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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Variability of Lidar-Derived Particle Properties Over West Africa Due to Changes in Absorption: Towards an Understanding" *by* Igor Veselovskii et al.

Anonymous Referee #2

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The manuscript contains a well-elaborated scientific study about the link between lidar-derived optical properties of dust and dust-smoke mixtures in western Africa and the basic aerosol microphysical and dust chemical composition. The manuscript is well written and appropriate for ACP. The key part deals with the relationship between measured extinction-to-backscatter ratios at two wavelengths (and corresponding wavelength dependence) and the imaginary part of the refractive index. However, conclusions are drawn (from observations and simulations) which are not at all trustworthy (to this reviewer) as long as a spheroidal shape model for the unknown irregular shape of mineral dust particles must be assumed in the modelling approaches and the influence of this assumption on the final results remains unknown, at least is not discussed and

quantified. Another critical point is that a reader may get the impression that the findings hold for mineral dust in general, and not only and specifically for western Saharan dust and dust/smoke mixtures. However, the literature contains numerous papers that are in contradiction with the findings presented here and in Veselovskii et al. (2016). For example, such large values of up to 1.5 for the ratio of the 355 to 532nmlidar ratios as reported here together with particle depolarization ratio of larger than 30 % and thus indicating pure dust conditions has never been reported in numerous others papers in the literature. Larger backscatter in the green than in the blue is reported sometimes but only as an exception. Furthermore, all these inversion procedures(lidar, photometer) assume particles with radius < 15 μm, but the lidar field site (Senegal) is almost in a desert source region so that the probability of the occurrence of giant particles is never zero. This can be another 'efficient' error source.

Detailed comments and suggestions:

P2 L65-67: One should even include the third part of the entire SAMUM-SALTRACE series and the related papers of Gross et al 2015, Rittmeister et al., 2017, and Haarig et al. 2017.

P2 L69-77: I am confused that the backscatter coefficient (a pure scattering parameter) is very sensitive to the imaginary part of the refractive index (describing the absorption efficieny. . .), but the extinction coefficient (absorption plus scattering coefficient) is insensitive to the imaginary part (to say it again. . .. describing the absorption features of aerosol particles). Can you please provide more background information on this 'apparent contradiction' that a scattering process is influenced by the imaginary part, but absorption features not?

P2 L126: The paper emphasis the usefulness of the lidar ratio (S) wavelength dependence. Why not writing down the relationship between all the different Angstrom exponents, AE(ext), AE(bsc), AE(S), that is AE(S) = AE(ext) – AE(bsc) as introduced by Ansmann et al., JGR, 107, 10.1029/2001JD001109, 2002. In this way, it is much more easy to follow the discussion later on, when AE(ext) is frequently around zero, because then we have simply: AE(S) = -AE(bsc).

P4 L120 – P5 L138, A lot of equations.... Are all these quantities used later on?

P7 L206 – 209: Here again the confusing relationship between backscatter coefficient and imaginary part ..... I am still confused by the high negative AE(bsc) values. Unfortunately, extinction profiles (with rather reasonable wavelength dependence) are frequently shown, but the profiles of the backscatter coefficients for these high negative AE values are never shown. Such observations are rare in the literature and may be related to the region of western Africa and proximity to dust source regions (and the omnipresence of coarse and giant dust particles).

The authors argue, it has to do with the chemical composition of the dust particles, and then with the imaginary part? But my question is continuously: If we use an alternative shape model what would then be the result...? Gasteiger et al., and modelling groups in Finland introduced very different shape models.... And found very different results in terms of dust optical properties. And if we ignore all the giant dust particles in the complex data analysis, how large can be the damage?

P7 l 210-220: The lidar ratios for dust (355 vs 532 nm) are so different (60-70sr vs 45-60sr), although the depolarization ratio shows (almost) pure dust conditions, this is a very surprizing result and in contradiction with the literature (Tesche et al, 2011, Gross et al., 2011). Any explanation for this? Maybe... it has to do again with missing coarse and giant, irregularly shaped dust particles? Because AE(S) = - AE(bsc), why is backscatter Angstroem exponent so pronounced, why is that so negative? Is everything ok with the backscatter retrieval (calibration in the reference height)? My explanation would be: smoke is responsible for the strong lidar ratio wavelength dependence, but then the depolarization ratio should indicate that.

P9 L266-267: There are also smoke lidar ratios from the SAMUM Cabo Verde campaign (Tesche, Tellus, 2011, second paper).

P11 L325-326: The lidar ratio depends on complex refractive index and size distribution, BUT ALSO ON SHAPE of the particles. Again the conclusion from my side: Are all the simulations and retrieval products trustworthy and reflect the reality when a spheroidal shape model is needed and used? Can we trust the main findings and conclusions? And again: What about the impact of missing giant, irregularly shaped particles? They are there over-near source regions!

P11 L355-356: observed low values of AE(bsc) cannot be produced without accounting for a spectral dependence of the imaginary part..... Yes, for the case of the assumed spheroidal shape model and size distribution up to about 10μm radius.

AERONET is used for comparison. But here (photometer) also a spheroidal shape model is needed for dust particles. So, no independence between lidar and photometer products.

P12 L368-369: ... the authors write...again...: .... the observed S355/S532 ratio and AE(bsc) can be explained by the spectral dependence of the imaginary part of CRI. My answer again: Yes, for spheroidal particles. So again... the reader is left with the question, and what is now the true impact of the imaginary part..... for irregularly shaped particles for which we do not have a model?

P12 L371: S355/S532 is 1.5 !!! and the depol ratio >0.3 indicates pure dust! Such a result I have never seen in the literature, and thus must have to do with the field site (and probably to the omnipresence of very large dust particles). I have no other explanation. Disregarding, whether my comment makes sense or not, the findings in this paperseem to be specific for Senegal, or maybe regions in the vicinity of dust source regions. That should be clearly written.

P13 L418: The authors write: The relative humidity varied from episode to episode. .... To my knowledge, different RH conditions usually show different air masses and different aerosol properties from different sources. At least it seems to be a difficult task to correlate RH with found aerosol properties, and to draw trustworthy conclusions on the hygroscopic (water uptake) properties.

P14, L447-448 Ok, this is said now but that comes very late.

P14 : When modelling lidar ratios for smoke, do you have proper smoke particle size distributions as input?

P15 L476-478: Also the size distribution changes a bit when the relative humidity increases. Is that considered in the simulations?

P17 after L545-550: At, at the end of the summary and conclusion section, one could mentioned as an outlook, that such studies as presented here should be repeated at many different places around the world, e.g. in the Middle East, Central and East Asia, Australia, and North Amerika. . .. in order to improve our knowledge on real-world aerosol optical properties needed in optical modelling and climate relevant atmospheric modelling.

My final summarizing comment: Disregarding the criticisms made here, my overall impression is: The discussion and sensitivity studies are nice and very helpful to interpret sophisticated multiple wavelength polarization Raman or HSRL lidar observations of complex dust and dust-smoke mixtures. The authors are experts and contribute with an important contribution to the field of dust atmospheric studies and interpretation in terms of dust size distribution, shape properties, and chemical composition. However, the discussion should be more sensitive to all the shortcomings (unknown shape, ignored giant particles). The more clearly the short comings are presented the more exciting the story and the probability to trigger further studies. The field is interesting, and the more questions are left at the end, the more interesting is this research field for the next generation of researches. To define the right questions is often better then to present some (questionable) answers. . .

Figures:

Fig.1: Please skip the symbols, just show different colored thin lines. It is hard to see

anything, except the ups and downs in the curves.

Fig. 5a: The lidar ratios of 70 sr at 355 nm and 50 sr at 532 nm clearly shows the strong impact of smoke to my opinion. The extinction profiles and the depolarization ratio > 0.3 indicate pure dust! I am very much confused.

Fig.5b: The same here. . .. And the modelling results are at all even more extreme . . . and thus totally unrealistic to my opinion . . .. with the spheroidal model as input. . .almost 40 sr at 532 nm and 70 sr at 355 nm for pure dust conditions! Has this something to do with any dust reality? And AE(S) of 1.2 or and AE(Bsc) of -1.2 from the model?

Fig 6: Here one could discuss AE(S)=AE(ext)-AE(bsc) when showing AE(ext) and AE(bsc). AE(ext) atleast shows what we know from AERONET and the many dust lidar observations. But the backscatter wavelength dependence remains unique and seems to be quite special for this part of the world, whatever the reason may be.

Fig. 8: Again, nicely measured 355 and 532 nm extinction profiles (a), and (surprisingly) ‘well-known’ lidar ratios in the upper part of the dust layer, and very trustworthy depol ratios (b) and trustworthy AE(ext) profile (b), and even AE(bsc) and thus AE(S) in the upper part of the layer, the same for (c) and (d). This is what we know from the literature and all the SAMUM and SALTRACE findings for Saharan dust and aged outflow dust transported to the west.

And then again this huge contrast: AGAIN these strange model results. Lidar ratios up to almost 70sr at 355 nm and down to 40sr at 532 nm, and at the same time, these very reasonable lidar measurements. The authors seem to provide me (voluntary) with the wappons I need to blame the modelling effort and point on the used shape model as the main source for these errors. What shall we believe at the end, from all the conclusions the author derive, when we see this? Fig.8 is an excellent figure to corroborate my opinion: Ido not believe in any of the result!

Fig 10: The depolarization ratio shows no change in the dust conditions over the plotted one month period and is so high (PURE DUST), but the lidar ratio is sensitively changing. . .., and again probably solely dependent on the backscatter coefficient which is a strong function of refractive index and particle shape! So, what to do? Ok there is a correlation with the refractive index from AERONET. But AERONET also assumes a spheroidal model to produce dust-related results. So, what can we believe at the end.

Fig 12: Similar curves , good! But what does it help?

Fig 13: Size distribution from AERONET and from lidar (at what height?). Differences are visible but both size distributions stop at 10 microns. What about bigger particles when measuring very close to the source region. . . Is it possible that the used size distribution in all model runs underestimate the impact of the giant particles?

Fig. 14: I do not believe the Imag curve (stars) as long as we have another errors source (the spheroidal shape model). AE(Bsc) = - AE(S) can be used her in the discussion because AE(ext) is almost zero.

Fig 17-Fig 19: The RH study and discussion makes the paper quite long, and all the efforts are again quite speculative. . . Fig 19 is not convincing.

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