

Referee Report on:  
Aerosol particle size distribution in the stratosphere  
retrieved from SCIAMACHY limb measurements

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**General Comments:**

The explanations and modifications of the manuscript brought by the authors greatly improve the quality of the paper and address the main issues in a satisfactory way. Also the quality of the English language is greatly improved.

I still have a few minor issues and questions I think worth to be addressed.

**Specific comments:**

2. Instrument and applied algorithm:

p.6, 1.29-30: “The particle number density (...) at 35 km”. I suggest slightly modifying the sentence to make it fully clear that this is the model you considered, and no observation. E.g.: “The particle number density,  $N$ , is assumed to decrease (...)”.

My previous comment on p.6, 1.25: Above 35 km, it is known that all  $H_2SO_4$  is only present in gas phase. It is irrelevant to retrieve sulfate aerosol above this altitude.

*Authors' response: 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  $0.5 \text{ cm}^{-3}$  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.*

This response is, in my opinion, only half satisfactory. (1) The choice of the aerosol number density takes indeed into account the presence of particles above 35 km, but the difference in composition (mainly meteoritic dust, unlike the sulfate aerosol dominating at lower altitudes) is not taken into account in the size distribution retrieval. (2) Even if the authors are able to show afterward that neglecting the difference in aerosol composition has no consequence on the final result or even may help stabilizing the solution, I find problematic that the difference in composition is just ignored, what looks like propagating wrong information in a scientific journal (although the authors admittedly use the expression “\*are assumed\* to be sulfate droplets”). I think there is no problem to discuss this point in the paper using the response given in the author's reply, saying that despite the difference in aerosol composition below and above 35 km, a

similar composition is assumed to simplify the processing and to stabilize the solution, and that they show later in the paper that this is without consequence on the validity range of the solution.

### 3. Sensitivity studies:

General comment: I understand now the authors' rationale, and why my remark about the typical variability of  $N$  of several orders of magnitude was not relevant. I think that, to be fully clear and convincing, the authors might consider writing at the beginning of the section that they are assessing the sensitivity of the solution found at a given location. It looks obvious, but it seems I missed that point, and the other referee seems also to have some problem with this study.

Then, maybe a very clear way to approach the problem would be to look in the literature (Bingen et al.; Deshler et al., etc.) what is a maximal range for the variability of the 3 parameters (i.e. large value with respect to the typical uncertainty found for any ground-based and space-based technique). If the variation used in the present paper for the different sets of parameter is large with respect to this typical range assessed from the literature, I think the rationale followed by the authors will be fully convincing.

The explanation given to Referee 2 using the expression of the scattered light intensity is also quite useful to illustrate the reason of smaller sensitivity of the size distribution with respect to  $N$ . I suggest adding it in the text.

I also wonder if this study leads to satisfactory results for cases of volcanic aerosols. The variability observed during the ENVISAT period was up to a factor  $\sim 8$  for the extinction (thus probably on the same order for  $N$ ) in the UTLS, see e.g. Kremser et al., 2016 (op. cit.). Obviously, such cases are particularly interesting and probably the most wanted (cf. the importance of tropical eruptions during the period covered here). Hence, I think it is relevant to add some comment on such case – even if the authors have to state that the study is not valid in such a case.

p.11, l.4: Is the significant increase of the deviation in  $w$  up to 40% in the “volcanic ( $2N$ )” scenario related to the fact that a background size distribution is assumed in the calculation of the forward model, making it less appropriate to describe a volcanic case ?

p.11, l.21: See my remark 2 paragraphs above.

My previous comment: 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).

*Author's response: 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  $0.01 \mu m$  (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.*

(...)

I am afraid there must be a misunderstanding. Is the method followed by the authors as follows:

1. Choice of a baseline scenario => gives the “true values”; e.g.:
  - a. “small” =>  $\{R_{\text{mod}}=0.06 \mu\text{m}; \sigma=1.7, N \text{ following the exponential vertical profile described in p.6, ll 29-30}\}$ ;
  - b. “volcanic (2N)” =>  $\{R_{\text{mod}}=0.20 \mu\text{m}; \sigma=1.2, [N \text{ following the exponential vertical profile described in p.6, ll 29-30}]x2\}$
2. Choice of a set of perturbed scenarios; e.g.:
  - a. “small” =>  $\{R_{\text{mod}}=0.06 \mu\text{m}+\text{Gaussian noise}; \sigma=1.7+\text{Gaussian noise}, N \text{ following the exponential vertical profile described in p.6, ll 29-30}\}$ ;
  - b. “volcanic (2N)” =>  $\{R_{\text{mod}}=0.20 \mu\text{m}+\text{Gaussian noise}; \sigma=1.2+\text{Gaussian noise}, [N \text{ following the exponential vertical profile described in p.6, ll 29-30}]x2+\text{Gaussian noise}\}$
3. For these perturbed scenarios: computation of the corresponding limb radiance
4. For the computed “perturbed” limb radiance, retrieval of  $R_{\text{mod}}$  and  $\sigma$ .
5. Comparison of all retrieved “perturbed”  $\{R_{\text{mod}}, \sigma\}$  with the initial set chosen for the baseline scenario in point 1.

?

If this is what the authors are doing, then I don't know why they call the 3<sup>rd</sup> scenario “unperturbed” nor why the “true values” of  $\{R_{\text{mod}}, \sigma\}$  have such a behaviour in the case “volcanic (2N)” in figs. 4-6, but I discard my comment on the case “volcanic (2N)”.

On the contrary, if, in the case “volcanic (2N)”:

- the baseline scenario is  $\{R_{\text{mod}}=0.20 \mu\text{m}; \sigma=1.2, N \text{ following the exponential vertical profile described in p.6, ll 29-30}\}$ ,
- the perturbed scenarios are  $\{R_{\text{mod}}=0.20 \mu\text{m}+\text{Gaussian noise}; \sigma=1.2+\text{Gaussian noise}, [N \text{ following the exponential vertical profile described in p.6, ll 29-30}]x2+\text{Gaussian noise}\}$ ,

then I maintain my criticism, because the Gaussian distribution of all perturbations should be centred on the baseline scenario, which represent the “truth” you want to retrieve.

The authors should clarify the method used in the paper.

#### 4. Results and discussion:

p.13, l. 14: I know I used the expression “individual profiles” (vs. “monthly profiles”) in my previous referee report, but I am not sure that, here, out of the context of my previous remark, this expression is clear of any reader. The authors might consider being more explicit, by using for instance an expression such as “profiles retrieved from individual measurements”.

#### 5. Comparison with SAGE II:

p.15, ll. 4 and 14-16: Considering the time series depicted in Fig. 15, especially at 18.0 km, I must say I don't find that the "tendency to show the same features" is striking, nor that differences around 30% (depending on the reference chosen, it might be estimated to more than 50% in some cases) look particularly a good performance, since this difference exceeds from far the variability found over the whole 3-year period. Therefore, I suggest removing the part of the sentence about the tendency to show the same features in l.4, and the highly subjective estimation of "good result": they weaken the author's argumentation and do not bring much to it. The discussion concerning the clear bias and its interpretation is far more interesting.

#### 6. Conclusions:

p.15, l.24: Please qualify in "for the considered unperturbed N profile".

Additional comment for this section: As discussed above, in the forward model used to retrieve the extinction from limb scattering sounders, the assumption of background aerosols is used, and it is not clear to me in which extend this assumption influences the particle size retrieval in case of volcanic aerosols. This point should be at least mentioned, even if the answer to this question is unknown or uncertain.