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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric and stratospheric wildfire smoke profiling with lidar: Mass, surface area, CCN and INP retrieval" *by* Albert Ansmann et al.

Anonymous Referee #2

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The paper by Ansmann et al. describes a methodology to determine smoke particles' mass, volume and surface area concentrations and then the condensation nuclei (CCN) and ice-nucleating particles (INP). These features are determined for smoke layers in upper troposphere and lower stratosphere as measured by lidars. The estimates are based on profiles of particles backscatter coefficient retrieved from lidar measurements and a series of conversion coefficients determined from photometer (Aeronet) measurements. Two case studies are shown: for stratospheric smoke observations with Calipso and ground based lidar. In my opinion, the paper is acceptable for publication after minor revisions. Please address all the items discussed below.

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Discussion paper

General and specific comments

Pp3, l 2 (Introduction) and Section 3. Please make it clear that in the present study no polarization is used. Thus, it should be clearly stated that from Poliphon method you only use the formulas for conversion factors (based on Aeronet).

Pp7, l 19-21. If I understood correctly, the studies by Haarig and Ohneiser do not do any separation technique for the pollution layers (Poliphon like). So basically, it is just an assumption, eventually based on literature values for LR, depolarization. Thus, I guess that the statement "So we assume pure smoke layers when the smoke is detected in the stratosphere" is enough.

Pp 7, l 23-26: Why you did not use Poliphon for particles separation as you had depolarization information (Fig 1 and Calipso)? Is it just because you assume dust is not present in UTLS? If so, Polihon method can be applied only in lower troposphere. Please clearly state it.

Pp 7, l 32 - pp 8, l 1-7: how do you chose 'an appropriate lidar ratio SL'? Basically, from Table 2 we have a range of 40-110 sr @ 532nm. Based on extensive literature review (39 papers reviewed) shown by Adam et al. (2020), LR at 532 ranged from 26 sr (aged smoke, Muller et al., 2005) to 147 sr (aged smoke, Mariano et al., 2010). Minimum and maximum values found for fresh smoke were 30 sr (Burton et al, 2012) and 100 sr (Mylonaki et al, 2017). Please see Table S3 in Adam et al (2020). Thus, it is not trivial how to choose an 'appropriate' value. Which are the criteria for your recommendations of LR range? Which uncertainty do you assume for them? Please comment more. I do not see using the colour ratio of the lidar ratio in this study and consequently, I am not sure if you should mention this (pp 8, l 5-7). From what I understand, this study is based on backscatter coefficient only. Thus, it is no need to mention LR and further CR of LR (they are not useful here).

References mentioned above:

Adam M. et al: Biomass burning events measured by lidars in EARLINET – Part 1: Data analysis methodology, 13905–13927, https://doi.org/10.5194/acp-20-13905-2020, https://doi.org/10.5194/acp-20-13905-2020-supplement, 2020.

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Burton S. P.: Aerosol classification using airborne High Spectral Resolution Lidar measurements – methodology and examples, Atmos. Meas. Tech., 5, 73–98, https://doi.org/10.5194/amt-5-73-2012, 2012.

Mylonaki M. et al: Aerosol optical properties variability during biomass burning events observed by the EOLE-AIAS depolarization lidars over Athens, Greece (2007-2016), 28th ILRC, Bucharest, Romania (2017).

Pp 8, l 15-16: how can we estimate the few percent BC in the smoke?

Pp 8, l 25: is it 500 nm or 50 nm?

Pp 9, l 8: you used n60 in Mamouri/Ansmann (2016). In that paper it is mentioned n50 in the abstract. In section 2.1 it is mentioned both. In 3.2 you determine n50=c60*sigmaˆx. In the current paper (table 1), you have n50=c50*sigmaˆx. Basically, c is different not n. Please rephrase. Maybe I did not understand (apologies for this).

Pp 12, l 20 (eq. 9): why did you use 440 and not 500? Why did you use 500 in Ansmann et al. (2019a)? Please argue.

Pp 13, l 7 (eq 12): as we use photometer data, it should be mean value of the Vj/tau_j (as in eq. 10). vj and sigma_j are characteristics for lidars only. It is similar for eqs

13 and 14. In my opinion it should be divided by tau_j while having N250 and S (as for column integrated). For eq. 16 I think it should be log(N50) versus log(tau_j). Shinozuka uses also AOD.

Pp 13, l 12: I do not see eq. 15.

Pp 14, l 21: please add date/time of the injection

Pp 14, l 30: how did you chose 0.45? why not 0.4 or 0.5? which criteria did you use?

Pp 15, l 2: same questions for 0.07

Pp 15, l 7: please add info about smoke layer geometry (from Table 1, Ohneiser) as well as about inversion method (i.e. did you use 3 backscatter and 2 extinctions?)

Pp 15, l 21: can you comment on the quantitative differences?

Pp 15, l 30: Figures. . .

Pp 16, l 1: I still don't understand the reason of dividing by D (here, 1000) unless there is a comparison with lidar retrievals. Please revise:

so that ïĄş (in Mm-1) divided by 1000 yield the basic AERONET 532 nm AOT value. Should be: so that ïĄş (in Mm-1) multiplied by 1000 yield the basic AERONET 532 nm AOT value.

Pp 16, l 17-18: you mention the larger spread because you plot both fresh and weakly aged. Why did you plot them together in the first place? Could you please plot them separately? Your statement does not hold for a) (both are spread). For b) and c) there is a better correlation for Yellowknife.

Pp 16, l 27. Why didn't you determine n50 for each fresh and weak aged in Fig. 9? Please add regression coefficients for both fresh and weak aged (besides their cumulative estimation).

Pp 16, l 1-5: how did you compute the conversion parameters (cv and cs) shown in

Figs 7 and 8 for fresh + weak aged? According to the numbers in Table 4, for the mixture f+wa, you have cv=0.181 (cs is the same in figures and table).

P 17, l5 (section 4.3): please clearly mention that the values in Table 4 correspond to the data shown in Figs 6-9. Thus, first row, for aged smoke, shows the result for data shown in Fig. 6 (and Fig 9 for n50). For fresh and weakly aged smoke, you have the results based on Fig. 7 data (and Fig 9 for n50). Please mention that f+wa is the average of each individual fresh and weak aged smoke. Please mention why you did not compute n50 for fresh only and for weak aged only. On table 4 and Fig 9 you show only for mixture.

Pp 17, l 10-11: Why is kext important to show? To show that it is higher for aged smoke? Ansmann et al 2012 does not contain kext for smoke so I am not sure it should be cited here. Several values of kext are given for different aerosol mixtures and different mean diameter of the particles in Horvath (chapter 16 of Atmospheric Particles, ed. Harrison and van Grieken, 1998, ISBN: 978-0-471-95935-9). For mixed absorbing particles, one can see the largest value for kext at 500 nm.

Pp 17, l 25: how is the "appropriate SL of 95 sr" determined? Is it from photometer AOD constraint? Earlier you mentioned (as for Table 2) a range of 65-80 sr at 532 nm. Please comment. Which error did you consider for SL when computing extinction error and further which error did you consider for density when computing M?

Pp 18, l 4: should be n250 instead of n50?

Pp 19, l 25: How did you select SL=80 sr?

Pp 19, l 26: you say you use conversion factors for fresh + weakly aged. In Fig 16 caption it is mentioned fresh only. Please correct. Why you did not show n50 for the two Calipso cases? According to Table 4, there is no n50 calculation for either fresh or weakly aged. There is a calculation only for fresh + weakly aged. As said before, why?

Pp 31, Table 1: change c100 with c50

Pp 33, Table 4: please add info for clarity: Yellowknife (for fresh smoke) and Churchill (for weakly aged smoke) (Figs 7 and 9) and from observations at Punta Arenas, CEILAP-RG, and Marambio (for aged smoke) (Figs 6 and 9). Latest row: I guess it is Lidar (Manaos). Use city instead of region (as for the others). As mentioned above, why you did not calculate c50 and x for fresh and weakly aged? Please calculate them and add them in the table.

Pp 38, Fig 6: change dust with smoke.

Pp 39: see comments above about cv. According to table 4 it should be cv=0.181.

Figs 7-8: I suggest deleting 'except' from 'except for' (which I understand it as "with the exception of"). I may be wrong so please check with a native English speaker. Why do we need fig 8? If you represent the lidar data on Fig 7 is to crowded? I guess it is an overlap of the lidar data (stars in Fig 8) with aged values in Fig. 7.

Figs. 6-9 and corresponding text: please make it clear that equations for cv, cs, c50, c250 (12-14, 16) are based on the data shown in Figs 6-9 (you mention Fig 2 – which has AOT). Thus, in Table 4 we have cv (and other coefficients) for aged smoke based on Fig 6 (and Fig 9 for n50). For cv and the others for fresh + weak aged we have 4 values based on Fig. 7-8 (and Fig 9 for n50).

Pp 43, Fig. 12: Sorry I missed it. What are the two profiles for each of INP representing? We see two red, blue and green profiles.

Pp 45, Fig. 16: used fresh conversion or fresh + weakly aged conversion?

General question: I did not revise deeply section 3 and thus I do not question all the parametrizations and various assumptions. I would like you to comment on the high uncertainty for n50, n250 and INP. We have uncertainties with a factor of 2-3 (i.e. 200 % - 300%). Please comment on CCN and INP values and their uncertainty as input in other studies (e.g. models). If I remember correctly, various models provide some variables with such high uncertainty.

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