Overview

The authors present an improvement on a previous technique for inferring particle size distribution (PSD) parameters from the SCIAMACHY data record. The proposed technique performs a 2-parameter (r_g and σ_g) retrieval that is *not* limited to the tropics (an improvement over Malilina et al. 2018). The authors evaluated the impact their assumptions have on the end products and compare their PSD values (including the calculated effective radius (r_e)) to those measured directly by the University of Wyoming optical particle counter (UWY OPC) as well as those inferred from the SAGE II and SAGE III/M3M instruments. Further, they used their derived PSD parameters to calculate extinction coefficients (they labeled this *Ext*, I refer to this a k herein) and compared these to extinction coefficients as measured by the SAGE instruments and OSIRIS.

Overall this is an interesting paper that the community will benefit from. Overall, it is well written and the major claims are more-or-less substantiated (details below) and I believe this should be published if the authors can satisfactorily address the more salient points below.

Finally, I want to congratulate Dr. Pohl and the coauthors for the high-quality work that went into preparing this manuscript.

Major Remarks

The stated goal of this work is to extract PSD parameters (for the sake of brevity I include r_e under the PSD label as appropriate) from SCIAMACHY data. This can be best achieved through comparison with the UWY OPC record, but comparison with OPC data is very limited. Instead, the authors devote a substantial portion of the paper to comparing their PSD parameters to those derived from the SAGE missions. Ultimately, this results in an evaluation of the assumptions in each model, which is a distraction from the intended purpose of this work. Direct comparison with the OPC record removes at least half of these assumptions and gets to the heart of the matter. This comparison should be expanded.

An alternative to direct comparison with OPC data is using the SCIAMACHY-derived PSD parameters to calculate extinction coefficient. While the authors did this the evaluation was presented in a bulk-statistics manner (Figures 5 & 8) and it would have been much more informative to expand these figures to include more meaningful information (please see specific comments below). Further, this type of comparison removes the assumptions that went into the SAGE estimates and thereby provides a more robust and meaningful comparison.

Another alternative is to use the SCIAMACHY-derived PSD parameters to calculate backscatter coefficients and compare those directly to the numerous ground-based lidars as well as CALIOP. This seems like a grossly overlooked opportunity (a potential gold mine of data) that would have significantly increased the number of intercomparison opportunities as well as the geographic coverage. If this should not be don then can the authors at least address, in the paper, why this should not be done?

There are numerous ambiguities throughout the paper that must be addressed before publication. Without correction the reader cannot understand the presented work and cannot reproduce it.

Finally, the current version of this manuscript suffers from a substantial flaw that prevents the reader from understanding and appreciating the impact and applicability of this work. The authors state that the intent of this method is to infer r_g and σ_g from SCIAMACHY data, but limit their discussion of these parameters. Instead, the authors spend more time discussing the comparison to SAGE-derived PSD parameters that are strongly dependent on the assumptions that go into the SAGE algorithm. Further, the reader is not afforded the opportunity to see the overall performance of the SCIAMACHY-derived PSD estimates. Indeed, inclusion of PSD time-series plots (whether line plots with error bars or contour/mesh plots) would communicate a wealth of information to the reader not only on how the PSD parameters changed over the lifetime of the instrument (including volcanic impacts), but would also inform the reader of the stability of this retrieval algorithm. Such a figure would no longer limit the authors to coinciding with other instruments and would allow the authors to display the entire SCIAMACHY PSD record (all within a single figure!). In my view this paper cannot be published without this type of information content. To publish without this information leaves the reader with a knowledge gap that should not be there. Why is this so important? Because this informs the reader and end-user of how stable and reliable these estimates are. I agree that, in aggregate, comparison with SAGE/OSIRIS is generally good, but there are many unanswered questions. Does the PSD algorithm become unstable at certain altitudes? Does it perform better seasonally? How much do volcanic perturbations influence the PSD estimates? What about wildfires events? Without answering these questions I cannot ascertain the utility of these estimates.

This is a potentially fantastic paper, but in its current state it is incomplete. For these reasons I recommend that the paper undergo major revisions and be resubmitted for review.

Specific Remarks

- **page 2, line 44:** "catalysers" should be "catalysts"?

- **Section 4:** There is a lot of repetition within this section. It reads as if it was written twice and never cleaned up. Please consolidate the information and rewrite to be concise and precise.

- **page 7, line 209:** Why a relative humidity of 0%? Does your algorithm have some dependence in RH? This seems important since an RH of 0% is never true.

- **page 7, line 212:** Please define R_{mod}.

- **page 7, line 212:** You stated that R_{mod} and σ_g are arbitrarily chosen, but why these specific numbers (σ_g of 1.37 is a precise number, why not 1.4)?

- **page 8, line 220:** What is meant by "Above 35 km, the PSD profile remains unchanged."? Please provide a reference to support this claim.

- **page 8, lines 220–222:** You use both r_g and R_{mod} in this sentence. I understand the difference between the 2, but it seems you use them interchangeably here. Please clarify.

- **page 8, lines 220–224:** Herein you stated that r_g has a lower-limit of 0.05 μm and σ_g is not limited. Surely r_g has an upper limit as well as a lower limit. Surely σ_g was also limited. Not to be pedantic, but σ_g cannot be less than 1...could it, in theory, be >5? Could r_g be 10μm? These values must be fundamentally limited by your model, please provide those limits here.

- **page 8, line 225:** Here you state that "step 2" solves for 3 parameters (r_g, σ_g, and albedo). Section 2 stated that "step 2" *only* solves for r_g and σ_g. Please clarify.

- **page 9, line 255:** Please confirm that the 0.15 and 10.0 μm values are radii and not diameters (newer OPC instruments have a lower limit diameter of $\approx$150 nm).

- **page 10, lines 301–302:** This is not correct. The grating spectrometer did not include the 1550 nm channel. The 1550 nm channel was an InGaAs photodiode. Please clarify for the reader.

- **page 12, line 356:** "Their PSD profiles are specified below." I assume this refers to Table 1. Please include reference to appropriate table or figure.

- **Figure 1:** I am not colorblind, but I still have a very difficult time reading this figure. This is one of the key figures of your paper. Please update to make it more readable.

- **page 13, line 375:** "...with latitudes north of 26°N in summer and 23°S in winter." It is unclear what this means. Do you never go farther south than 23°S? Please clarify.

- **Sections 6.1 and 6.2:** This is a critical aspect of this paper. Ultimately you do not know the PSD parameters so you cannot definitively calculate the error in your derived PSD parameters that is caused by the Lambertian assumption and the assumed N profile. Therefore, how do you propagate the error from these 2 assumptions into your estimates for r_g, σ_g, and extinction coefficient (e.g., in Fig. 4)? Is this uncertainty ignored for the rest of the paper, or is it accounted for?

- **page 14, lines 403–404:** Here you state that you averaged over all angles from 20°– 96°. If I understood Section 4 correctly the variation in scattering angle has no impact on this process. If that is correct then please ignore this comment. If there is a dependence then would you please clarify here.

- **Figures 2 & 4:** Do I understand this correctly that there is no variability in the inferred number density? You should be able to infer N as well (I thought that's what you did in Figure 7), so I expect some spread in the profiles. Please clarify.

- **page 15, line 413:** "...correct selection of the a priori N is crucial..." That's not how I interpret Figure 2. It looks like the algorithm is sensitive to the N profile and how close it is to reality. However, "crucial" may be an overstatement. The r_g is mostly within $\pm$25% up to $\approx$30 km. The r_e is even better. I think the text, as written, misleads the reader. Please revise.

- **page 15, line 418:** "...latitudes south of 26°N in summer and 23°S in winter." Please see similar comment above.

- **Figure 3:** Please consider plotting panel (a) on a log scale. Maybe not necessary, but it helps readability. If the authors disagree then please disregard this comment.

- **page 16, line 424:** "2002 and 2011" Why not "2002 and 2012"?

- **page 16, line 426:** "...these processes are evidenced in Fig. 3." This level of information cannot be ascertained from a simple extinction coefficient plot. Please revise.

- **Figure 4:** How were the relative error statistics calculated? Did you first calculate the relative errors then average, or did you calculate the average r_g then the relative error?

- **Figure 4:** This is possibly the most important figure in this paper (comparing to OPC) and should receive more attention. You put all of the data into one plot (I realize the OPC data are sparse), but this leaves me wondering if information is lost in the bulk statistics. Did volcanic activity significantly change the performance of your method? Breaking this analysis into 2 paradigms (volcanically perturbed/not-perturbed) would be illuminating. Without this information it is difficult to realize the value of this method.
- **page 17, lines 441–442:** "...and one based on balloon-borne measurements over Wyoming before 2002..." This is confusing. This sounds like you use an OPC-based climatology as your N profile, which has the potential to significantly bias your evaluation. This raises several critical questions regarding the methodology.
  1. Are you now using an OPC-based climatology for your N profile? If so, did you also use this climatological profile in creating Figure 2?
  2. You stated this N profile is based on WY OPC data collected before 2002. Does this include data collected throughout the entire record (i.e., back to 1971)? If so, how do you handle the differing OPC instruments? How do you handle extreme outlier events like El Chichon and Pinatubo? How did these events influence the overall performance of your algorithm?
  3. From Figure 4, it looks like the OPC-based N profile yields better performance than the ECSTRA profile (better errors, the profile shape is in better agreement with reality, etc.), yet you never state which N profile you use in the operational algorithm. I assumed you use the ECSTRA profile, but Figure 4 indicates the OPC profile is better. Please clarify.
- **page 18, lines 448–449:** "...which can be explained by a small reference value (Fig. 4 (i))." Did you mean Fig. 4 (d)? Also, I'm not sure Figure 4 supports this claim. Extinction coefficients on the order of 1E-4 (i.e., z $\leq$24 km) are not small. What you are dealing with here is, in fact, relatively large differences in the derived extinction coefficients.
- **Figure 5:** You allude to this figure throughout the text and reference statistics from this figure. However, I cannot read this figure. There are so many colors on top of each other I cannot tell where they all begin/end...and I cannot tell if the quoted numbers quote are correct (I assume they are, I just cannot verify). Please improve the readability of this figure.
- **Figure 5:** Why the stark difference between this figure and Figure 4 (i)?
- **Figure 5:** Why not use the same method to calculate error as used in Figure 4?
- **page 19, lines 489–490** "...with the differences decreasing with altitude." This is not what Figure 6 shows. All differences increased (except panels f and g). Please clarify.
- **Figure 6:** This figure exemplifies why the comparison with OPC (or even a comparison of k) is of more value than comparing with the SAGE-derived PSD parameters. The extreme slope in panels b–e indicates a systematic error in the SAGE PSD values. How is the reader to draw meaning from this analysis? Can the authors account for this extreme slope? To be honest, the extinction comparison looked promising, so I was somewhat shocked to see this odd behavior in these profiles (I do note that the authors plotted here data from both SAGE missions and that this is a figure for r_e (a derived product that is based on derived

products, which adds another layer of obfuscation)). Given the ambiguities, it is unclear how this figure is helpful, especially since the intended purpose of this work is to extract r_g and σ_g from SCIAMACHY...not r_e.

- **page 20, line 501:** "...utilizes a fixed number density profile..." What is this profile? Is it the OPC-based climatological profile, or the ECSTRA profile?

- **page 20, line 509:** "...can be by up to 51.1%." I have 2 points. First, should this be "...can be up to 51.1%"? Second, the error in N (including error bars) goes well beyond 51% (maybe even over 100%). Please clarify.

- **Section 7.3 and Figure 8:** It is unclear how the 1989–2002 time period is relevant to this study. This seems like wasted space and text. Figure 8 would be much more meaningful to the reader if the authors were to remove the 1989–2002 period (this would also allow them to use consistent scales) and create a single time series (i.e., 2002–2012). The figure could be further improved by breaking it into 2 figures (1 for extinction, 1 for r_e), with each figure containing sub-plots for different altitudes (e.g., panel (a) could be 30 km, panel (b) is 25 km, etc.). The value of doing this is it shows the reader the relevant information and provides the reader a much better understanding of the performance of this algorithm at multiple altitudes. Finally, showing this as a time series (instead of the aggregate profile statistics) allows the reader to appreciate the influence volcanic perturbations have on this method.

- **Figure 8:** It seems the first legend (titled "Symbols: Comparison of") is unnecessary. There is no "comparison" plotted in these panels, so it is unclear what the legend title means. The 2 legends could be consolidated to make the figure more easily interpreted.

- **page 23, lines 532–533:** "...due an increasing median radius with a simultaneously decreasing geometric standard deviation." First, should this be "...due to an..."? Second, this claim is conjecture and is not supported by the analysis. Since you are dealing with inferred values at this point I suspect that everything you see here, including the changes in r_g and σ_g, are symptoms as opposed to the root cause. Can the authors provide additional support for this claim or reword this sentence?

- **page 23, line 535:** What is the constant in the SCIAMACHY process? Undoubtedly it is N, but which N (UWY OPC or ECSTRA) is it?

- **Figure 9:** Again, this figure is an excellent example of why the authors should make the comparison with the UWY OPC products their first priority and give this analysis the most weight. This is also why the second priority should be comparing SCIAMACHY-derived extinction coefficients with SAGE/OSIRIS extinction coefficients should receive more weight and be second priority. This figure clearly demonstrates that the comparison is dominated by the assumptions in the SAGE estimates.

- **page 27, lines 648–652:** I have several points about this text.

  1. I saw nothing in the paper to indicate that "the median radius and geometric standard deviation are fully reliable *only* in the northern hemisphere." (emphasis mine). If this was in text then I sincerely apologize. Since the authors are limited to UWY OPC data (i.e., northern hemisphere) for validation I don't know how you could determine this. Please clarify.

2. If the southern hemisphere SCIAMACHY PSD parameters are "bad" then how do you justify inferring extinction coefficient and r_e from them? It would seem you are getting a more-or-less right answer for the wrong reasons. Please clarify.

3. The previous 2 points seem to be contradictory. Please clarify in the text.

4. The authors stated that the intent of this method is to derive PSD parameter (r_g and σ_g) from SCIAMACHY data...and do so globally. If the southern hemisphere r_g and σ_g are, per the authors' statement, unreliable then has the intent of this work failed? If so, the abstract must be updated to reflect the limited applicability of this method.

– **page 27, line 666:** The "link" must be updated.

– **Appendix A:** While this appendix is interesting, I fail to see how it makes a substantive contribution to the paper and should be removed (unless the authors can justify its inclusion, of course).