

With this document we would like to respond to the comment published by anonymous reviewer #2 on 20 September 2019. We thank the reviewer for this helpful comments, which were essentially all considered in the revised version of the manuscript.

Referee comment:

"Here is a paper describing a method to analyze monthly mean lidar color ratios to infer: profiles of the median radius of lognormal size distributions which model the measurements, the lidar ratio, and finally extinction from backscatter measurements. Yet no examples of the fundamental data are shown."

Our response:

We changed this and included now an example of the measured data. In section 2 we have included a new Figure 1 showing measured backscatter ratio profiles for 532 and 1064 nm with error bands together with the corresponding color ratio.

Referee comment:

"The paper moves quickly, and with no justification, from distributions of quantities with respect to color ratio, the measurement, to distributions of derived quantities with altitude. There is no explanation of how this transformation is made, yet the results from the rest of the paper hinge on this."

Our response:

The general concept of our method is described in section 3. We added text with further explanations to section 3.2 and hope this point is now clearer. Especially the origin of the altitude scale should now be better understandable.

Referee comment:

"The method to derive lidar ratio merits almost no explanation, yet it is a method I have never seen before, and raises questions as to why others have not used this method."

Our response:

We are sorry, but we do not fully understand this comment. Our paper is wholly dedicated to explain the method, so we would appreciate if you could be more specific on this point. If the particle size distribution is known (or has been estimated), then the determination of the lidar ratio is indeed that simple. However, in many cases, this information is not available.

Referee comment:

*"There are errors in some of the equations, and the origin of equation (11) is unclear. I detail these comments and questions in the review below.
10 Budget"*

Our response:

Thank you, somehow the spell checking has overseen this.

Referee comment:

"17-32 This nice description of the importance of stratospheric aerosol would benefit from some additional appropriate references."

Our response:

Now we included more references as suggested.

Referee comment:

"Eq (5) Shouldn't the scattering term k be $k(sca)_{Ray}$?"

Our response:

Thank you, this was obviously wrong.

Referee comment:

"116 For consistency with the ratio on $k(sca)$ on line 115 change to "depend on aerosol/air densities. I don't believe that there is any Mie scattering from air molecules."

Our response:

We changed it.

Referee comment:

"118 The scattering cross section, σ , should also be defined here, or above."

Our response:

The σ was not mentioned in the text. We have improved this and provided a reference.

Referee comment:

"130 Why is $n_0(\lambda)$ given only for 532 nm? What about 1064 nm?"

Our response:

Now the value for 1064 nm is also given.

Referee comment:

"142 Probably should add a more standard reference for Mie scattering (e.g. Born and Huffman or Dave), to which the Oxford scattering calculations surely have probably been compared. Perhaps this is even referenced in their code."

Our response:

In the source code of the software package used (Mie scattering routines (2018)) is a short reference to Bohren and Huffmann (1998). We have added this reference to the text.

Referee comment:

"177-178 Why do monthly mean data rule out a distribution width of 1.1? If the aerosol signal was from uniformly narrow distributions over the month then this is possible. Background stratospheric aerosol is thought to be from generally a rather well constrained and somewhat stable size distribution. The authors need a better argument to rule out a width of 1.1. "

Our response:

The work of Langenbach et al (2019) shows that even at high altitudes between 23 and 32 km and on short time scales of several hours the stratospheric background aerosol layer is highly dynamic. Therefore, the assumption of an aerosol population with a very narrow distribution width during a relatively long time period of one month is at least problematic. We included this in the text in section 3.3

Referee comment:

"The authors could look to the literature. It should be quite straight forward to find a reference to a typical background aerosol size distribution, for example from in situ measurements, which would not be consistent with a narrow size distribution with a median radius near 300 nm. This would clearly rule out a width of 1.1."

Our response:

That's right. In section 5.1 we compare our results with works of McLinden et al. (1999), Bourassa et al. (2008), Ugolnikov et al. (2018), Bingen et al. (2004) and Deshler (2008).

Under volcanically quiescent conditions most of this studies are in good overall agreement with our results. Only the work of Bingen et al. (2003, 2004a, 2004b) which analyses SAGE II data yields much larger radii of several hundred nm, even in the late 1990s, when the Pinatubo aerosol has already almost entirely disappeared. These discrepancies may in part be a consequence of different sensitivities to the aerosol particle population in combination with errors in the assumed PSD.

The available studies providing experimental values on sigma show sigmas exceeding a value of 1.1. A direct reference to other works which derived aerosol distribution widths with values above $S=1.4$ is given in section 3.3, lines 175-178 (McLinden et al., 1999; Bourassa et al., 2008; Ugolnikov et al., 2018).

Referee comment:

"180-181 characterise should be characterize."

Our response:

Thank you, now it is corrected.

Referee comment:

"Figure 2 Why is the ordinate altitude and not color ratio as Figure 1. The factors involved in this transformation of the ordinate are not clear. "

Our response:

The transformation of the ordinate axis from colour ratio to altitude is now described in the text in section 3.2.

Referee comment:

"In any case it seems that for branch one the radius variation range is limited to about 20 nm once the distribution width is assumed. This is quite restrictive."

Our response:

The relatively small radius range retrieved using this approach is not directly restricted by the approach itself, but by the measurements used as an example in Figure 2. For $S=1.3$, e.g., particle radii from over a range covering about 100 nm are in principle accessible (see blue line in Figure 2).

Referee comment:

"183-185 For the reader to understand this statement they would have to know how the color ratio varies with altitude. Isn't it enough for both branches 2 and 3 to point out that in these cases approximately half of the color ratio range would not be covered?"

Our response:

As stated above we have now included example data in section 2 with a new Figure which show measured backscatter ratio profiles for both wavelengths and the corresponding color ratio in dependence on the altitude. We hope this improves the understanding.

Referee comment:

"190 Change 'reduces' to 'restricts'. It's easy to misinterpret the sentence, as I did, if reduces is used, to mean the application to radii < 150 nm is limited."

Our response:

We have changed this.

Referee comment:

"196 ' . . . eqs (3, 4, and 5) can be solved for'"

Our response:

We have changed this.

Referee comment:

"Figures 2, 3, 4. There is some important information missing which is required to allow the reader to understand and tie Figures 2-4 to Figure 1. That information is the vertical distribution of the color ratio and for figures 3 and 4 the vertical distribution of the scattering ratio. After Figure 1 the ordinate shifts from color ratio to altitude with no explanation of how the two are related. In Figure 1 the color ratio range is 0.1-3.8. So how is this color ratio distributed by altitude? Once this is known then maybe it will be clear how the following figures are generated."

Our response:

Thank you for pointing out the importance of the transition from colour ratio to altitude. As suggested we have added text to section 3.2 and included a new Figure with example data showing measured backscatter ratio for both wavelengths and the corresponding color ratio. We hope this helps to better understand this point.

Referee comment:

"The two altitude dependent quantities in Eq (11) are P_{Mie} through its dependence on r_m and the scattering ratio, R . But for the lidar ratio the authors claim that only P_{Mie} is required and the altitude dependence is through r_m and hence the color ratio. All the vertical profiles, except the lidar ratio at 1064 nm, decrease rather significantly at 23 km, right where r_m decreases from 80 nm towards 60 nm for $s=1.3$. Is this all that's driving this vertical structure? And if that is the case is the lidar ratio at 532 nm really that much more sensitive to a change in radius from 80 to 60 nm than the lidar ratio at 1064 nm. There should be more discussion on these points."

Our response:

The Rayleigh extinction coefficient k_{ray}^{sca} is also altitude dependent and decreases exponentially with the atmospheric scale height. For longer wavelengths the phase function for backscattering exhibits a weaker dependence on radius because with growing wavelength the size parameter approaches the Rayleigh limit. So, here an dissimilar behaviour can be expected. We added this explanation to section 3.4.

Referee comment:

*"Eq (11) How is this Equation used? The term $k_{Ray} * P_{Ray}(\pi)$ in the numerator on the RHS of Eq (11) is calculated from the molecular density profiles from ERA-interim, and this same term, handled the same way, appears in the denominator of (R-1). So if Eq (11) is simplified it is a simple statement that $k_{Mie} = k_{Mie} * P_{Mie}(\pi) / P_{Mie}(\pi)$ or $k_{Mie} = k_{Mie}$. Isn't this a tautology? So how is Eq (11) something more than the measured backscatter divided by the phase function for backscatter, which can be calculated once the particle size is assumed and the wavelength known?"*

Our response:

There would be an tautology if only theoretical values would be used, but this is not the case here. The essential point is, that with R real measured data enters the equation which contains information about the aerosol.

Referee comment:

"205-213 There has been a lot of previous work devoted to determining the lidar ratio, but I have not seen the approach here. Is it really as simple as inverting the backscatter phase function, with the assumption that the backscatter is just the scattering coefficient times the phase function? Don't equations 12 and 13 imply that $P_{\text{Mie}}(0)$ is 1?"

Our response:

It is that simple if the particle size distribution is known (or has been estimated). However, in many cases, this information is not available. Concerning equations 12 and 13 it should be kept in mind that in equation 13 with β_{mie} measured data is used which allows for computation of the scattering coefficient.

Referee comment:

"Earlier, line 114, $k_{\text{Mie}}/\text{Ray}$ were defined as scattering coefficients, now here that term is being equated to extinction, the sum of scattering and absorption. "

Our response:

In section 3.1 we write that aerosol absorption has only a negligible effect and therefore we set it to zero. We added a note that in such cases the scattering and extinction coefficients are the same.

Referee comment:

"Also if the lidar ratio is just the inverse of $P_{\text{Mie}}(\pi, r_m, \lambda)$, why isn't that method used by, for example Jaeger et al. (1995), to calculate the lidar ratio from measured size distributions?"

Our response:

Jaeger et al. (1995) is not able to compute the lidar ratio based on his data because it consists of measurements at only one wavelength. Therefore, there is no information about particle size present and the lidar ratio is unknown. He uses a different approach with particle counter measurements with which he computes a lidar ratio and with this the extinction coefficient.

Referee comment:

"217 Or for certain wavelengths, 1064 nm?"

Our response:

You are right, it is a good approximation for 1064 nm over the whole altitude range but for 532 nm only for a constricted range between 15-23 km. We have adjusted the text accordingly.

Referee comment:

"230 They are identical because they both use a calculated cross section integrated over the same size distribution, or? It's hard to believe they would be identical if they were derived from measurements. But so far we haven't seen any measurements."

Our response:

The profiles are derived from real measured data as stated above concerning the meaning of equation 11, an example of measured data is now included as suggested. They are identical, essentially because measurements at both wavelengths are used to determine the colour ratio and finally the particle size.

Referee comment:

"242-245 Why are errors in temperature and pressure stated if they are not required because they cancel out? This is just a waste of the reader's time."

Our response:

The influence of temperature and pressure cancels out only for the radius computation. Both values enter the retrieval when the extinction coefficient is derived, they are implicitly included in the value of k_{ray} . We have added this information into the text.

Referee comment:

"Figure 7 How is this figure different from an expanded version of Figure 2? It is basically the same figure. What are all the error contributions included? It was already stated that temperature and pressure cancel each other out because color ratios are used, so these are not included. It is not explained how a difference in the refractive index affects the retrieved radius."

Our response:

Figure 2 shows radius profiles obtained for different assumed distribution widths whereas Figure 7 shows the influence of parameter variation on a radius profile when a distribution width is already chosen, in our case it is $S=1.3$.

Figure 7 is embedded in section 4, which deals with the error estimation. The assumptions made for the estimation of errors are clearly stated in this section in our opinion and reference to Figure 7 is made. We apologize, if this is not fully clear. The content of the error contribution is specified in lines 249-251: "If the single error contributions are simply added to a total error separately for the two assumed deviations of the distribution width an absolute error range can be assigned to the retrieved radius profile as shown in Figure 7. " This also includes the influence of the refractive index.

Referee comment:

“Figure 8 What kind of a scale is on the abscissa. The minor tick marks cannot be used to state what the precision is exactly, but it appears to be less than 3% for 532 nm. This figure then indicates that a difference in aerosol radius between the min and max distribution width, which is about a factor of two in radius, lead to almost no effect on extinction? This is a surprising result, suggesting that the determination of the median radius is not that critical. There is a much larger effect at 1064 nm but it seems a bit odd that the effect is not symmetric.”

Our response:

Thank you for pointing out the obviously distorted Figure, we have corrected it. In section 4 we added a short explanation about the relative error used as abscissa scale in Figure 9 (since we added one figure in the revised manuscript this corresponds to Figure 8 of your comment). The influence of the distribution width has of course an influence but it is not that big. The differences between the two wavelengths are caused by a wavelength dependant sensitivity. The scattering cross section and the extinction coefficient are not linear functions of the radius. Therefore, a symmetric behaviour can not be expected here.

Referee comment:

“254-258 According to Eq (11) the only size distribution information used is the median radius, r_m , which appears in the phase function. Thus I don't follow this argument that the uncertainties in distribution width are compensated for by the opposite uncertainty in median radius. The uncertainty in distribution width leads to the radius uncertainty which is then used in calculating extinction, according to Eq (11).”

Our response:

If the assumed distribution width is too large, i.e. larger than its real value, then the retrieved median radius will be low biased and vice versa. These effects partly compensate each other for the determination of the aerosol extinction coefficients. For this reason, the extinction coefficient retrievals react less sensitively to an erroneous distribution width compared to the median radius retrievals.

Referee comment:

“Figure 9 What is the point of this figure. It is just a repeat of Figure 3 with the per cent uncertainties, already shown in Figure 8, added to absolute extinction, and it is much less helpful than Figure 8 in assessing this uncertainty.”

Our response:

This observation is right. We prefer to separate the retrieved profiles (without error ranges) from the error estimation. In our view this approach improves the understanding.

Referee comment:

"Eq (16) $c(z)$ is not defined. Shouldn't there be a ratio of wavelengths within the exponential term of the desired wavelength over the reference wavelength. "

Our response:

Thank you for pointing this out, there was no reference on $c(z)$ in the text, we have changed this.

Referee comment:

"Figure 11 Which lidar extinction profile is used in the Angstrom conversion to the satellite wavelengths. "

Our response:

This information is now included.

Literature used:

Craig F. Bohren and Donald R. Huffman, "Absorption and Scattering of Light by Small Particles", Wiley Science Paperback Series, Wiley Professional Paperback Edition 1998

Jäger, H., T. Deshler, and D. J. Hofmann, Midlatitude lidar backscatter conversions based on balloonborne aerosol measurements, *Geophys. Res. Lett.*, 22, 1729-1732, 1995.

McLinden, C. A., McConnell, J. C., McElroy, C. T., and Grifoen, E.: Observations of stratospheric aerosol using CPFM polarized limb radiances, *J. Atmos. Sci.*, 56, 233-240, 1999

Mie scattering routines, University of Oxford, Department of Physics, <http://eodg.atm.ox.ac.uk/MIE/index.html>, last access: 31 December 2018

Bourassa, A. E., Degenstein, D. A., and Llewellyn, E. J.: Retrieval of stratospheric aerosol size information from OSIRIS limb scattered sunlight spectra, *Atmos. Chem. Phys.*, 8, 6375-6380, 2008

Ugolnikov, O. S. and Maslov, I. A.: Investigations of the Background Stratospheric Aerosol Using Multi-color Wide-Angle Measurements of the Twilight Glow Background, *Cosmic Research*, 56, 2, 85-93, doi:10.1134/S0010952518020119, 2018, Original Russian Text, 2018, published in *Kosmicheskie Issledovaniya*, 2018, 56, 2, 95-102, 2018

