Reply to Review #2

by Qiaoyun HU

The paper discusses interesting measurements of aged biomass burning smoke with a unique lidar. The paper is well written and appropriate for ACP. The measurements are performed with a recently introduced advanced lidar that combines multiwavelength lidar, Raman lidar, polarization lidar, and (new!) fluorescence lidar techniques.

Minor revisions are necessary.

p3, l75: When discussing INP, please keep in mind that these aged smoke particles are organic aerosol particles, the organic properties (of humic-like substances) count, and not the ones for soot or fly ash. Therefore, Knopf et al. 2018 …. is appropriate as reference.

Knopf, D. A., Alpert, P. A., and Wang, B.: The role of organic aerosol in atmospheric ice nucleation: a review, ACS Earth and Space Chemistry, 2, 168–202, https://doi.org/10.1021/acsearthspacechem.7b00120, 2018.

Reply: added.

p4, l96: One may cite Baars et al., 2021: Baars, H., et al. (2021). Californian wildfire smoke over Europe: A first example of the aerosol observing capabilities of Aeolus compared to ground-based lidar. Geophysical Research Letters, 48, e2020GL092194. https://doi.org/10.1029/2020GL092194

Reply: added.

p4, l105: Please use 'pyroCb' instead of 'pyCb'!

Reply: Corrected.

p4, l112: Figure 1(b) is mentioned, and then (l114) Figure 3 is mentioned. Figure 2 is left out.

Reply: The order of Figure 2 and 3 is swapped.

P5, l126: No one should introduce Figure 2!

Reply: Reference to Figure 2 added.

p5, l128: AE decreased…

Reply: Corrected. Yes, AE decreased.

p5, l133: To my understanding, Cimel (AERONET) is unable to correctly measure AODs>4.0. And now we have peak AODs of 5.8!

Reply: The new photometer is able to measure AOD up to 7.0. The AOD of 5.8 remains in the Level 2 data, which means it has been validated.

The Case study section is a bit boring, one should better emphasize the deviation of the optical properties on 17-18 Sep 2020 in Figure 5 from the rest, to make the entire story more exciting.

Reply: The presentation of case studies is now modified in the following ways:

1. In the Case 1, the introduction of BBA characteristics is kept and simplified.

2. In Case 2, the differences of BBA characteristics compared to Case 1 are emphasized.

So the repetition of numbers is avoided and the comparison of BBA properties is more clear.

Section 4: Discussion

I miss a clear structure of this section. The discussion could be shortened and should clearly highlight the added value now available in terms of the fluorescence information. Please state clearly: What is new! The discussion should be some kind of a review of the recent Veselovskii papers 2020 (general method), 2021 (on pollen) and the recent one on smoke/cirrus observations (also submitted in 2021) together with the present article on North American smoke.

I would leave out any speculation. For example, the discussion on age of smoke as a function of height. This is just speculation, and usually depends on many different factors such meteorological conditions, fire type, burning material, size of burning area and so on....).

Some suggestions that should be considered. The smoke particles are usually glassy in the upper troposphere and stratosphere (see the review article of Knopf et al.) The organic coating means that the INP properties are controlled by organic (humic-like) material. When discussing heterogeneous ice formation, do not restrict yourself to mixed phase clouds and temperatures higher than -35C. Heterogeneous ice nucleation also occurs at -50 to -70C (in cirrus). All this should be mentioned.

Furthermore, PLDR (or better, ... the shape properties) seem to depend on relative humidity (availability of water vapor) and further gases that can condense on smoke

particles to make them spherical. And the concentration of the gases are high in the lower troposphere and then obviously decrease with height from the middle to the dry upper troposphere and the extremely dry stratosphere.

page 9, line 267-274: I would leave out such a discussion.

Reply: The discussion has been condensed and restructured in the following ways:

1. Speculations and unnecessary discussions about modeling are removed.

2. Section 4.1 and 4.2 merged.

3. The discussion about smoke acting as INP shortened and condensed.

4. The discussion is organized in four paragraphs, each with one different topic: PLDR, lidar ratio, aerosol fluorescence and smoke acting as INP.

page 9, line 275-280, please state clearly how you calculate the lidar ratio, You cannot combine extinction and backscatter values obtained with DIFFERENT smoothing lengths.

Reply: After checking the code of lidar data processing, I confirm that the smoothing length for extinction and backscatter profiles is the same ! I forgot that I had considered the possible artifacts of using different smoothing length when developping the code longtime ago, so the statement in the manuscript was wrong and it has been removed. Thanks a lot for this remark !

page 9, line 287 to page 10, line 297: I would leave this discussion out as well. The paper deals with fluorescence. Please clearly state what is new…! Provide clear facts, what the added value is!

An extra section 4.2 on BBA as INP is not needed in this fluorescence-related paper. A paragraph on the impact of smoke serving as INP is sufficient, but please cover the full range of clouds from mixed phase clouds to cirrus (-25C to -70C), and then a reference to the recent Veselovskii paper on smoke-cirrus interaction is needed.

Veselovskii, I., Hu, Q., Ansmann, A., Goloub, P., Podvin, T., and Korenskiy, M.: Fluorescence lidar observations of wildfire smoke inside cirrus: A contribution to smoke-cirrus – interaction research, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2021-1017, in review, 2021.

Reply: Section 4.1 and 4.2 have been merged.

The conclusion section should finally also be better organized and structured. I do not agree that the fluorescence information is the better information to identify

smoke. It is an additional one, more precise, another independent one, besides all the useful information on PLDR spectrum and lidar ratio spectrum.

Reply: The conclusion section has been re-organized. The sentence--"the fluorescence information is the better information to identify smoke" is ambiguous and has been removed from the conclusion and elsewhere. We want to emphasize that the fluorescence is very sensitive and is accessible even at low aerosol concentration and high altitude, while the calculation of lidar ratio requires smooth and at least moderate concentration of aerosols. But for sure the fluorescence is a supplementary information and should not replace extinction or backscattering measurements.

Figure 3: mixed-phase cloud at 10 km height? Impossible!

Reply: This mixed phase cloud was detected in a subtropical region at (37.91N, 85.41W) by CALIPSO. The temperature at this altitude was roughly about -40 degrees, but the uncertainty could be large and the occurrence of super-cooled liquid water is still possible. In Lille (50.6N, 3.1E), we observed supercooled liquid water clouds at 8 km height and mixed phase clouds at higher altitudes in September. The appearance of ice crystals can be confirmed with the depolarization ratio of 0.2--0.4. These ice crystals appeared in a smoke layer and were possibly initiated by smoke particles. But the nucleation pathway cannot be revolved with the information we currently have and CALIPSO did not observed any mixed phase cloud. So I decided to changed "mixed phase cloud" into "ice crystals mixed with BBA" in order to avoid inaccurate expression.
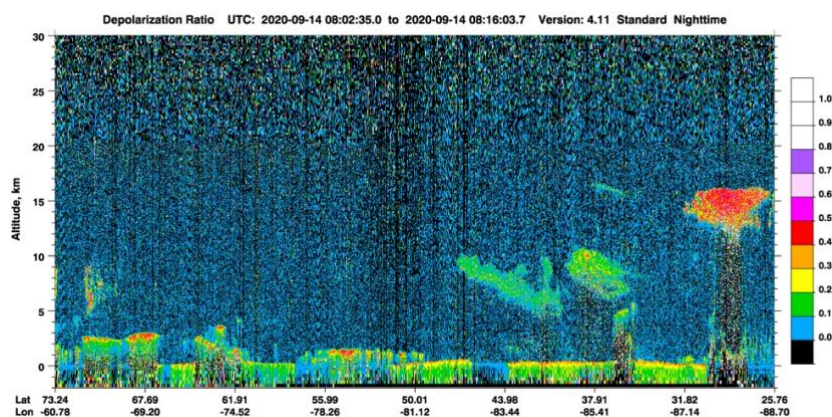

Fig 1. The depolarization ratio at 532 nm, CALIPSO measurements on 14-09-2020
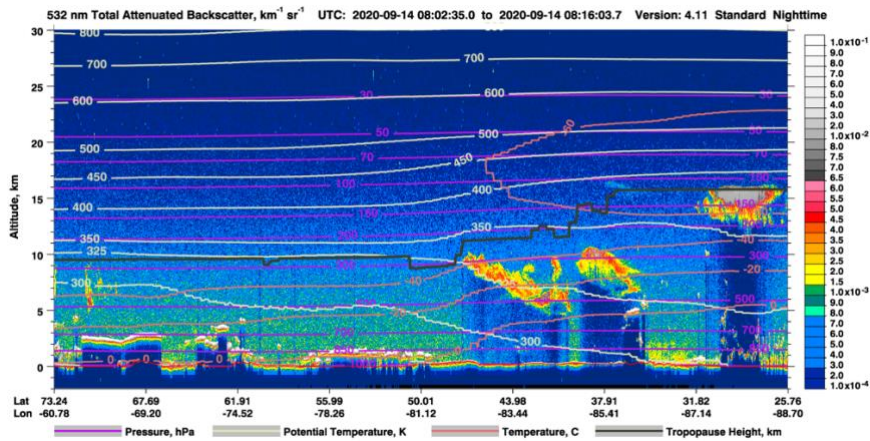
Fig 2. The total attenuated backscatter at 532 nm, CALIPSO measurements on 14-09-2020 overlaid with temperature and potential temperature.

Figure 4: All axis text must be enlarged, ... is much too small at the moment.

Reply: Label size increased

Figure 5: (d) Y-axis EAE/BAE is confusing, better write .... EAE, BAE. In the caption, please explain explicitly which intense parameters are shown. What does P1-P9 mean? Please state that P1-P9 are listed in Table 1....

Reply: Corrected.

Figure 7: Again, all axis text must be enlarged, much too small at the moment.

Reply: Ticks are enlarged.

Figure 8: I would recommend to explain clearly what parameters are shown. Figures should be widely self-explaining.

Reply: More explanations have been added.

Figure 9: Again, all axis text must be enlarged, much too small at the moment.

Reply: Axis ticks are enlarged.

Figure 10: Here one could then state: Same as Figure 8, except.....

Reply: Caption updated.

Figure 11: Again, what is shown... should be stated.

Reply: Caption updated.

Regarding all the figures, keep in mind that many readers may not be lidar specialists and need a lot of information.

Reply: More information has been added in the caption of figures.