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Interactive comment

Interactive comment on "Is the near-spherical shape the "new black" for smoke?" *by* Anna Gialitaki et al.

Anonymous Referee #3

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General comments:

The more frequent occurrence of pyroCb smoke plumes lofted into the stratosphere is a rich target for analysis into a kind of aerosol about which we know relatively little. I'm particularly interested to see apparently successful modeling of three-wavelength values of both linear particle depolarization ratio and lidar ratio from lidar measurements of one such plume. This is a new result and potentially quite useful, since previous particle modeling studies did not have access to such a complete lidar observation and also because they, in general, resorted to much more complicated models than what the current authors have found to be useful. So, for this reason, primarily, I would like to see this paper published.

On the other hand, many aspects of the manuscript seem rather weak and unconvincing, so I believe major revisions are appropriate. First, the discussion and elimination of other models is not convincing. It may not be strictly necessary to show that other models perform worse if the near-spherical model is able to reproduce all the available measurements (because a simple model that fits all the observations has benefits over a more complicated model just by virtue of being simpler), but since an attempt is made to do so, it should be done thoroughly and correctly. Second, an unsubstantiated claim is made that this could improve AERONET retrievals. The idea of the new model improving or complementing AERONET is potentially quite appealing, and if done right this could be a major focus of the paper, so again it should be addressed thoroughly, not haphazardly. Finally, the speculation about the role of sulfuric acid for explaining the depolarization measurements seems a bit far-fetched and very difficult to validate while other potential explanations have not been adequately discussed. In my opinion, this is the weakest part of the manuscript and the best solution may be to simply not offer explanations for the shape at all, but rather to present this work as an advancement in the modeling of the optical properties alone. Otherwise, if the authors want to keep this, then a better, more thorough discussion of alternate theories and ways to distinguish between theories is needed.

Specific comments:

page 3, line 10, I think rather than "an explanation that could justify" the values, you're more fundamentally searching for "a model that can reproduce" the observations. This is a more precise statement of what this calculation is able to do and valuable enough at this stage.

page 3, line 10. "non-typical spectral dependence". What do you mean non-typical? Compared to what? It's my understanding that there are only a very small number of observations of three wavelengths of smoke particle linear depolarization ratio and not much discussion of two-wavelength observations. So, how do we know what spectral dependence is typical?

Besides just listing three papers at page 3, line 14, the introduction should discuss how this study is similar and different to the modeling studies in those and other papers, including those that use other modelled particle shapes other than near-spherical. Besides the 3 references listed, consider Kahnert (2017), Liu and Mishchenko (2018), Kanngießer and Kahnert (2018), Ceolato et al. (2018), and Luo et al. (2019) (some of these are mentioned later in the manuscript but not the introduction). Also Mishchenko et al. (2016) used multiple particle shapes, not just the near-spherical.

Kahnert, M.: Optical properties of black carbon aerosols encapsulated in a shell of sulfate: comparison of the closed cell model with a coated aggregate model, Optics Express, 25, 24579-24593, 10.1364/OE.25.024579, 2017.

Liu, L., and Mishchenko, M.: Scattering and Radiative Properties of Morphologically Complex Carbonaceous Aerosols: A Systematic Modeling Study, Remote Sensing, 10, 1634, 2018.

Kanngießer, F., and Kahnert, M.: Calculation of optical properties of light-absorbing carbon with weakly absorbing coating: A model with tunable transition from film-coating to spherical-shell coating, Journal of Quantitative Spectroscopy and Radiative Transfer, 216, 17-36, https://doi.org/10.1016/j.jqsrt.2018.05.014, 2018.

Ceolato, R., Gaudfrin, F., Pujol, O., Riviere, N., Berg, M. J., and Sorensen, C. M.: Lidar cross-sections of soot fractal aggregates: Assessment of equivalent-sphere models, Journal of Quantitative Spectroscopy and Radiative Transfer, 212, 39-44, https://doi.org/10.1016/j.jqsrt.2017.12.004, 2018.

Luo, J., Zhang, Q., Luo, J., Liu, J., Huo, Y., and Zhang, Y.: Optical Modeling of Black Carbon With Different Coating Materials: The Effect of Coating Configurations, Journal of Geophysical Research: Atmospheres, 124, 13230-13253, doi:10.1029/2019JD031701, 2019.

How are the ranges arrived at that are shown in Table 1, and also specifically the fixed values for the distribution widths?

Can you eliminate water or ice cloud as an explanation for the measurements in CALIOP?

Figure 7 had a panel of water vapor mixing ratio but no explanation of this measurement or description of what impact this figure has on the analysis of this case.

For Figure 8 and Table 4 and others, please define the meaning of the error bars. Is this a true calculated uncertainty including both random and systematic error, or is this the standard deviation of available measurements, or something else? If it's standard deviation, how well do you think this captures the actual uncertainty of the lidar measurements at each wavelength? I ask because you mentioned that the 1064 nm extinction measurement is more challenging to make and we also know from literature that particle depolarization ratio in particular can be subject to significant systematic error in some circumstances (e.g. Burton et al. 2015, Freudenthaler 2016, Belegonte et al. 2018).

Freudenthaler, V.: About the effects of polarising optics on lidar signals and the ∆90Âăcalibration, Atmos. Meas. Tech., 9, 4181-4255, 10.5194/amt-9-4181-2016, 2016.

Belegante, L., Bravo-Aranda, J. A., Freudenthaler, V., Nicolae, D., Nemuc, A., Ene, D., Alados-Arboledas, L., Amodeo, A., Pappalardo, G., D'Amico, G., Amato, F., Engelmann, R., Baars, H., Wandinger, U., Papayannis, A., Kokkalis, P., and Pereira, S. N.: Experimental techniques for the calibration of lidar depolarization channels in EARLINET, Atmos. Meas. Tech., 11, 1119-1141, 10.5194/amt-11-1119-2018, 2018.

page 7, line 20-21. Lidar ratio increase from UV-visible suggests that it also increases from visible to near-IR. This should be deleted. There's nothing in Muller et al. (2007) that addresses the lidar ratio values in the near-IR one way or the other.

page 7, line 22 "far from typical". I urge you to reword and avoid "typical". Muller et al. 2007 was a quite valuable paper, but the cases in it are somewhat limited, and it is now more than a decade old. Something that does not conform to Muller et al. 2007 is not necessarily "non-typical". We are still seeing new and different observations, by now including many of depolarizing smoke. I would say that this manuscript and other recent papers make a more convincing case that there is no "typical" for smoke or else that we do not have sufficient observations yet to know what is "typical", rather than that the depolarization ratios in this case are non-typical.

The notation in Equation 8 is confusing and doesn't really make sense. Please define what are i and n? Is this a summation over the three wavelengths? In that case, you have two subscripts (i and lambda) that mean the same thing? Or maybe i is binary and means lidar ratio and depolarization ratio, but in that case, you do not show how the different wavelength measurements are combined.

Equation 8 furthermore should arguably have the measurement uncertainty rather than the measurement itself in the denominator. This would be a more meaningful cost function considering you intend to compare the result to the measurement uncertainty in Eqn 9. Doing this could have a significant impact on your results, specifically the result for the Chebyshev model that is shown in Figure 12. The only simulated point that doesn't fit the measurements is the 532 nm depolarization which has a very small reported uncertainty and therefore not much tolerance. But if the cost function reflected the error bars as well, you might find there is a solution that fits that point at the expense of slightly larger discrepancy in another quantity where the uncertainty tolerance is much larger (e.g. lidar ratios).

I'm also curious how many solutions fit the criteria in Eqn 9 (or revised criteria) besides the minimum. Looking at this would give some insight into the uncertainty of your modeled results and the degree to which the set of measurements is sensitive to the complete set of free parameters in your model.

In the figure 10 comparison with AERONET, the use of a generic biomass burning solution instead of a solution for the same smoke plume seems like a needless shortcut that undercuts your ability to draw conclusions from it. I realize there were no precisely coincident AERONET measurements, but a previous paper studying the same event that you already reference by a coauthor (Haarig et al. 2019) shows an AERONET retrieval that is at least of the same smoke plume, and also apparently better agreement, so clearly it's possible to get better fidelity than the unrelated generic case given by Dubovik et al. (2002).

Furthermore, comparing a fit to a monomodal size distribution to the fit from a bimodal size distribution and then noting that the modes don't line up is not particularly useful, and it's not obviously tied to to the presence or absence of near-spherical particles per se. If you must compare a monomodal fit to a bimodal fit, then at least calculate the effective radius and variance (quantities that are more comparable from different distribution types) from each of them and compare that instead.

On a related note, could there be a coarse mode that your model is ignoring that might explain some of the features of your observations? Have you tried to eliminate the possibility of an optically significant coarse mode?

Fractal aggregates: Fractal aggregates require a lot of parameters to describe them, and some of them you held fixed instead of varying. If you cannot explore the full parameter space, how do you know that fractal aggregates can't fit the observations?

At line 25, you then point out that previous authors (not just Ishimoto et al. 2019, but see also Kahnert 2018, Kanngeisser and Kahnert 2018, Luo et al. 2019 and Ceolato et al. 2018, as noted above) have already established that bare aggregates are not able to reproduce measurements as well as coated aggregates, so I don't see a lot of value in running a model type that has already been shown not to work without also running a related model type that has been used with some success in the past (granted with less complete measurements in the past; indeed, the new measurements are the real strength of this contribution and where you have the opportunity to go beyond prior work).

Page 7, line 10. This brief sentence about extending AERONET to include near-spherical particles is an interesting idea, but unsupported. To address this properly, please consider at least these 3 points. First, as stated above, there would have to be a fair comparison between AERONET results and your results for similar cases. As part of this, there would have to be an assessment of not just the size distribution, but also a reconstruction of the lidar measurements using the AERONET solution. Can you show that the AERONET retrieval fails to reproduce the lidar ratio and linear particle depolarization ratio adequately? Conversely, how does your near-spherical model do in reproducing AERONET radiances at all AERONET wavelengths? Whatever the answer to each of these questions, there's something to be learned. If the near-spherical model does a better job of reproducing lidar measurements and is also better at modeling AERONET measurements, then it could genuinely be an improvement for AERONET. If it doesn't improve the AERONET fits but improves the lidar fits, it might be less useful for AERONET alone (at least it would suggest that it might be hard for AERONET to operationally use the model if there is not sufficient measurement information content to distinguish near-spherical from spherical particle shapes), but might still be potentially of significant value for combined AERONET-lidar retrievals (e.g. constrained backscatter lidar retrievals). Even if the near-spherical model does a worse job at modeling AERONET measurements but a better job at modeling lidar measurements, it at least points us to the need for further modeling studies to find a single model that can unify both types of measurements.

page 7, line 16. See also Kablick et al. 2018 for another case discussing ice.

Kablick III, G., Fromm, M., Miller, S., Partain, P., Peterson, D., Lee, S., Zhang, Y., Lambert, A., and Li, Z.: The Great Slave Lake PyroCb of 5 August 2014: Observations, Simulations, Comparisons With Regular Convection, and Impact on UTLS Water Vapor, Journal of Geophysical Research: Atmospheres, 123, 12,332-312,352, 10.1029/2018jd028965, 2018.

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page 7, line 17. In 2015, Burton et al. could not have extensively discussed studies that were published 3 or 4 years later. Specifically there's no discussion in Burton et al. 2015 about the ice hypothesis. It would be better for this manuscript's authors to take the opportunity to address these newer theories more completely here.

page 7, line 17-18. I'm not following the statement "soil lifting ...could explain the ....observations presented in this study". Do you mean to eliminate this possibility or support this possibility?

page 7, line 18. The Angstrom exponent is indeed confusing, primarily because we don't know what wavelengths you're referring to either in the measurements or in the comparison dataset that causes you to say this is "low". It's not uncommon for smoke measurements to have significant curvature in the spectral AOD (or extinction) (see Eck et al. 1999) and in fact Haarig et al. 2018 show a significant difference between the 355-532 nm and 532-1064 nm Angstrom exponents for their analysis of this smoke plume. Taking this into account, do you still believe the Angstrom exponent for this case indicates coarse mode particles? Please add a more complete discussion.

Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, Journal of Geophysical Research: Atmospheres, 104, 31333-31349, 10.1029/1999JD900923, 1999.

page 7, line 21 "surface roughness alterations"? I don't follow where this idea comes in the paper. Is this what the Chebyshev particle shape is meant to represent? There are other model representations of surface roughness (e.g. Liu et al. 2013, Kemppinen et al. 2015) so this label is probably too non-specific (vague) in this context and should be made more precise.

Liu, C., Lee Panetta, R., and Yang, P.: The effects of surface roughness on the scattering properties of hexagonal columns with sizes from the Rayleigh to the geometric optics regimes, Journal of Quantitative Spectroscopy and Radiative Transfer, 129, 169-

185, https://doi.org/10.1016/j.jqsrt.2013.06.011, 2013.

Kemppinen, O., Nousiainen, T., and Lindqvist, H.: The impact of surface roughness on scattering by realistically shaped wavelength-scale dust particles, Journal of Quantitative Spectroscopy and Radiative Transfer, 150, 55-67, https://doi.org/10.1016/j.jqsrt.2014.05.024, 2015.

page 10, line 2, what do you mean by "superposition" in this context? The papers above that address coatings (e.g. Kahnert 2017) suggest that the mixing of soot aggregates and coatings has a rather complicated impact on the optical properties, which I believe you would have to model rather explicitly and not just average, but perhaps the reference you're quoting suggests otherwise. Please describe more explicitly.

page 10, line 8 "two criteria need to be fulfilled". This seems like a spuriously specific conclusion. The lab study showed that the optical properties change in the presence of H2SO4, but does it follow that it must be H2SO4 and not other coating substances that are more likely to be found in a smoke plume? Or that there are not other mechanisms unrelated to this lab result that might explain the depolarization?

page 10, lines 15-20, the two hypotheses that the volcanic plume is required to explain the observations and that the smoke plume and volcanic plume intersected each seem like a stretch. There been several pyroCb plumes displaying elevated depolarization ratios in CALIPSO data, as well as the high tropospheric case described by Burton et al. 2015. Do you propose that they all intersected with volcanic plumes? If so, I guess it would be straightforward to check and you should do so. Also, the volcanic plume itself should be traceable with CALIPSO at least for part of its lifetime. Investigating this could help determine more clearly if the two plumes could have intersected at the same altitude and location.

Later on page 10, lines 29-30, is the suggestion that lower RH with aging of the plume decreased the depolarization ratio of the plume. This seems somewhat counter-intuitive, since with coated particles in the troposphere, at least, we would not expect that losing a coating could make particles more spherical. Does the lab study support the idea that losing the coating makes the particles more spherical? Does the lab study, or your proposed follow-on, take the colder stratospheric temperatures into account?

Alternately, is there an indication that the size of the particles are changing (from losing the coating) for instance a change in the Angstrom exponents?

What results does your near-spherical model retrieval produce when applied to this later less-depolarized observation? Can you reproduce the reduction in linear particle depolarization ratio and also reproduce lidar ratio and extinction spectral dependence? If so, are the particle sizes smaller, or what other differences do you observe compared to the solution for the earlier time frame?

page 10, line 25, "ongoing research investigates whether this concurrence, or the the large amounts of SO2 “internally” released by the fire, or even the stratospheric sulfate background already pre-existing at the stratospheric smoke injection height, is the reason behind the unique large values of stratospheric PLDR." While you are obviously not required to describe your future research here in any detail, this teaser is so broad that it invites skepticism. I’d be interested to see it replaced with a more concrete description of what falsifiable question(s) you will attempt to answer and whether your proposed research is a lab study, based on in situ measurements, or a theoretical study.

Minor comments:

While informal titles can catch people’s attention, I wonder if this one is really a good idea? "The new black" means "fashionable" or "popular", which is probably not quite what you’re hoping for from your new smoke particle model. In general I appreciate funny titles but if it is a pun, I’m not getting it, since I don’t see what scientific meaning "black" has in this title. Also, the phrase "the new black" is itself something of a fad which may fade quickly, leaving readers 5 or 10 years from now completely confused about what the title means. But of course it is up to the author; this isn’t a comment that needs a response, just a perspective on how one reader sees it, in case you find this helpful.

page 2, line 26. The sentence starting "implication for enrichment of smoke plumes with dust particles" should probably be deleted. Later on, you mention several possible explanations from previous literature, but here you only mention one without discussion that you later dismiss. Better to delete it here and hold the discussion until you are ready for it in the later section.

page 7, line 21 "not currently supported by other observation evidence found in the literature". Probably should be reworded. It seems like you're saying there is other evidence found in the literature that doesn't support your finding, but my understanding is that there is no contradictory observational evidence because lidar ratio at 1064 nm for this kind of observation has never been reported before! Please make this more clear. Having unique measurements is a real strength! No need to muddy the picture with imagined controversy.

page 7, line 11, probably delete the brief mention of pollen, which has not been addressed at all in this paper. We have no way of knowing whether the near-spherical model has any success in modeling pollen.

page 9, line 12, "results in near-spherical shapes" should be reworded. Logically, the finding that the near-spherical particle model reproduces a set of measurements better than a few other models could still be merely a very useful approximation. It does not necessarily mean the particles are literally shaped like Figure 2.

page 9, line 12, "previous studies" should be "some previous studies" (i.e. but not all, see for example Murayama et al. 2004, Burton et al. 2015 who specifically dispute it for certain other cases) Murayama, T., Müller, D., Wada, K., Shimizu, A., Sekiguchi, M., and Tsukamoto, T.: Characterization of Asian dust and Siberian smoke with multiwavelength Raman lidar over Tokyo, Japan in spring 2003, Geophys Res Lett, 31, 10.1029/2004gl021105, 2004.)

page 9, line 14, Sugiomoto et al. what year? (typo)

page 9, line 23, "advocate dissuasive towards" should be changed to (e.g.) "argues against"

page 9, line 26, "while up to now the LR values are not reproduced either". Again, I feel like this should be reworded, because it's making a confusing (or perhaps just incomplete) point when a much stronger one is indicated. Most previous modeling papers did not have the opportunity to reproduce three-wavelength lidar ratios because these observations have only just recently been published. The strength of the current manuscript is these new and unique measurements.

page 9 line 30. I think 15 micrometer monomers must be a typo.

page 10 line 4. "Thought" should be "Although"'

Table 4 caption. Please specify the instrument that made these measurements.

Figure 6. The red dashed lines appear to be in the wrong place.

It would be good to include a figure explaining what Chebyshev particles look like.

Figure 7. What do the white pixels near the top of the layer signify, in both the linear depolarization ratio and the water vapor mixing ratio?

Figure 7. What do the black down-arrows on the latitude and longitude axes represent?

Figure 7. Please indicate in 7c the portions of the track that are represented in 7a and 7b.

Figure 14. Please mark the location of the smoke plume and consider plotting this on an altitude (rather than pressure) scale and with altitude range more comparable to the ranges shown in Figures 6 and 7.

Please also consider putting an indicator of distance scale on Figures 6, 7, and 14.

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