Review of "The unprecedented 2017–2018 stratospheric smoke event: Decay phase and aerosol properties observed with EARLINET" by Baars et al. (hereafter B19).

B19 offer a new contribution to the quite interesting set of studies examining a pyrocumulonimbus (pyroCb) smoke plume in the stratosphere that in several regards seems to have been unprecedented in remote sensing data sets going back decades. This work offers a valuable new perspective, involving two ground-based lidar networks: EARLINET and POLLYNET. There are additional additional lidars in Europe that are not involved in this work, but these two networks together have an impressive geographic spread over the continent and operation frequency that enables a sophisticated temporal and regional analysis of an evolving plume. Moreover, the capabilities of certain instruments (e.g. multispectral probing and polarization) give B19 the ability to assess microphysical and cloud-nucleation properties in addition to optical properties of an aerosol plume. An example of the strength of these networks as applied to this smoke event has already been demonstrated by several papers that B19 cite, wherein the nascent phase of this plume over Europe was characterized. Here they follow the plume until it was undetectable above noise levels, six months after the pyroCb injection.

The manuscript is well crafted, logically organized, straightforward to follow, and careful to characterize the levels of uncertainty in the data and processing methods.

B19 present valuable new results. This perspective of the decay of the Pacific Northwest Event (PNE) smoke plume illustrates new constraints on the sensitivity of these lidars to the stratosphere relaxing back to background conditions. They make a strong argument in the Conclusions section for the strategic value of these lidar networks, bolstered by the results they show. Hence this is appropriate for consideration in ACP. However, I have one major concern that keeps me from recommending this for publication. That will be discussed next, followed by a few minor and technical issues.

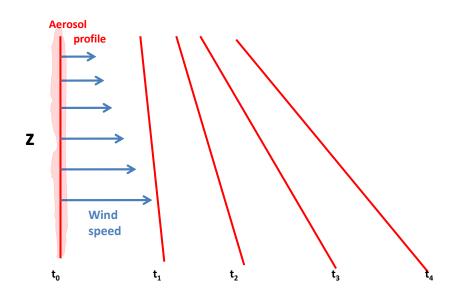
Major Concern:

Considerable attention is devoted to the visualization of a backscattering object referred to variously as, e.g. an "apparently ascending smoke layer," and "coherent, apparently upward moving structure." B19 then explore the most plausible physical mechanism for the supposed ascent. It has already been shown convincingly that the PNE smoke plume underwent diabatic ascent, in a paper cited by B19 (Khaykin et al. 2018). Khaykin et al. utilized a global aerosol data set (CALIPSO), which in my opinion is a requirement for ascertaining diabatic rise. Applying geographically bounded data sets such as these Eurocentric lidars to the task of quantifying diabatic ascent and assigning causal mechanisms is vulnerable to misinterpretation. Inferring diabatic rise from an upward sloping aerosol feature in an altitude vs. time analysis (using single or multiple lidars) is hampered by the additional plausible explanations for that slope that are impossible to resolve locally. For example a

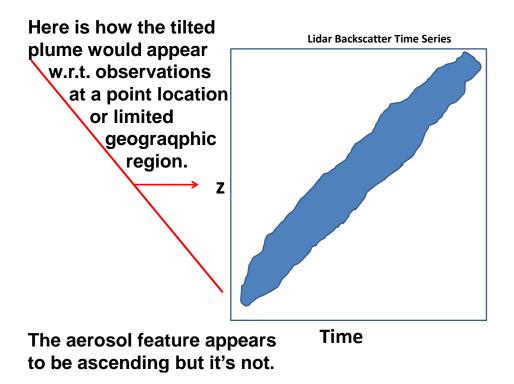
sloping feature might simply be attributable to wind-speed shear operating on an evolving plume. For a plume below the jet max, the effect is an apparently descending slope. For a plume above the jet max, i.e. in the lower stratosphere, the effect would be an apparently ascending layer. The tropospheric example was brought up in the discussion of a previously published ACP paper:

https://www.atmos-chem-phys.net/10/11921/2010/acp-10-11921-2010-discussion.html

The stratospheric analog is shown in the following schematic.



Plume above jet max (dv/dz < 0)



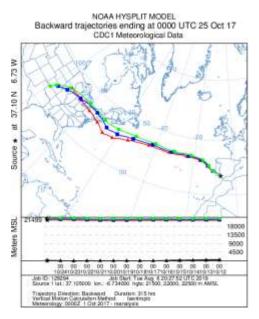
B19 refer to an "ascending...layer" in the context of a time series plot comprising a season's worth of data. In the course of that time it is hard to conceive of the aerosols blowing over that limited domain as simply a "layer." The reader might take "layer" to mean a coherent physical object that is wholly within the sampling beam of those lidars. This of course is not what was occurring within the narrow string of lidars spanning the Mediterranean Sea and Iberia that B19 grouped for this season-long analysis. Over the course of the latter half of 2017 the smoke that blew into that regional swath had a vertical and horizontal history that is totally outside B19's domain.

Complicating the interpretation of diabatic ascent are two established facts about the PNE plume. 1. Above southern Europe the plume was as high as 20 km by late August, according to B19's Figure 3 and Khaykin et al. (2018), Figure . 2. The plume was already higher than 20 km in early September (Khaykin et al., 2018), when B19's nascent smoke signal is constrained to 15-17 km (Figure 7a--the figure central to B19's ascent argument). Hence it can be argued that the representation of smoke over the Mediterranean swath was biased quite low at the outset of the Fig. 7a time series. Given the results from southern France reported by Khaykin et al., B19 are compelled to argue for a physically-based distinction between the plume observed early over Iberia and eastern Mediterranean versus the plume in the French Mediterranean area in late August reported by Khaykin et al..

The fact that PNE smoke was higher than ~21 km in the extratropics by the first week of September means that any smoke observed at such altitudes at any time after that, in the extratropical latitudes of EARLINET and POLLYNET, may owe their altitude to a mechanism other than that put forward by B19. Wind shear meteorology may be one possible factor. The wind speed profile is also a climatological reality and hence may play a role in differential transport with respect to altitude on a longer time scale like that of Figure 7a. I'm not sure how much merit this has; I'd just ask B19 to consider broadening the discussion of the possible forces involved in an upwardly sloping feature such as that seen in Figure 7a.

B19 give a thorough survey of the mechanisms that might explain diabatic transport of absorbing aerosol such as smoke. They find that the only candidate consistent with their data is a pathway proposed by Kloss et al. (2019) (K19). K19, which is at this time still under review, argue for ascent of the PNE smoke by way of a combination of horizontal transport to the tropics and subsequent diabatic ascent driven by the Brewer Dobson Circulation (BDC). If this is indeed the precursor condition and setup for B19's "apparent ascending layer," the entire upward slope in Figure 7a must be the consequence of a coherent, continuous flux of tropical air to B19's Mediterranean lidar belt during the entre fall and early winter of 2017. If this was the driving transport mechanism, it would also be necessary to argue why the "ascending layer" in Figure 7a had apparently little impact on any other European lidars near the Mediterranean belt and presumably downwind of that flux from the tropics. In short, B19's argument was not convincing to me.

I ran an experiment by computing back trajectories from an observation at El Arenosillo (NASA MPLNET) of smoke at 22 km on 25 October 2017, about midway along the Figure 7a time series. The purpose was to ascertain the general trajectory direction at that time/altitude. The results suggest that transport to the observation location was on westerly winds and did not appear to be consistent with flow from the tropics. Hence the smoke at that time over southern Iberia was more likely to have an extratropical history (at least for the preceding two weeks) than tropical. What is not shown be straightforward to ascertain is that along this path one will find CALIPSO aerosol-layer coincidences, at the approximate altitude of the 25 October layer, at several points along the trajectory path. By this example one can argue that individual observations in Figure 7a can be fully and quantitatively explained by simple meteorological transport, in this case from extratropical plume sightings. Separating this history from one imposed by the BDC would seem to be a tall order, and perhaps unnecessary. I'd ask B19 to comment on whether this experiment is well conceived, and if it raises a question as to the pathway of the smoke that is in the apparently ascending layer in Figure 7a.



The robustness of K19's analysis and conclusions is still in question, given that the manuscript is currently under review. Issues were raised with K19's analysis that are analogous to those raised in this review. The discussion is available here:

https://www.atmos-chem-phys-discuss.net/acp-2019-204/

To the extent that B19's conclusions w.r.t. plume ascent depend on the work of K19, it is suggested that B19 assess the issues raised in both reviews and then revisit the question of the causes of the upward slope in Figure 7a.

Minor Concerns:

Regarding the discussion of smoke ice-nucleating properties, B19 make a plausible hypothesis that the huge abundance of smoke at the tropopause could have had a discernible impact on cirrus formation over Europe (and presumably beyond). I would understand if B19 consider further exploration of this as beyond the scope of this paper. However it made me wonder if cirrus occurrence was indeed perturbed on fall 2017. Perhaps B19 could add a brief statement as to whether this is being explored or just generally a topic for future work.

P10, L20-22. Discussion of the relation of depolarization ratio to particle size. This is presented as if it is common knowledge, but I don't think it is. For instance, is this true for dust and volcanic ash? Please elaborate and/or cite the literature establishing this.

Technical Concerns:

Abstract, L5 (and elsewhere): The term "soot" should be defined; otherwise it is ambiguous. E.g. sometimes "soot" is applied to aircraft emissions.

P2, L10: Change "ascent" to "ascend."

P2, L17: Change "2018" to "2017."

P3, L10: "The particles obviously reached the stratosphere as pure soot particles..." What makes this "obvious?" Might it be better to use "apparently" instead?

P3, L12: Change "lead" to "led."

P3, L28: There is another paper that directly deals with this issue. Please consider citing Campbell et al. (2012). <u>https://www.sciencedirect.com/science/article/pii/S135223101100968X</u>

P4, L4: "part of the smoke particles" suggests a micro-level. Please reword. P5, L1: Is there a difference between "ACTRIS-2" and "ACTRIS" is used thereafter? If so, please clarify.

P5, L12: "PBL" should be spelled out at first usage.

P5, L26: "analysis were performed" should be "analysis was performed"

P7, L18: "indicate" should be "indicates" to agree with the singular subject "set."

P7, L21: "(because of the low tropopause height)" What does the tropopause height have to do with the altitude of the smoke layer? Please elaborate or reword.

P11, L6: Change "ascend" to "ascent."

P12, L14: "...could be lifted before." This is an incomplete sentence. Please modify.

P12, L24: "The unprecedented event of ..." is awkward. Perhaps "The unprecedented occurrence of..." instead?

References: Gialitaki et al. Is it proper to cite a paper as "to be submitted"?