Review of "Tropospheric and stratospheric wildfire smoke profiling with lidar: Mass, surface area, CCN and INP retrieval", manuscript version 3, by Ansmann et al., 2021

This latest draft represents a prodigious rewrite of the authors' original manuscript. In it they address all of the major criticisms I noted in my initial review. By reorganizing the structure of the paper, they have also expanded their exposition in several places where I thought additional details would be particularly helpful. In my opinion, the revised manuscript can and should be published. Nevertheless, below I offer a few minor comments, questions, and suggestions. While addressing these is entirely optional, I believe that doing so might help further clarify a few of the points made in the paper.

Page 8, equations 1 through 4 : the symbol β has not yet been associated with backscatter. Ideally, both β and L would be defined in the paragraph immediately following the first use of the symbols.

Page 8 : the authors recommend default values for smoke lidar ratios "if there is no possibility to obtain actual lidar ratio information from Raman lidar or High Spectral Resolution Lidar (HSRL) observations". Do they consider the lidar ratios obtained via constrained solutions of elastic backscatter lidar measurements (e.g., Prata et al., 2017) to be of insufficient accuracy? A comment on this would be helpful for potential users of the authors' technique.

Page 9, line 23 through page 12, line 7 : I did not revisit this material, as I assumed it was essentially unchanged from the first version of the paper.

Page 12, lines 8–19 : I expect readers attempting to implement the authors' technique will appreciate the addition of these caveats.

Pages 12–14, Section 4.1 : I'm especially pleased to see the inclusion of the AERONET data from the sites in Alta Floresta, Mongu, Mukdahan, and Singapore.

Page 16, line 23 : Since the authors are presenting an "AERONET-based correlation analysis", I'm more than a little surprised not to see correlation coefficients reported for the relationships shown in figures 6–10. Having uncertainty estimates for the coefficients derived from the individual fits included either in the caption or as part of the figure annotations would also be a very sweet addition. (According to page 18, line 7, some of these numbers are given in Table 3.)

Page 17, line 8 : regarding figure 4, the basis for asserting the "smallest particles found at Alta Floresta indicate rather fresh smoke, probably just a few hours after emission" is not immediately apparent. Best I can tell, this is the only example that is not associated with a specific fire, and hence the only example in which the age of the smoke is directly inferred from the size distribution. Since Figure 4 is shown to illustrate "the shift of the size distribution towards larger particles with age of the observed smoke", it seems that have an actual fire (and hence an excellent estimate of age) to associate with the Alta Floresta smoke is reasonably important.

Page 17, line 17 : the caption for Figure 5 should define the 'SPM' acronym. (Should readers assume that SPM is shorthand for 'sun photometer'?)

Page 17, lines 18–19 : Regarding Figure 5, the authors state, "The weak coarse mode may result from aerosols in the boundary layer (marine particles, soil and road dust). The lidar observations do not show this coarse mode." This distinction between full column measurements (AERONET) and range-resolved retrievals (lidar) is well worth emphasizing. When assessing the properties of lofted layers, using AERONET data or parameterizations will always introduce some uncertainties.

Page 17, lines 21–22 : suggest changing "However, in practice, such an approach is not useful" to "However, in all likelihood such an approach would be impractical and/or unreasonably difficult". Separating the contributions from fresh and aged smoke would no doubt be useful. But, as the authors point out, reliably accomplishing the separation would be damnably difficult.

Page 18, line 10 : I wonder what explains the large dispersion of the Table Mountain data seen in Figure 17b?

Page 20, line 27 : perhaps it's worth reminding the reader what the variable x represents? (I had to page back to section 3 to remind myself.)

Page 21, line 6 : first and foremost, the authors deserve a huge round of applause and thanks for the addition of Table 4.

Page 21, line 6 : having 'backscatter lidar' appear twice in the column headers for Table 4 is incredibly confusing. Yes, the explanation is given in the caption. But I strongly believe that it's worth using up a bit of extra page real estate to clear differentiate between ground-based and space-based backscatter lidars in the column headers.

Page 21, line 14 : The authors quite rightly call out the Achilles Heel of elastic backscatter lidar retrievals: "The lidar ratio is even required as input in the basic determination of the backscatter coefficient profiles." This point is, I believe, well worth emphasizing in this manuscript.

Page 21, line 16 : "we assume an uncertainty of 25% in Table 4". Uncertainty estimated for the CALIPSO aerosol backscatter coefficients are given in the aerosol profile products. Their calculation is described in Young et al., 2013 and in the supplementary material for Young et al., 2018.

Page 24, line 14 : regarding Figure 14, and in my role as reviewer #2, I'm disappointed that the authors chose to exclude the third, stratospheric smoke plume from this figure. I doubt other readers will know or care.

Page 24, lines 20–26 : I have several comments on this paragraph.

a) I'm pleased to see the multiple scattering issue specifically acknowledged. For dense aerosol layers, multiple scattering can (and, no doubt, does) introduce substantial unquantified error into the CALIOP retrievals of particulate backscatter and extinction coefficients (e.g., Wandinger et al., 2010; Liu et al., 2011). And to answer a question posed by the authors in their 'responses to the reviewers', I too am of the opinion that the multiple scattering impact in the case of opaque smoke layers is largely unknown. However, the opaque smoke layers identified by the authors in the initial version of their manuscript may offer an opportunity to begin quantifying this impact (albeit very crudely at first).

b) I'm disappointed that only one of the two smoke plumes shown in Figure 14 is further analyzed in Figure 15. In my opinion, the authors are missing an opportunity to establish some practical limits on the application of their method.

c) In the fourth line in the caption for Figure 15, I suggest changing "The CALIPSO backscatter coefficients…" to "The CALIPSO aerosol backscatter coefficients…".

Page 25, line 27 : the DOI for the CALIPSO aerosol profile products is 10.5067/CALIOP/CALIPSO/LID_L2_05KMAPRO-STANDARD-V4-20 (see https://asdc.larc.nasa.gov/project/CALIPSO/CAL_LID_L2_05kmAPro-Standard-V4-20_V4-20)

One final comment: the authors' responses to the reviewers (https://acp.copernicus.org/preprints/acp-2020-1093/acp-2020-1093-AC1-supplement.pdf) claims that "Sections 1-3 are not changed". But even a cursory scan of the revised manuscript shows that this is not exactly true. In fact, the original section 3 ("POLIPHON method: smoke retrieval") has been entirely replaced by a newly titled ("Methodological background: Microphysical properties from backscatter coefficients") and substantially revamped section 3. In my view, this is welcome change.

References

Liu, Z., D. Winker, A. Omar, M. Vaughan, C. Trepte, Y. Hu, K. Powell, W. Sun, B. Lin, 2011: Effective lidar ratios of dense dust layers over North Africa derived from the CALIOP measurements, *JQSRT*, **112**, 204–213, https://doi.org/10.1016/j.jqsrt.2010.05.006.

Prata, A. T., S. A. Young, S. T. Siems, and M. J. Manton, 2017: Lidar ratios of stratospheric volcanic ash and sulfate aerosols retrieved from CALIOP measurements, *Atmos. Chem. Phys.*, **17**, 8599–8618, https://doi.org/10.5194/acp-17-8599-2017.

Wandinger, U., M. Tesche, P. Seifert, A. Ansmann, D. Müller, and D. Althausen, 2010: Size matters: Influence of multiple scattering on CALIPSO light-extinction profiling in desert dust, *Geophys. Res. Lett.*, **37**, L10801, https://doi.org/10.1029/2010GL042815.

Young, S. A., M. A. Vaughan, R. E. Kuehn, and D. M. Winker, 2013: The Retrieval of Profiles of Particulate Extinction from Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations (CALIPSO) Data: Uncertainty and Error Sensitivity Analyses, *J. Atmos. Oceanic Technol.*, **30**, 395–428, https://doi.org/10.1175/JTECH-D-12-00046.1.

Young, S. A., M. A. Vaughan, J. L. Tackett, A, Garnier, J. B. Lambeth, and K. A. Powell, 2018: Extinction and Optical Depth Retrievals for CALIPSO's Version 4 Data Release, *Atmos. Meas. Tech.*, **11**, 5701–5727, https://doi.org/10.5194/amt-11-5701-2018.