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Atmospheric Measurement Techniques Discussions

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

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

General Comments:

This paper presents a new algorithm for the retrieval of aerosol particle size distribution (PSD) from SCIAMACHY. As a result of a sensitivity study of the three mode

parameters (particle number density N, mode radius Rmod, and standard deviation σ), the authors conclude that the sensitivity is higher for Rmod and for σ than for N, and in order to alleviate the problem of limited degree of freedom, they decide to retrieve the 2 first parameters and to assign the N parameter using ECSTRA, a climatology published in the literature. This paper is an important milestone since it concerns the first PSD retrieval from limb scattering measurements. The study has been conducted carefully, but the order of magnitude considered for the various mode parameters may be not well suited, leading, in my opinion, to erroneous conclusions. Fortunately, I don't see any reason why it would invalidate the methodology used. Further details are given below. The authors are invited to revise the English language: sentences are sometimes very long and confusing, and their structures and the use of some word is incorrect (e.g. many confusions between "a" and "the"). It might be useful to let read the manuscript by a native speaker.

Working on the comments of the reviewer, we found a typo in the formula, which defines R_{mod}. For that reason the main comment on the unrealistic order of magnitude of R_{mod} and σ is resolved, as the reviewer's calculations were done with the wrong formula. Based on the comments we have also realized, that some of our formulations might have been misleading, which resulted in the reviewer's partial misinterpretation of the manuscript. We hope, that the revised manuscript solves these issues. English language was improved in the revised version.

Specific comments:

Abstract:

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

Here we meant, that the usage of aerosol particle number density together with aerosol particle size distribution parameters is more optimal approach to describe stratospheric aerosol than its extinction coefficient, because extinction coefficient can be calculated with these parameters but not vice versa. The text has been revised to avoid possible confusion.

1. Introduction:

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

The comment repeats the previous one, and in the marked part of the manuscript particle number density is not mentioned. For that reason we have considered this comment as a typo.

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

The main source of OCS has been specified.

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

The information about OCS emissions has been added.

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

The paragraph has been rewritten in accordance with the reviewer's comments.

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

We meant, that three particle size distribution parameters describe stratospheric aerosols in more optimal way, than aerosol extinction, because aerosol extinction can be recalculated from the PSD parameters. The paragraph has been rewritten to avoid possible confusion.

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

The paragraph has been rewritten to avoid possible confusion.

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

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

The text has been revised to avoid terms "linear space" and "logarithmic space" instead "$\frac{dn}{dr}$ distribution" and "$\frac{d\ln(n)}{d\ln(r)}$ distribution" have been used. Eq. (1) has been changed respectively.

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

"Spectral information" has been replaced by "limb radiances".

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

The paragraph has been revised and the definition has been added.

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

The word "a priori [values]" has been replaced by "initial [values]". The choice of the

initial value is described in the following paragraphs.

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

We have rephrased this part of the text to make it less confusing. The statement, that value of 1% was selected empirically to achieve a trade-off between the retrieval stability and sensitivity has been added to the text.

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

The issue has been addressed in the previous comment. The word "empirically" has been added.

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

Although there are not so many data above 35 km, that is true, that H_2SO_4 is strongly decreasing at higher altitudes and is presented just in the gas phase. For that reason we retrieve aerosol particle size distribution parameters from 18 to 35 km. However, it is known that there is some aerosol above 35 km (e.g. meteoritic dust), but in very small concentrations. It is taken into consideration by our aerosol number density profile, which is about 0.5 cm^{-3} at 35 km and is decreasing exponentially with increasing altitude. We define the aerosol profile above 35 km and below 18 km as sulfate aerosol in order to avoid jumps and unreasonable values at the lowermost and the uppermost retrieval altitudes. In Sect. 3.2 we show, that the trustworthy altitude range is 18-32 km.

p.6, l.30-32: Please specify the issues.
The issues have been specified in the revised manuscript.

p.6, l.33: Please explain.

In the revised manuscript the phrase "To reduce profile oscillations" has been replaced by "To minimize the need for constrains and avoid additional errors related to e.g. altitude interpolation".

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

Based on this and next comments of the reviewer we realized, that there might be a misunderstanding of the general concept of our study. In our method the particle

number density profile is considered to be known, and R_{mod} and σ are retrieved. We believe, that based on the text of the original manuscript the reviewer assumed, that one value of N was used for all altitudes, but that is not the case. A background number density profile from ECSTRA model was used. The text of the manuscript, where particle number density has been mentioned, has been revised to resolve this issue. The profile is changing exponentially from 15.2 cm^{-3} at 18 km to 0.5 cm^{-3} at 35 km, thus, the vertical variations of number density, mentioned by the reviewer, were taken into account. The uncertainty due to changes in the particle number density profile with the time remains an issue, but as it was shown in the Fig. 1 of the manuscript and further with the synthetic studies, the sensitivity of the algorithm to N is rather small. Our number density profile is of the same order of magnitude as the one for $r > 0.15$ presented in Fig. 17 of Kremser et al., (2016). Also in Deshler, (2008) in Fig. 5 the results of the campaign in Darvin, Australia (23 November 2005) are presented. The profiles for $r > 0.15$ are very close to the one, which we are using. Since Darvin is located in the tropical zone, and time of the campaign is within the SCIAMACHY operating period, we believe that used particle number density profile is a good approximation. Also important to mention, that we believe that the variation of number density profile within the factor of 2 is quite realistic for SCIAMACHY observation period, as at the time there were no colossal Pinatubo-like eruptions, all the eruptions were times smaller. According to Fig. 1 in Deshler, (2008), where the particle size distributions 1 year, 3 years and 15 years after Pinatubo eruptions are presented, the variations of N_1 (fine mode particle number density) are within a factor of 2 for background and Pinatubo period, and the coarse mode number density (N_2) is rather small in comparison to N_1).
The reviewer also mentions very small R_{mod} and σ, but as we realized from the comment to p.10, Table 10, there was a mistake in the formula for R_{mod}, which lead to that conclusion. The values for R_{mod} and σ are realistic and coincide with the ones from Bingen et al, (2004), and Deshler et al., (2003).
Another important remark, Fig. 1 shows the intensity for one tangent height (25 km),

and thus the altitude variations of the particle size distribution parameters do not play an important role there. The values of R_{mod}, σ and N near the tangent point are most important.

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

Please see the answer to the previous comment. Text has been corrected to resolve the issue.

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

We disagree with the reviewer on this point. First, as was shown in the manuscript, the R_{mod} errors in the small scenarios are around 0.01 μm (relative error 10-20%). Implementation of the mean profile would noticeably increase uncertainty for the background cases, while for the volcanic cases, where the relative error is about 20-25%, the uncertainty will remain. Second, there are no known assessments of changes in the particle number density profiles during the Envisat operating period, all the known assumptions were based on SAGE II climatology, which included the colossal eruption of Pinatubo, which is not representative for 2002-2012.
SCIAMACHY and SAGE II overlap period is considered to be volcanically quiscent,

thus the lower biases cannot be a result of the particle number density underestimation. This comparison is in general quite problematic. First, we emphasized, that the sample is quite small. Second, as the reviewer mentions, SAGE II is insenstive to the smaller particles typical for the background period, thus, SAGE II effective radius is expected to be biased high. For that reason the relative error of 30% even with systematic negative bias considered to be a good result.

p.8, l.26: What do the authors mean by "geometric variations in tropics" ?

The phrase has been replaced by "variations in observational geometry (viewing angle and solar zenith angle)".

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

Here we meant the shape of the averaging kernels. The sentence has been changed to avoid a possible confusion.

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

As we mean the set of parameters, used for the retrieval we think that the word "settings" suits better, than any other.

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

For the synthetic simulations we chose just one combination of the viewing angle and solar zenith angle, which are typical for SCIAMACHY measurements in the tropics. In contrast to widely used transmission measurements, the single scattering angle and solar zenith angle for most of the limb viewing instruments, and SCIAMACHY in particular, are dependent on the geographical latitude. This has been clarified in the revised manuscript.

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

We distinguish 3 types of the simulations, based on the perturbations in the particle size parameters. The explanation has been added to the manuscript.

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

The term "unperturbed" is related to the perturbations in the particle size distribution parameters. To reduce the use of the word "(un)perturbed" and "perturbation" the text has been slightly changed.

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

As explained before, the altitude dependency of the particle number density profile is

accounted for, and now it is clearly stated in the manuscript. For the synthetic studies R_{mod} and σ were modelled without altitudinal changes, because there is no information on the realistic behavior of this parameters with the height. Furthermore, the errors have a weak dependency on the specific values. R_{mod} and σ used in the study have the same order of magnitude, as the ones from (Desheler et al., 2003; Bingen et al., 2004; Deshler, 2008). We revised the sentence in the manuscript.

p.10, Table 10: The author might usefully add the values of rg and w. If my understanding is right, the values of rg for the 5 scenarios are: {17, 18, 11, 6.6, 6.6}.10-3 micron, what is irrealistically small, and in the size range of condensation nuclei, which seems not realistic to describe the cases mentioned here. As mentioned in Kremser et al., 2016, only median radii $>$ 0.2 micron are observable for instruments like SAGE. SCIAMACHY's spectral range is not very different, and this instrument is not able to observe such small particles. Values of w are found accordingly very small, as 0.12,0.09, 3.7, 1.24, 1.24.10-3 micron.

We are very thankful to the reviewer for that comment, as it helped us to identify a typo in the formula for R_{mod}. In reality $R_{mod} = r_g/exp(ln^2(\sigma))$, the formula was corrected in Sect. 2.2. Our calculations though were correct, and r_g and σ values are realistic and consistent with other studies (e.g. Deshler et al. (2003); Bingen et al. (2004a, b); Deshler (2008)).

The true values for r_g and w were added to Tab. 1 and 2 of the manuscript.
The reviewer is appealing to Kremser et al. (2016), and authors of that paper are citing Thomason et al. (2008). The introduction of Thomason et al.(2008) ends with a phrase "Since visible and near-infrared wavelength aerosol extinction is insensitive to particles with radii smaller than 100 nm, the robustness of SAD (surface area density) estimates based on these measurements is questionable. Therefore, we have also included a study of the limitations of SAD estimates based on SAGE II

aerosol extinction measurements." We agree, that aerosol extinction in the occultation measurements is insensitive to the small particles ($r < 0.1$ μm), although for the limb scattering measurements the situation is slightly different. As can be found e.g. in Chandrasekhar (1960) for the scattered light $I \sim \sigma_s N p$, while for the transmission $\ln I \sim \sigma_s N$, where I is intensity, σ_s is aerosol extinction cross section, and p is the scattering phase function, which depends on (R_{mod}, σ). Thus, limb radiances are differently sensitive to the aerosol parameters than the occultation measurements. As an example we simulated the intensities for 3 different distributions: R_{mod}=0.06 μm, R_{mod}=0.08 μm, R_{mod}=0.10 μm as well as the intensities with no aerosol. For all distributions σ was chosen so, that $w \approx$0.01 μm. We calculated the relative differences of the intensities ($(\Delta I = I_{aer} - I_{noaer}) * 100/I_{noaer}$). This relative differences are plotted in the Fig. 1 in the supplement to the answer. As it can be seen from the Fig. 1, for the distribution with R_{mod}=0.06 μm ΔI is 1% which is around the sensitivity threshold, although for the distributions with R_{mod}=0.08 μm, R_{mod}=0.10 μm ΔI is about 5 and 15% respectively. Thus, we believe, that SCIAMACHY limb measurements are more sensitive to the smaller particles than SAGE II.

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

The calculations of the reviewer were not correct, as there was a typo in the formula (see previous comment).

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

As mentioned in the two previous comments, there was a typo in the formula, and

because of that the calculations of the reviewer were not correct. The scenarios are realistic and are consistent with Deshler et al. (2003); Bingen et al. (2004a, b); Deshler (2008).

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

This criterion was chosen to eliminate the distributions with the unrealistic values rather than clouds. As it was mentioned in the comment to "p.10, Table 10", the distributions with R_{mod}=0.06 μm and σ $\approx$1.1 is on the lower limit of the sensitivity of SCIAMACHY measurements, but in reality this distribution is highly unprobable. According to our data for the distributions with R_{mod}=0.06 μm mean σ value is around 1.7. For the distributions with R_{mod}=0.03 μm and σ around 1.6 the threshold ΔI value of 1% is reached. We have changed the text of the manuscript to make this paragraph more clear.

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

The criterion was applied everywhere, however, as follows from Fig. 1 in (Bourassa et al., 2010), the daily mean values for aerosol extinction at 750 nm never reached the value higher than 0.0015 even after the eruption. In their research Bourassa et al.(2010) use 0.0015 as the highest value for their color bar, thus we believe that this value will not exclude volcanic plumes.

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

In this manuscript we do not aim to analyze volcanic plumes, but rather to understand the general state of the atmosphere in 2002-2012, and for that purpose monthly mean values serve quite well. We added a statement about the purpose of the evaluation, and the issues of the monthly mean values in the revised manuscript.

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

The word "small" was changed to "smaller" and citation to Bingen et al.(2017) was added.

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

The distributions are individual profiles. This is explicitly mentioned in the revised manuscript.

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

As N is just a multiplicative factor of $n(r)$, and N is decreasing exponentially with the height, we chose N=1 cm^{-3} for each altitude to show the form of the distributions with the same y-axis for all altitudes. The explanation has been added to the text.

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

By "heavier tails" we meant "stronger relative contribution of the larger particles". In the revised manuscript this formulation has been corrected.
In the revised manuscript we also mention the eruption on 4 March 2006. This explains, why at 18 km the cyan line did not return to the background state as indicated by the green line.

5. Comparison with SAGE II:
p.14, ll. 18-25: See comment above.

Please, see the answer to that comment.

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

Conclusions have been slightly revised. However as we believe the main issue raised by the reviewer has been clarified, we have not done any major revisions.

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

Changed to "for now unique".

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

The clarification of this phrase is provided in the revised manuscript.

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

The systematic negative bias has been mentioned.

Technical corrections:
*p.1, l. 7: Please revise the sentence.*The sentence has been revised.

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

The suggested phrase might be misinterpreted as keeping the particle number density constant with the height. This is why we left the original phrase adding the word "profile": "the aerosol particle number density profile remains unchanged".

p.1, l.14: "Overall" instead of "Generally" ? Has been replaced by "typically".

p.2, l.5: "colossal" : I am not sure this word is used in this context.

According to the database of the Smithsonian Institution, cited in the manuscript, and other sources Mount Pinatubo eruption 1991 had VEI=6, which is classified as colossal eruption.

*p.2, l.12: "for modelling the processes"*Corrected.

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

p.3, l.3: "Otherwise". Has been change to "Additionally".

*p.3, l.4: 'unambiguously"*As we haven't found any place, where this word occurs at the marked page and line, we cannot address this comment.

*p.3, l.9: "Stratospheric aerosol properties"*The sentence has been revised.

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

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

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

p.3, l.31: "ever" instead of "possible". The sentence has been rephrased, and "possible" has been replaced by "among".

p.4, l.3: "international"; otherwise the country should be mentioned. Corrected.

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

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

*p.4, l.21: "indicating" instead of "responsible" ?*Corrected.

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

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

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

*p.5 and further: "a priori" instead of "apriori" !*Corrected.

*p.6, l.11: "the root mean square between two subsequent iteration": unclear.*Sentence has been revised.

p.7, l.10: "on a statistical approach". Corrected.

*p.9, l. 7: Please revise the sentence.*The sentence has been removed.

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

p.10, l.11: "it is obvious". Corrected.

p.13, l.7: "its" Corrected.

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

*p.14, l.15: "compared to" instead of "opposite to".*Corrected.

p.15, l.7: "an instrument" Corrected.

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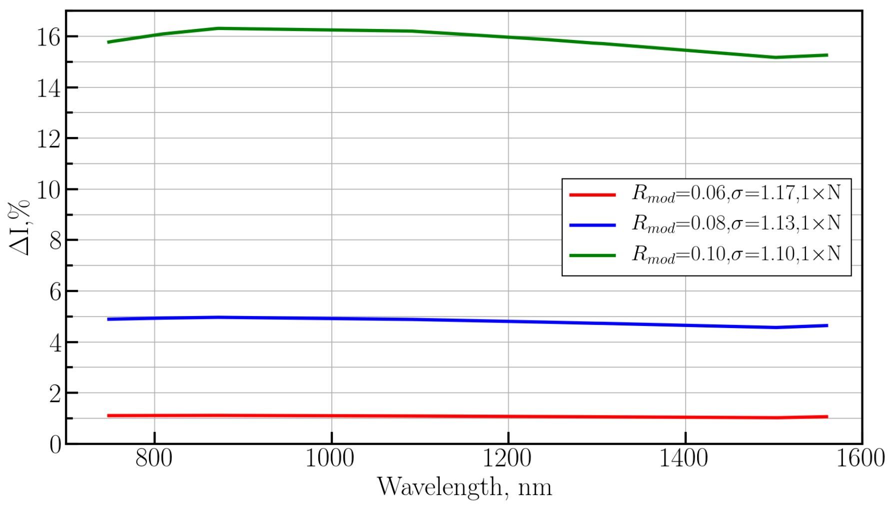

Fig. 1. The relative differences of the intensities ((ΔI = Iaer − Inoaer) * 100/Inoaer) for the scenarios with and without aerosol loading. Rmod and σ for the scenarios are shown in the legend.