the results for limb geometry depicted in panel (a) and for occultation geometry in panel (b). For ~~each simulation the~~ the simulations the whole profile of ~~one of the following parameters was increased by 10%:~~ either temperature (magenta dots) ~~,~~ or atmospheric pressure (green dots) ~~and~~ was increased by 10%. This perturbation is related to the estimated uncertainties of these parameters in the stratosphere. In the additional simulation ozone concentration (cyan dots) ~~. For simplicity reasons, we choose to perturb the whole profile, rather than values at the particular altitudes.~~ was enhanced by 5%. Changes in ozone concentration by ~~10~~5% are reasonable as they reflect the remaining uncertainties in the ozone profiles retrieved from the space-borne measurements across the relevant altitude range (Tegtmeier et al., 2013). For simplicity reasons, we perturb the whole profile, rather than values at the particular altitudes. Additionally, we depicted with the blue dots the changes in the radiances by assuming the concentration of the aerosol particles with $r <=$0.06 μm being equal to zero (cut off at 0.06 μm) and with the red dots assuming the cut off at 0.10 μm (particles with $r <=$0.10 μm were not considered). Figure 2 shows, that for the limb geometry the relative changes in the radiance due to changes in temperature, pressure or ozone concentration are within 0.8% ~~in the considered wavelength interval~~for most wavelengths. The exception is 525 nm wavelength, where changes in the ozone concentrations are about 1.5%. For the PSD cutoff at 0.06 μm the changes are about 0.03%, however, for the cut off at 0.10 μm the changes are ~~slightly larger than~~ about 1% ~~for the wavelength interval shorter than~~ between 700 nm and 1000 nm, and and reducing to 0.6% at the longer and shorter wavelengths. For the occultation geometry the changes are somewhat different. Namely, the changes in pressure or temperature are comparable with the changes in the PSD cut off for all wavelengths, but the changes in ozone concentration contribute up to ~~1.4~~3.5% in the radiance at 525 nm and 1.4% at 750 nm. Such behaviour of relative changes in the intensities shows that in the considered wavelength interval 1% provides a realistic estimation of the uncertainties from the radiative transfer modelling for both geometries, even though this uncertainty is caused by different factors. ~~For limb geometry, the changes due to PSD cut off at r=0.06 μm are within this threshold, however, with the cut off at r=0.10 μm the relative difference in the radiances exceeds it, and thus this change is considered as detectable. For the occultation measurements, both PSD cut offs result in smaller changes than those coming from the radiative transfer modelling uncertainty.~~

Summarizing Sect. 3.2, it can be concluded, that due to differences in the underlying radiative transfer processes, limb and occultation instruments have intrinsically different sensitivity to stratospheric aerosol parameters. Limb radiances are more sensitive to the smaller aerosol particles ~~, which~~ when measurements in the visible and near-infrared wavelength interval are used. This is expected to result in a more accurate PSD parameters retrieval if this spectral range is used. This is particularly the case during the background aerosol loading periods when smaller particles prevail. ~~However,~~ Ext retrieval from limb instruments suffers from the uncertainties due to assumed PSD and the SSA dependency. On the contrary, the retrieval of Ext from the occultation radiances is more straightforward, but the threshold of the sensitivity of the occultation measurements to the aerosol particle sizes is somewhat lower in comparison to the limb instruments. More information on the smaller particles from the occultation measurements can be obtained if the ultraviolet part of the spectrum is considered.

4 Error assessment

4.1 Extinction coefficients errors

As discussed above, aerosol extinction coefficient data bases are widely used in the stratospheric aerosol research. In order to make SCIAMACHY aerosol PSD product comparable with the products from other satellite instruments, Ext at four wavelengths were calculated and then used to derive the Ångström exponents at two wavelength pairs.

As it was mentioned in Sect. 2.1, SCIAMACHY PSD product provides R_{mod} and σ. These parameters were retrieved assuming a fixed N profile. Thus, during the volcanically active periods, an inadequate assumption of N results in errors in the retrieved R_{mod} and σ. To assess how this assumption affects resulting Ext and Ångström exponent, synthetic retrievals were performed. The limb radiances were simulated with the known "true" parameter settings and then used in the retrieval instead of the measurement spectra. Applying this approach, Malinina et al. (2018) showed, that R_{mod} was retrieved with an accuracy of about 20% even if the true value of N was a factor of 2 higher than the value assumed in the retrieval. For scenarios with the perturbed N profile, σ is retrieved with about 10% accuracy, while an accuracy of about 5% is reached for the profiles with unperturbed N.

To extend the error assessment of SCIAMACHY PSD product, we use the same scenarios and the same modelled radiances as ~~(Malinina et al., 2018) to evaluate~~ Malinina et al. (2018) to evaluate noise and parameter errors in Ext resulting from the errors in the retrieved R_{mod} and σ. For this study Ext was calculated employing Mie theory and Eq. (3) at 525, 750, 1020 and 1530 nm using the retrieved PSD information and the N profile assumed in the retrieval (exponentially decreasing from 15.2 cm^{-3} at 18 km to 0.5 cm^{-3} at 35 km). In addition, Ext_{750} was retrieved from the simulated radiances using an algorithm similar to SCIAMACHY v1.4 (Rieger et al., 2018), but with the normalization to the solar spectrum and using the phase function which was calculated from the retrieved R_{mod} and σ at each altitude. For this retrieval the albedo value was set to the value resulting from the PSD parameters retrieval. To distinguish the two approaches to calculate extinctions we denominate Ext, obtained with Eq. (3) as "calculated" and Ext, retrieved using the corrected PSD as "retrieved".

Five scenarios for a typical observational geometry in the tropical region were used. Four of them: "small", "background", "unperturbed" and "volcanic" were simulated with the same N profile as the one used for the retrieval and the aerosol PSD parameters listed in Tab. 1. For the scenario "volcanic (2N)" N profile was multiplied by the factor of 2 below 23 km (as discussed in Malinina et al. (2018) this approach is considered to be realistic for the SCIAMACHY operation period). The scenarios are summarized in Tab. 1. For all scenarios the surface albedo was set to 0.15 at all wavelengths (perturbed by 0.35 in comparison to the first guess value 0.5). The measurement noise was simulated by adding the Gaussian noise to the simulated radiances. The signal-to-noise ratios were assessed from the SCIAMACHY measurements. The retrievals were done using 100 independent noise sequences to ensure reliable statistics.

Panel (a) of Fig. 3 shows the median calculated Ext_{750} profiles with solid lines and median retrieved Ext_{750} profiles with dashed lines. The "true" values are shown by the dotted lines. The colors corresponding to each scenario are listed in Tab. 1. Panel (b) shows the median relative parameter errors for both calculated and retrieved Ext_{750}. Solid shaded areas show ± 1 standard deviation for the calculated profiles, while the striped ones denote ± 1 standard deviation for the retrieved profiles. The maximum relative errors for the calculated aerosol extinction coefficients at other wavelengths is presented in Tab 1. As the altitudinal behaviour of the extinction coefficients at the other wavelengths is the same as for the Ext_{750} profiles, we show the results only for one wavelength.

As it follows from Tab. 1, the relative parameter errors for the scenarios with unperturbed N do not exceed 20%. As expected, for the "volcanic (2N)" scenario, the errors are slightly higher and vary depending on the wavelength from 19% to 31%. For all scenarios the largest parameter errors are observed for Ext_{525}. This is most likely because for the retrieval of PSD parameters only the wavelengths longer than 750 nm were taken into consideration, while the information from the visible and UV parts of the spectrum has no contribution to the retrieval.

Table 1. Selected scenarios and associated maximum relative parameter errors in the calculated extinction coefficients.

Name	True		Color*	Perturbation	Max.			
	R_{mod}	σ			$\epsilon_{Ext_{525}}$	$\epsilon_{Ext_{750}}$	$\epsilon_{Ext_{1020}}$	$\epsilon_{Ext_{1530}}$
Small	0.06 μm	1.7	cyan	R_{mod}, σ	20%	17%	15%	12%
Background	0.08 μm	1.6	blue	R_{mod}, σ	10%	9%	8%	7%
Unperturbed	0.11 μm	1.37	brown	unpert.	4%	4%	4%	4%
Volcanic	0.20 μm	1.2	green	R_{mod}, σ	7%	9%	10%	11%
Volcanic (2N)	0.20 μm	1.2	red	R_{mod}, σ, N	31%	26%	22%	19%

* Color of the lines in Fig. 3.

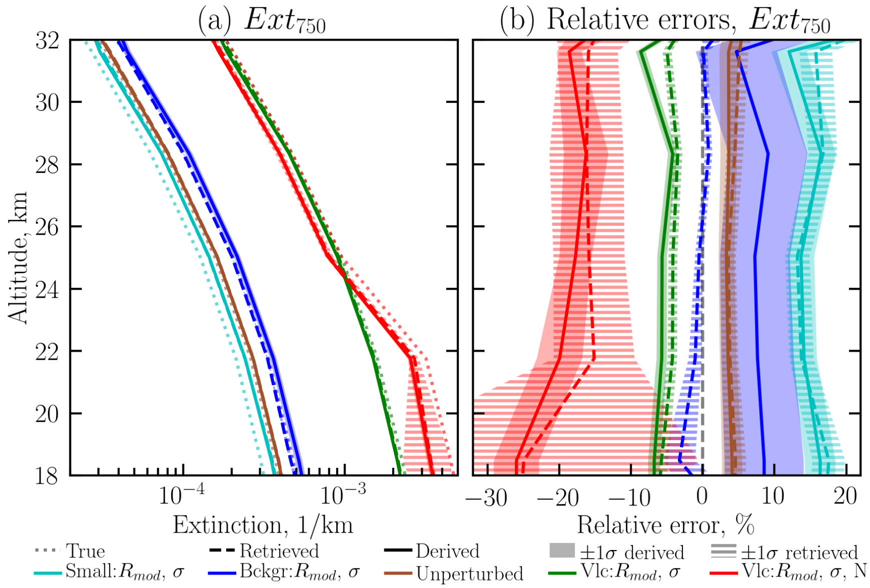

Figure 3. Profiles of Ext_{750} (a) and their relative parameter errors (b) for a typical tropical observation geometry. The solid lines show the Ext_{750} profiles calculated from PSD product~~profiles, dashed~~. Dashed lines depict the directly retrieved profiles, while the dotted lines represent the true values. The shaded areas stand for ± 1 standard deviation. The scenarios used for the simulations are listed in Table 1.

Analyzing Fig. 3 it is important to mention that retrieved Ext_{750} barely differs from that calculated. For "small", "unperturbed" and "volcanic" scenarios the solid and dashed lines are very close to each other, and the median relative errors of the retrieved profiles lay mostly inside the standard deviation of calculated profiles. For the "background" conditions the retrieved and the calculated profiles have the same shape, but the retrieved profiles are about 8% more accurate. As it was suggested that the difference between calculated and retrieved profiles of Ext might possibly be used to correct N for the PSD retrieval, it is most important to analyze the Ext profiles for the scenario with the perturbed N profile ("volcanic (2N)"). As it can be seen from the Fig. 3 (a) the retrieved Ext_{750} shows similar altitudunal behaviour as the calculated profile, although the retrieved profile has larger standard deviation at the lowermost altitude. Similarly to the calculated profile, the retrieved profile is about 25-30% lower than the "true" one. This leads to the conclusion, that an additional retrieval of Ext with the corrected PSD or fixing Ext during the retrieval does not provide any additional information about the aerosol PSD, and some other independent data or constraint is needed to retrieve all three parameters. One possible way would be to combine limb and occultation measurements, using the later to constraint N. Another possibility to constraint N would be through the use of collocated profiles of stratospheric H_2SO_4 concentrations. For SCIAMACHY the use of the MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) H_2SO_4 volume mixing ratio data set (Günther et al., 2018) might be appropriate. However, synergistic use of the data from two different instruments is not straightforward, and is a subject ~~of the~~ for further studies.

4.2 Ångström exponents errors

Combining Eqs. (3) and (4), the Ångström exponent can be obtained as

$$\alpha_{\lambda_1/\lambda_2} = -\frac{\ln(\beta_{aer}(\lambda_1, r_{med}, \sigma)/\beta_{aer}(\lambda_2, r_{med}, \sigma))}{\ln(\lambda_1/\lambda_2)}. \tag{10}$$

Equation (10) shows, that Ångström exponent is not directly dependent on N, thus only the errors from R_{mod} and σ influence the derived $\alpha_{\lambda_1/\lambda_2}$. To assess this influence, the Ångström exponents were calculated using Eq. (10) and the calculated Ext from the synthetic retrievals discussed in Sect. 4.1. While from SCIAMACHY PSD product the Ångström exponents can be calculated at any wavelength pair, SAGE II and OSIRIS provide only $\alpha_{525/1020}$ and $\alpha_{750/1530}$ respectively. Thus, we limit our analysis to those values. The "true" Ångström exponent values, the scenario summaries (described in the previous section) as well as the maximum absolute and relative parameter errors for $\alpha_{525/1020}$ and $\alpha_{750/1530}$ are presented in Tab. 2. As for Ext analysis, we show the altitudinal behavior only for one wavelength pair ($\alpha_{750/1530}$), while the results for the other pair are very similar. In panel (a) of Fig. 4 the median derived Ångström exponents are presented with solid lines, and the "true" values are shown by dashed lines; in the panel (b) median relative parameter errors for the chosen scenarios are depicted. For both panels shaded areas show ± 1 standard deviation.

Analysis of Tab. 2 and Fig. 4 leads to the conclusion that the relative parameter error in the Ångström exponent for all scenarios is below 10% for $\alpha_{525/1020}$, and less than 5% for $\alpha_{750/1530}$. As expected, the largest errors are seen for the "volcanic (2N)" scenario, where N profile was perturbed and the parameter errors in R_{mod}, σ and Ext were the largest. For all scenarios the largest errors are observed at the lowermost retrieved altitude, e.g. for $\alpha_{750/1530}$ the errors above 21.3 km do not exceed 2.5%. The absolute error for $\alpha_{525/1020}$ is less than 0.12 for the scenarios with unperturbed N and about 0.2 for the scenario with N perturbed by a factor of 2. For $\alpha_{750/1530}$ the absolute parameter errors are even smaller, in particular, for the scenarios with the same N as used for the retrieval the errors are smaller than 0.1, and for the "volcanic (2N)" scenario the difference between the true and derived Ångström exponent is 0.15.

Summarizing Sect. 4 it can be concluded, that parameter errors in the calculated Ext for the background scenarios do not exceed 20%, and are about 20-25% for the cases with the perturbed N. The largest errors are observed for the aerosol extinction coefficient at 525 nm, most likely due to the missing information from the visible spectral range in the PSD retrieval. The retrieval results for Ext with the retrieved PSD parameters barely differs from the aerosol extinction coefficients calculated

Table 2. Selected scenarios and associated maximum absolute (relative) parameter errors in Ångström exponents.

Name	True				Color*	Perturbation	Max.	
	R_{mod}	σ	$\alpha_{525/1020}$	$\alpha_{750/1530}$			$\epsilon_{\alpha_{525/1020}}$	$\epsilon_{\alpha_{750/1530}}$
Small	0.06 μm	1.7	2.18	2.76	cyan	R_{mod}, σ	0.12 (4.8%)	0.10 (4.3%)
Background	0.08 μm	1.6	2.22	2.84	blue	R_{mod}, σ	0.05 (2.4%)	0.06 (2.1%)
Unperturbed	0.11 μm	1.37	2.76	3.36	brown	unpert.	0.01 (0.3%)	0.01 (0.2%)
Volcanic	0.20 μm	1.2	2.41	3.12	green	R_{mod}, σ	0.04 (1.6%)	0.03 (1.0%)
Volcanic (2N)	0.20 μm	1.2	2.41	3.12	red	R_{mod}, σ, N	0.20 (8.3%)	0.15 (4.7%)

* Color of the lines in Fig. 4.

In the last 2 columns maximum absolute error for the profile is given by the number without brackets, while the maximum relative error is presented in brackets.

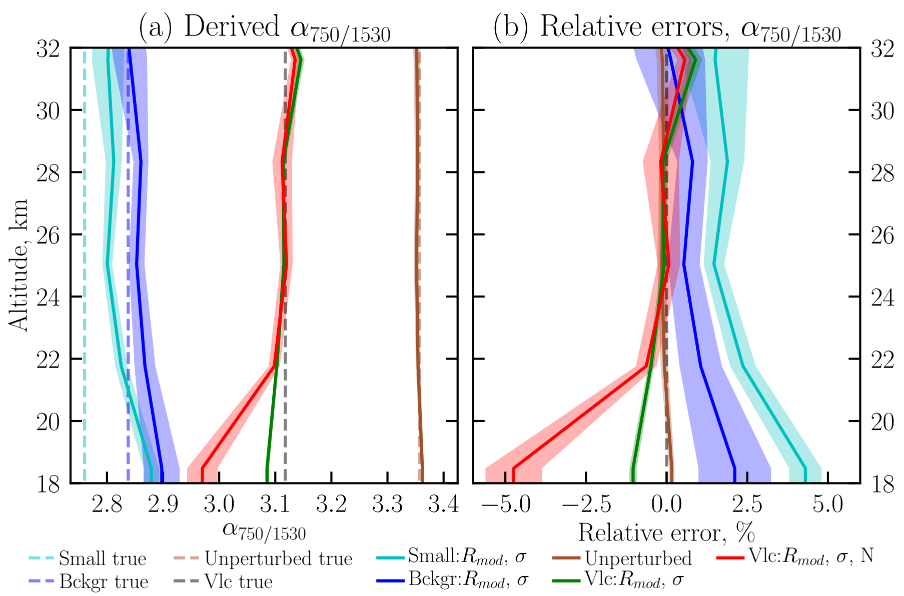

Figure 4. Ångström exponent profiles ($\alpha_{750/1530}$) (a) and their relative parameter errors (b) for a typical tropical observation geometry. The solid lines show the derived from PSD product profiles, while the dashed lines represent the true values. The shaded areas stand for ± 1 standard deviation. The scenarios used for the simulations are listed in Table 2.

from PSD product, and thus, cannot be used to improve the knowledge of N. For the Ångström exponent the parameter errors are much smaller, as they cancel out in the ratio of the aerosol extinction coefficients and depend only on the aerosol PSD. For $\alpha_{525/1020}$ the relative parameter error does not exceed 10%, and for $\alpha_{750/1530}$ the relative error is below 5%. For both Ångström exponents the absolute parameter errors are less than 0.2 for the cases with the perturbed N profile and are even less than 0.1 for unperturbed N.

5 Comparison of the measurement results

5.1 Extinction coefficients comparison with SAGE II

As discussed in the previous sections, SAGE II was an outstanding instrument, providing aerosol extinction coefficients, and there have been several comparisons performed using its data. For the SCIAMACHY PSD product the comparisons with SAGE II are associated with some challenges. First, SCIAMACHY and SAGE II have only a 3 year period of overlap. Second, SAGE II was an occultation instrument, providing about 30 vertical profiles per day, which resulted in 57 collocated profiles with SCIAMACHY (collocation criteria are ±5° latitude, ±20° longitude and ±24 hours) for the entire overlap period because the SCIAMACHY PSD retrieval is currently limited to completely cloud free profiles in the tropical zone (20°S-20°N). This amount of collocated profiles is not enough for an indepth investigation, however a rough assessment of the consistency of the data from both instruments can be provided. Another issue related to the SCIAMACHY and SAGE II comparison is the difference in the measurement techniques and thus a different sensitivity to aerosol properties. As was discussed in Sect. 3.2, SAGE II as an occultation instrument using visible-near-infrared spectral information for the PSD assessment is less sensitive to the smaller particles~~, although~~ . At the same time, Ext retrieval from SAGE II is associated with smaller uncertainties. The limb measurements from SCIAMACHY, in turn, are more sensitive to the particles with r <0.10 μm, although the direct retrieval of Ext is associated with some issues (see Sect. 3.2). The comparison of the r_{eff} from SAGE II and SCIAMACHY, presented in Malinina et al. (2018) is expected to be influenced by these differences, because the instruments overlap period is considered to be volcanically quiescent with a prevailing amount of smaller particles. Here, we present the comparison of Ext retrieved from SAGE II and that calculated from SCIAMACHY PSD product, which is expected to be more reliable than direct comparison of r_{eff}.

To perform the comparison, SCIAMACHY aerosol extinction coefficients at 525, 750 and 1020 nm were calculated with Eq. (3), considering the same N profile as used in the PSD retrieval. As SAGE II did not have a 750 nm channel, Ext_{750} for this instrument was calculated with Eq. (4) from Ext_{525} and Ext_{1020} using $\alpha_{525/1020}$. To assess a possible uncertainty associated with the usage of the Ångström exponent when calculating Ext_{750} from SAGE II data, SCIAMACHY Ext_{750} was additionally calculated using the same approach. To distinguish between two different methods of Ext_{750} calculation, we use $Ext_{750}(PSD)$ for the one derived from PSD product with Eq. (3), and $Ext_{750}(\alpha_{525/1020})$ for that calculated using the Ångström exponent. As SCIAMACHY and SAGE II have different vertical resolution, SAGE II data was smoothed to the coarser SCIAMACHY vertical resolution and then interpolated onto the SCIAMACHY vertical grid. The mean relative differences between SCIAMACHY and SAGE II for Ext_{525} (blue line), Ext_{1020} (green line), $Ext_{750}(PSD)$ (red line) and $Ext_{750}(\alpha_{525/1020})$ (magenta line) are presented in Fig. 5. The shaded areas show the standard error of the mean. We prefer to depict the standard error of the mean instead of the standard deviation to make the figures less busy.

As it can be seen from Fig. 5, for all wavelengths the ~~errors~~ differences for the derived extinction coefficients are below ±25%, which is within the reported precision of the extinction coefficients for SCIAMACHY. For Ext_{1020} the shape of the relative difference follows the one reported earlier in Malinina et al. (2018) for the differences between SCIAMACHY and SAGE II effective radii, but with slightly different values (-20 to 10% for Ext_{1020} versus -30 to 0% for r_{eff}). Such behaviour

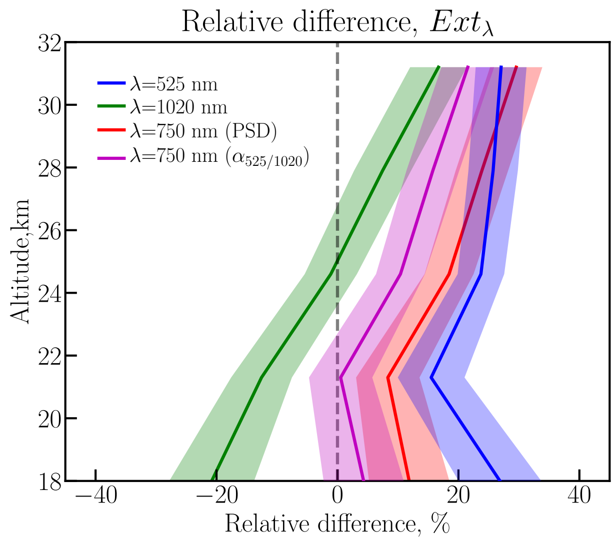

Figure 5. Mean relative difference (200 ×(SCIAMACHY-SAGE II)/(SCIAMACHY+SAGE II)) between extinction coefficients at 525 nm (blue line), 1020 nm (green line), 750 nm obtained directly from PSD (red line) and 750 nm converted with $\alpha_{525/1020}$ (magenta line) from collocated SCIAMACHY and SAGE II measurements. Shaded areas show standard error of the mean.

is expected, because r_{eff} from SAGE II is obtained using the Ext_{1020} and Ext_{525} (see Sect. 2.3). The differences in the Ext_{525}, $Ext_{750}(PSD)$ and $Ext_{750}(\alpha_{525/1020})$ are fairly constant with height and vary from 15 to 25% for Ext_{525}, from 10 to 25% for $Ext_{750}(PSD)$ and from 0 to 15% for $Ext_{750}(\alpha_{525/1020})$. The discrepancy between two different ways of computing Ext_{750} derivation is quite remarkable, as with the consistent methods a better agreement is obtained. Though it should be highlighted once again, that for SAGE II 750 nm is not a measurement wavelength, and 525 nm is not considered in the SCIAMACHY retrieval. To highlight the uncertainties coming from the different approaches to derive the Ext_{750}, we depict in Fig. 6 the mean relative differences between $Ext_{750}(\alpha_{525/1020})$ and $Ext_{750}(PSD)$ for the whole SCIAMACHY data set with a solid line, and ±1 the standard deviation shown as the shaded area. From Fig. 6 it is clear that the calculation of the extinction coefficient with Ångström exponent results in about 8% negative bias. This result is consistent with the result presented in the supplements to Rieger et al. (2015). Thus, it is important to consider this uncertainty when comparing the aerosol extinction coefficients from different instruments measuring at different wavelengths.

5.2 Ångström exponents comparison with SAGE II

To extend the comparison to the data from SAGE II, $\alpha_{525/1020}$ were calculated from its aerosol extinctions, and compared to the ones derived from SCIAMACHY PSD product. The mean differences between SCIAMACHY and SAGE II Ångström exponents are presented in Fig. 7 with the solid blue line, with relative differences in panel (a) and absolute differences in panel (b). Following the previous comparison, the standard error of the mean is shown with the shaded area.

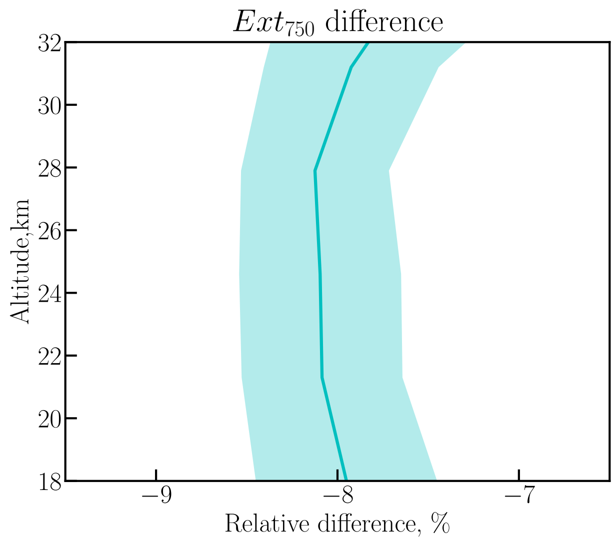

Figure 6. Mean relative difference between $Ext_{750}(PSD)$ obtained from PSD product with Eq. (3) and $Ext_{750}(\alpha_{525/1020})$ calculated using the Ångström exponent ($100\times(Ext_{750}(\alpha_{525/1020})$-$Ext_{750}(PSD))/Ext_{750}(PSD)$) from all available SCIAMACHY measurements. Shaded area shows ±1 standard deviation.

The relative difference between the instruments is altitude dependent, showing ~~slightly~~ different shape than Ext_{1020}. SCIAMACHY $\alpha_{525/1020}$ is about 40% higher than that from SAGE II at 18 km, about 20% higher between 21 and 28 km, and the difference at 31 km is around 10%. In the absolute values the difference varies from 0.8 at the lowermost altitude to 0.2 at the uppermost one. The ~~bias~~ observed differences in this comparison ~~is~~ are expected. As it was discussed in the previous section, the differences between SCIAMACHY and SAGE II for Ext_{1020} and Ext_{525} are both about 20%, but have the opposite sign, which results in the amplification of the error when calculating $\alpha_{525/1020}$. As for the r_{eff} and Ext_{1020}, the reason why the difference between SCIAMACHY and SAGE II is altitude dependent is still under investigation. To address this question the amount of collocations needs to be significantly increased to provide better sampling. This can be done by extending the SCIAMACHY PSD retrieval algorithm to other latitude bands and applying it to the profiles with cloud contamination.

5.3 Ångström exponents comparison with OSIRIS

Although comparison of SCIAMACHY with SAGE II provides some information on the agreement of the products, this comparison is not sufficient to draw any robust conclusions. Additional information can be gained from the comparison with OSIRIS, which was operating at the same time as SCIAMACHY and also provided particle size information. Generally, comparison between SCIAMACHY and OSIRIS is more robust than with SAGE II, as both instruments employ the same measurement technique, use information at the same wavelengths (750 nm and 1530 nm), and provide a similar amount of measurements per orbit. Applying the same collocation criteria as for SAGE II, 4603 coincident profiles were found, which

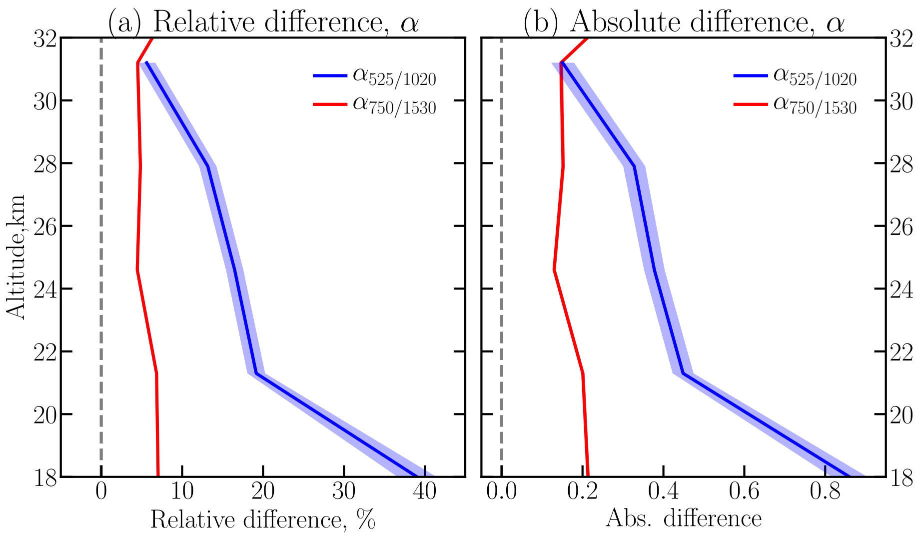

Figure 7. Mean relative (panel (a)) and absolute (panel (b)) differences (200×(SCIAMACHY-instrument)/(SCIAMACHY+instrument)) between Ångström exponents from collocated SCIAMACHY and SAGE II ($\alpha_{525/1020}$: blue lines) and SCIAMACHY and OSIRIS ($\alpha_{750/1530}$: red lines) measurements. Shaded areas show standard error of the mean.

is about half of the available SCIAMACHY profiles. The obtained amount of collocations is sufficient to ensure a reliable comparison. It is important to highlight once again that all the comparisons were done for the tropical region (20°N-20°S).

Differences between SCIAMACHY and OSIRIS $\alpha_{750/1530}$ are presented in Fig. 7 by the red line. OSIRIS $\alpha_{750/1530}$ was interpolated onto the SCIAMACHY measurement grid. The difference in vertical resolution was not accounted for as SCIAMACHY and OSIRIS have similar specifications. As for SAGE II, in panel (a) of Fig. 7 the mean relative differences are plotted with a solid line, while in panel (b) the mean absolute differences are depicted. The standard error of the mean is shown by the shaded area; it is, however, within the thickness of the solid line. As follows from Fig. 7, the relative difference between SCIAMACHY and OSIRIS is about 7% for the lower altitudes and about 4% for the altitudes above 25 km. In absolute values the difference is about 0.2 below 25 km, and less than 0.15 at the higher altitudes. Taking into consideration the $\alpha_{750/1530}$ errors from SCIAMACHY (5%), estimated in Sect. 4.2, and the errors reported by Rieger et al. (2014) for OSIRIS Ångström exponents (10%), it can be concluded, that the Ångström exponents obtained from both instruments agree well with difference being smaller than the reported errors.

To evaluate the temporal behaviour of the differences between SCIAMACHY and OSIRIS results the monthly zonal means of $\alpha_{750/1530}$ and its deseasonalized values (anomalies) at 21.3 km are plotted respectively in the panels (a) and (b) of Fig. 8. The deseasonalization of $\alpha_{750/1530}$ for the both instruments is justified, firstly, by the seasonality of stratospheric aerosols, which was reported in multiple studies including Hitchman et al. (1994); Bingen et al. (2004), and secondly, by the dependency of the limb radiances on the seasonal changes of SSA. The deseasonalized values for each instrument were calculated individually by subtracting an averaged $\alpha_{750/1530}$ over all corresponding months in the whole observation period from each monthly mean

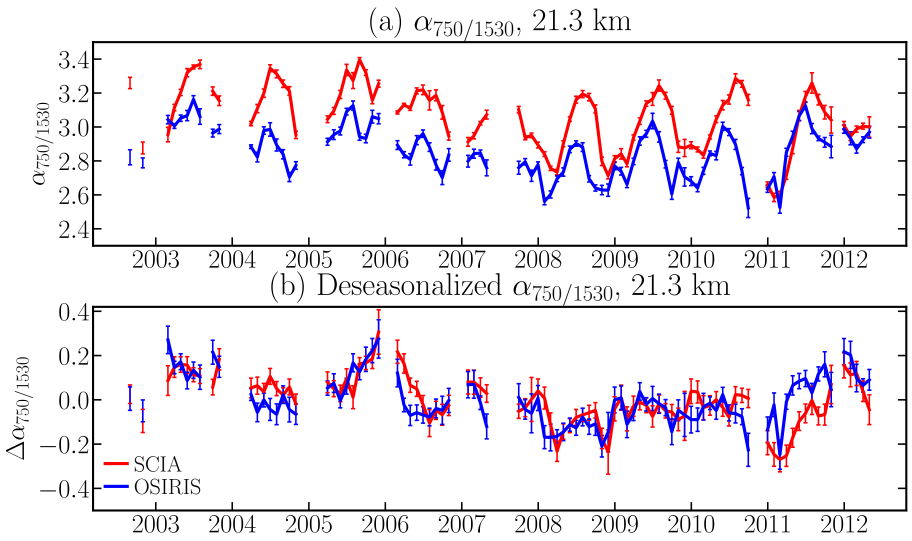

Figure 8. Zonal monthly mean (panel (a)) and deseasonalized (panel (b)) Ångström exponents ($\alpha_{750/1530}$) from collocated SCIAMACHY and OSIRIS measurements. Vertical bars show standard error of the mean.

value (e.g. the averaged $\alpha_{750/1530}$ for all the Julys from 2002 till 2012 was subtracted from the monthly mean value of each July in 2002-2012). Months with less than 10 collocations were excluded from the comparison. SCIAMACHY data is presented in red and OSIRIS in blue, the vertical bars show the standard error of the monthly mean value.

As seen in panel (a) of Fig. 8, the Ångström exponents retrieved from both instruments show very similar behavior, although the absolute values of $\alpha_{750/1530}$ from SCIAMACHY are systematically higher than those from OSIRIS. A high degree of consistency is found between the results from both instruments in the comparison of the deseasonalized time series (see panel (b) of Fig 8). Generally, the blue and red lines overlap or lay within the error bars and follow the same pattern, except at the beginning of 2006, when SCIAMACHY values are slightly higher, and 2011, when OSIRIS is slightly higher. However, even in those periods the differences are rather small (about 0.05-0.08). As the differences between Ångtröm exponents from both instruments are fairly constant with the time, and do not show signatures of any remarkable events (e.g. volcanic eruptions), it can be concluded, that they originate most probably from the technical specifications of the instruments and differences in the retrieval algorithms.

Summarizing Sect. 5, the following conclusions are made: aerosol extinction coefficients, obtained from SCIAMACHY PSD product agree with SAGE II within ±25%, i.e. within the reported accuracy of the obtained Ext from SCIAMACHY. The best agreement was acquired for Ext_{750}, calculated for both instruments from Ext_{525} and Ext_{1020} with $\alpha_{525/1020}$. Additionally, it was shown that the recalculation of Ext using the Ångström exponent can results in about 8% uncertainty. Ångström exponents from SCIAMACHY were compared to the ones from SAGE II and OSIRIS. The differences with respect to SAGE II results are about 40% at 18 km, decreasing to 20% at 21 km and to 10% at 30 km. With respect to OSIRIS, the agreement is much closer, with the relative differences between 4 and 7%. The differences between the instruments are smaller

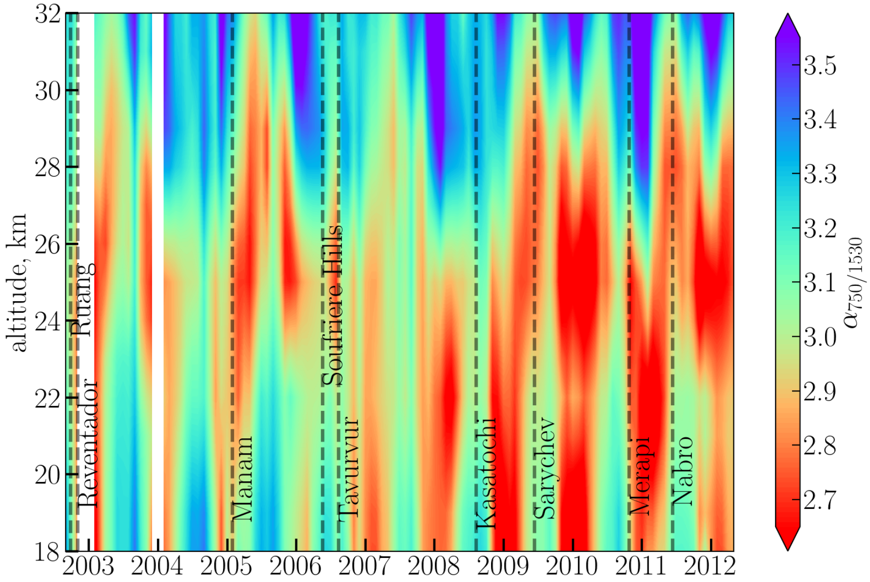

Figure 9. Monthly zonal mean values of the Ångström exponents ($\alpha_{750/1530}$) derived from SCIAMACHY limb data in the tropics (20°N - 20°S).

than the expected uncertainties, showing remarkable agreement between the instruments. The temporal behavior of $\alpha_{750/1530}$ anomalies is independent of any remarkable events, with the minor differences coming from the small technical differences of the instruments and retrievals. The much better agreement of SCIAMACHY aerosol values with OSIRIS than with SAGE II is explained by a better consistency of the data sets: the same measurement technique and the same spectral information were used to retrieve the parameters.

6 Discussion

In order to investigate the temporal behavior of the Ångström exponent and to understand its dependency on the PSD parameters, the SCIAMACHY data set recalculated from the PSD product was analyzed. Unfortunately, due to rejection of cloud contaminated profiles temporal sampling of the obtained product is too sparse to analyze volcanic plumes. For this reason the monthly zonal mean (20°S-20°N) $\alpha_{750/1530}$ as shown in Fig. 9 were considered. With exception of the upper altitudes (26-32 km), where the quasi biannual oscillation (QBO) pattern is obvious, the seasonal variation of $\alpha_{750/1530}$ represents the dominating pattern seen in Fig. 9. As it was mentioned in Sect. 5.3, the seasonality of stratospheric aerosols was discussed in several previous studies. To make the analysis of the Ångström exponent behaviour after the volcanic eruptions more clear, $\alpha_{750/1530}$ was deseasonalized using the same approach as discussed in Sect. 5.3. The deseasonalized $\alpha_{750/1530}$ time series are presented in Fig. 10. It should be noted, that in Fig. 10 the increased Ångström exponent values are shown in blue, and decreased in red, as the increased $\alpha_{750/1530}$ is often interpreted as a decrease of the aerosol particle size, and vice versa.

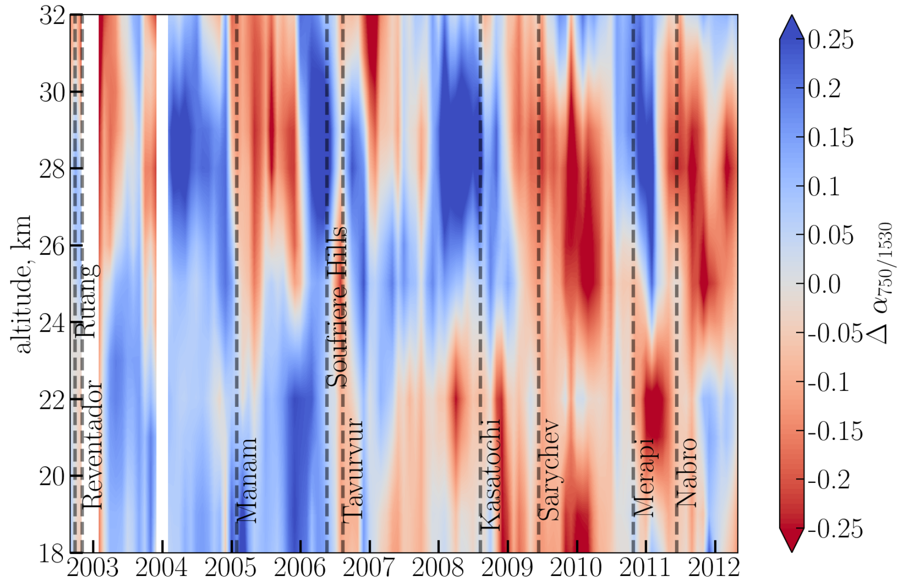

Figure 10. Deseasonalized time series (anomalies) of the Ångström exponents ($\alpha_{750/1530}$) derived from SCIAMACHY limb data in the tropics (20°N - 20°S).

Looking at Fig. 10, it becomes even more evident that the QBO pattern is well pronounced at altitudes above 26 km. This agrees with results reported earlier for SCIAMACHY aerosol products, e.g., Brinkhoff et al. (2015) showed similar patterns at around 30 km altitude for Ext_{750}. Later, the QBO signatures in R_{mod} and w were revealed by Malinina et al. (2018). The deseasonalized $\alpha_{750/1530}$ time series also demonstrate the influence of multiple volcanic eruptions. The slight decrease of $\alpha_{750/1530}$ was noticed after the Tavurvur eruption, and more significant after the extra-tropical Kasatochi and Sarychev eruptions. Almost no change in $\alpha_{750/1530}$ was observed after Ruang, Reventador and Manam eruptions. After the Nabro eruption $\alpha_{750/1530}$ increases at the 18-23 km altitude. As for R_{mod} and w from the same data set (Malinina et al., 2018), the changes in $\alpha_{750/1530}$ reach higher altitudes with a certain time lag (tape-recorder effect). Interestingly, the general behaviour of $\alpha_{750/1530}$ looks very similar to that of w (Malinina et al., 2018, Fig. 12). To evaluate it in more detail the dependency of $\alpha_{750/1530}$ on R_{mod} and w was analyzed.

It is well known, that $\alpha_{750/1530}$ is dependent on both, r_{med} and σ, but as R_{mod} and w are derived from these parameters, their impact on $\alpha_{750/1530}$ is not obvious. To investigate these relationships, the results from individual measurements in the tropical region over the whole observation period of SCIAMACHY are presented in Fig. 11. In panel (a), $\alpha_{750/1530}$ at all altitudes is presented as a function of R_{mod} and w, while in panel (b) the dependency of $\alpha_{750/1530}$ on r_{med} and σ is shown. The colors in Fig 11 depict the magnitude of $\alpha_{750/1530}$. From Fig. 11, it is clear that any particular value of $\alpha_{750/1530}$ can result from an infinite number of combinations of R_{mod} and w (or r_{med} and σ). However we note that retrieving a pair of R_{mod}/r_{med} and w/σ is not the only way to obtain the Ångström exponent. As was discussed in (Malinina et al., 2018), in the spectral interval from 750 to 1530 nm the radiance can be fitted by two out of three PSD parameters, for SCIAMACHY the

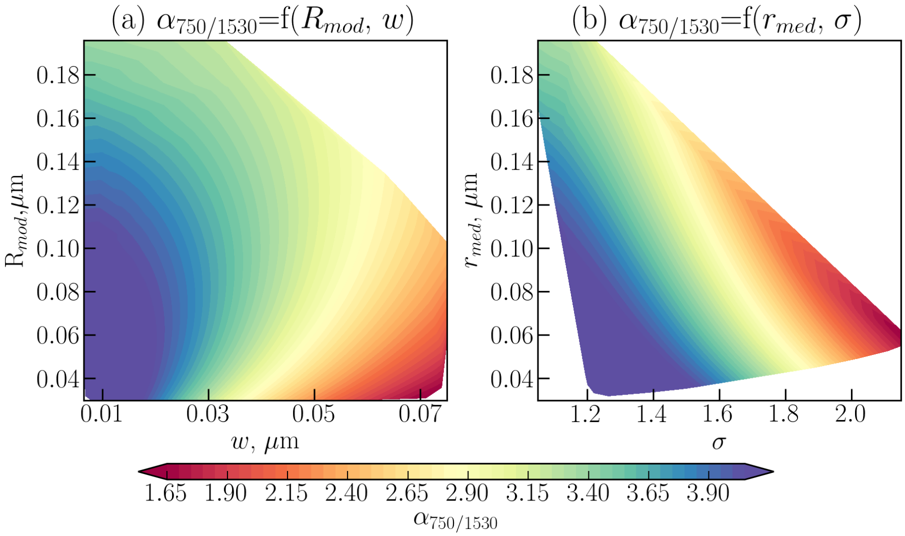

Figure 11. Dependence of the Ångström exponent ($\alpha_{750/1530}$) on mode radius, R_{mod}, and absolute distribution width, w (panel (a)). In panel (b) $\alpha_{750/1530}$ as a function of median radius, r_{med}, and σ is presented. Plot is based on the SCIAMACHY limb data in the tropics (20°N - 20°S).

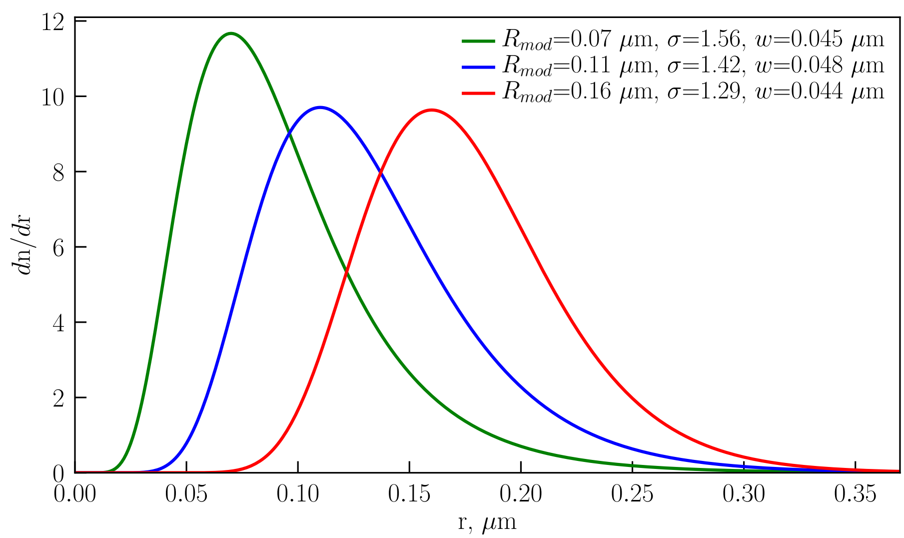

Figure 12. Aerosol particle size distributions with $\alpha_{750/1530}$=3.17. For convenience, N=1 cm^{-3}.

pair R_{mod} and σ was chosen as the limb radiances sensitivity to these parameters is higher than to N. However, it is possible to obtain a correct Ångström exponent also by retrieving, e.g., r_{med} and N (Rieger et al., 2014), although the accuracy of the PSD parameters may be not as high as for R_{mod} and σ.

Comparing panels (a) and (b) it can be seen, that $\alpha_{750/1530} = f(R_{mod}, w)$ is a non-monotonic function, i.e. up to a turn-around point the same value of $\alpha_{750/1530}$ is obtained by increasing both, R_{mod} and w, and then the same $\alpha_{750/1530}$ is a result of increasing R_{mod} and decreasing w. The function $\alpha_{750/1530} = f(r_{med}, \sigma)$ is monotonic with respect to both, r_{med} and σ, and there is the general rule: the larger is r_{med} or σ, the smaller is $\alpha_{750/1530}$. The same value of $\alpha_{750/1530}$ can be reached by increasing r_{med} and decreasing σ or vice versa. It is important to highlight, that completely different distributions might result in the same value of $\alpha_{750/1530}$. To illustrate this fact, we chose three pairs of PSD parameters with $\alpha_{750/1530}$=3.17, and plotted the distributions dn/dr in Fig. 12. The values of R_{mod}, σ and w used for the figure are listed in the legend. This figure disproves a widely spread belief often met in the scienfic discussions, that smaller value of an Ångström ~~exponents are~~ exponent is associated with the prevalence of larger particles and vice versa. As it can be seen, distributions with R_{mod}=0.07 μm and w=0.045 μm (green line) and R_{mod}=0.16 μm and w=0.044 μm (red line) have completely different amounts of large particles, but result in the same Ångström exponent. As it was already noted by Ångström (1929), α has only an "approximate coincidence with the average diameter directly measured". Thus, it can be concluded, that, firstly, there is no possibility to obtain a unique pair of PSD parameters from the known single value of $\alpha_{750/1530}$, and secondly, to provide a relevant information on the change in the particle size, $\alpha_{750/1530}$ should be accompanied by one of the PSD parameter. Here, we note, that our conclusions are valid for the Ångström exponent at one wavelength pair. If several Ångström exponents for different independent wavelength pairs are provided, more information on PSD can be derived. However, any conclusions about particle size based on the Ångström exponent at one wavelength pair without any additional PSD information are meaningless. Currenly, for all known space-borne instruments providing aerosol information in the stratosphere, only one value of Ångström exponent is reported in peer-reviewed publications, which makes our conclusions applicable to all of them. This statement is clearly not applicable to the datasets directly providing PSD information.

Summarizing Sect. 6, it can be concluded, that based on the available climatology $\alpha_{750/1530}$ can increase, decrease or remain unchanged after the volcanic eruptions. As for R_{mod} and w from the same data set, the tape-recorder effect after the volcanic eruptions as well as QBO signatures at upper altitudes (26-32 km) are observed. The pattern of $\alpha_{750/1530}$ changes is similar to that of the changes in w, although changes in both, R_{mod} and w, contribute to the changes in $\alpha_{750/1530}$. It was also shown, on the examples of the single measurements that infinite amount of R_{mod} and w (or r_{med} and σ) pairs result in the same $\alpha_{750/1530}$, and the statement that the large/small Ångström exponent means the prevalence of small/large particles is strictly valid only for increasing/decreasing r_{med} with σ remaining unchanged.

7 Conclusions

In this study, the stratospheric aerosol extinction coefficient and Ångström exponent have been discussed. From the investigation of the sensitivity of ~~limb-scatter and solar occultation instruments~~ space-borne measurements in visible-near-infrared spectral range to the aerosol particles of different size, it was shown that limb-scatter instruments are sensitive to the aerosol particles of smaller size, and thus, provide more accurate PSD information than solar occultation instruments, in particular, during periods with low aerosol loading. However, the sensitivity threshold of the occultation instruments can be improved, in case the UV part of the spectrum is considered. In contrast, occultation instruments provide aerosol extinction coefficients which are associated with smaller uncertainties than the ones from the limb instruments.

Here, we focus on the aerosol PSD product, which provides R_{mod} and σ (and recalculated w), obtained from SCIAMACHY limb measurements. In order to compare it with other space-borne instruments, aerosol extinction coefficients and Ångström exponents were recalculated using the PSD parameters. Error estimation based on the synthetic retrieval approach showed that the aerosol extinction coefficients are obtained with about 25% accuracy for the scenarios with high aerosol loading and with less than 20% uncertainty for the background period. It was also shown that by using the retrieved R_{mod} and σ from the same data set it is impossible to estimate N, or put another constraint on the parameters by implementing coupled or a consequent Ext retrieval. Ångström exponents calculated from the PSD parameters show less than 10% error for $\alpha_{525/1020}$ and less than 5% error for $\alpha_{750/1530}$. In the absolute values these errors are less than 0.2 and 0.15 respectively.

The recalculated aerosol extinction coefficients from the SCIAMACHY observations were compared to those from SAGE II. This comparison showed that differences are within $\pm 25\%$, which is within theoretically determined errors for SCIAMACHY. Ångström exponent ($\alpha_{525/1020}$) differences vary from 40% at 10 km to 10% at 30 km, with SAGE II values being systematically smaller. Furthermore, SCIAMACHY Ångström exponents ($\alpha_{750/1530}$) were compared to those from OSIRIS (another limb-scatter instrument). The relative difference between the instruments is decreasing from 7% at the lowermost altitudes to 4% at the uppermost altitudes. The absolute values of $\alpha_{750/1530}$ differ by less than 0.2, and both relative and absolute differences are within the theoretically determined errors of $\alpha_{750/1530}$ for those instruments. The time series analysis of the collocated data sets showed that the differences do not change significantly with time and are not correlated with any remarkable events, such as volcanic eruptions.

The Ångström exponent in the tropics was analyzed using the values, recalculated from SCIAMACHY PSD data set. It was shown that the monthly $\alpha_{750/1530}$ anomalies show distinct QBO signatures in the upper stratosphere, and the Ångström exponent can either increase, decrease or remain unchanged after a volcanic eruption. The analysis showed that changes in $\alpha_{750/1530}$ are driven by changes in both R_{mod} and w (or r_{med} and σ), and an infinite number of pairs of these parameters provides the same value of $\alpha_{750/1530}$. It was concluded, that it is impossible to derive any reliable information on the changes in the aerosol size based solely on the Ångström exponent for one wavelength pair. This can only be done if at least one of the PSD parameters is provided in addition.

Data availability. SCIAMACHY aerosol particle size distribution data is available after registration at http://www.iup.uni-bremen.de/sciaarc/. OSIRIS data set can be downloaded at http://odin-osiris.usask.ca/?q=node/280.

Competing interests. The authors declare that they have no conflict of interest.

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