Review of Ohneiser et al. (2022), "Self-lofting of wildfire smoke in the troposphere and stratosphere caused by radiative heating: simulations vs space lidar observations"
Reviewer: Mike Fromm

Ohneiser et al. explore the phenomenon of diabatic ascent of solar-radiation-absorbing smoke particles with a combination of satellite lidar- and visible-wavelength imager-based aerosol observations and modeling. The diabatic lofting in their focus is broken down into two categories: free-troposphere-to-stratosphere (hereafter "FTTS") and exclusively stratospheric. The latter of these two has been unequivocally observed in multiple case studies. The former (FTTS) has been hypothesized but not confirmed observationally.

It is the FTTS aspect that is the driver of this work, as revealed in the Abstract: "One of the main goals is to demonstrate that self-lofting processes can explain observed smoke lofting in the free middle and upper troposphere up to the tropopause and into the lower stratosphere without the need for pyrocumulonimbus convection." This is the logical extension of this author-group's prior publications that argued for non-pyroCb stratospheric smoke being the dominant particulate constituent in the Arctic in 2019-2020 (as opposed to Raikoke volcano sulfates, put forward in a host of additional recent publications). Hence, this manuscript attempts to test their hypothesis with observations and modelling support thereof.

The authors' observational test includes not only the Raikoke summer of 2019 but an apparently similar scenario in August 2021 when Siberian fires also produced dense, synoptic-scale, multi-day smoke plumes. They focus on CALIOP backscatter curtains for a few selected days each year over the smoky domain. From those curtains the authors mark vertical locations representing a smoke-layer centroid that presumably represents a traceable ascent of the smoke they attribute to self-lofting. In addition to the CALIOP observations, the authors show HYSPLIT forward trajectories in association with the CALIOP plume heights over the multi-day plume evolution. Back trajectories, briefly described and unshown are also invoked to argue for diabatic ascent of the smoke layers.

I found the entirety of the Siberia CALIOP and trajectory analysis (Section 4.4) to be confusing, arbitrary, speculative, error-prone, hence unconvincing. Below I will point out the various issues I found in greater detail. But in summary, my assessment is that the authors did not demonstrate observational evidence of diabatic self-lofting of Siberian tropospheric smoke in either 2019 or 2021. Absent that evidence, the reader has no basis on which to accept the modeling results presented herein and thus be convinced of the novel hypothesis presented in foundational work of Ohneiser et al. (2021; "O21").

Moreover, the authors clearly did not show incontrovertible observational proof of the FTTS diabatic self-lofting pathway required for the presumed stratospheric source term for the Arctic MOSAiC lidar-based conclusions of O21, on which Engelmann et al. (2021), Ansmann et al. (2021a), and Ohneiser et al. (2022) depended. From O21's Figure 13, one can project that the nascent, tropopause-level Siberian smoke resulting from FTTS transport would have an AOT of ~0.2 to 0.3 in August 2019. Layers embodying such an AOT would present as an unmistakable signal in CALIOP data. By their own analysis in Section 4.4 (Fig. 17), the Siberian smoke in 2019 had ascended only to 5 km (~5 km below the tropopause). Presumably, had there been detectable, efficient self-lofting to the tropopause, the authors certainly would have shown it. In the apparent absence of smoke FTTS proof, the authors were left to state: "Fractions of this smoke plume must have reached the tropopause and later on the lowest stratosphere." Thus, even if their observational interpretations are robust, the authors did not achieve one of their "main goals." The reader is left to rely on hypothetical modeling such as already presented in O21 and updated herein. From the Summary and Outlook section: "the large number of open parameters generates large uncertainty / sensitivity to the modeled lofting rates." Hence, we are no closer to a fire-emission/stratospheric-plume-pathway connection in boreal 2019 as before.

The authors' main goal was to support the contentions of O21, Engelmann et al. (2021), Ansmann et al. (2021a), and Ohneiser et al. (2022) that a boreal stratospheric smoke plume was present in 2019/2020 with AOT and persistence on par with certain volcanic injections and major pyroCb outbreaks. Their approach was to establish proof by observation in support of O21's hypothesis. My assessment is that the observational proof was not given in spite of expectations. If my assessment is accurate, the authors have strategically falsified the O21 hypothesis by looking for and not finding a suitable source term for the stratospheric smoke that O21 and associated papers proffered. This is a valid and valuable scientific conclusion. The current manuscript is thus positioned to challenge those previous conclusions. The authors are asked to consider using this current work to acknowledge the lack of observational evidence for Siberian smoke FTTS transport and the implications for O21, Engelmann et al. (2021), Ansmann et al. (2021a), Ohneiser et al. (2022), etc.

If my assessment is incorrect, it is incumbent on the authors to establish observational proof of the 2019 Siberian-smoke pathway to the stratosphere. The modeling work had essentially been done by O21; the modelling in this paper on its own does not serve as proof.

Regarding the observations and modeling the authors present on the second diabatic pathway, exclusively stratospheric, the simulations make a valuable contribution to the literature. As acknowledged by the authors, the stratospheric scenario is much cleaner and simpler to follow. Moreover, prior publications have already provided a foundation for tracking the smoke-plume features studied herein. Considering my concerns with the FTTS aspect of this manuscript and the implications for publication merit, I will focus my comments on that rather than the stratospheric part.

Major Concerns

L49, “Recently, smoke self-lofting was observed in the middle and upper troposphere (Ohneiser et al., 2021; Engelmann et al., 2021).”: It is more accurate to say that self-lofting was inferred. Only a single CALIOP curtain was displayed by O21, giving a static view of vertical aerosol placement. Inferences were made based on Lagrangian trajectories and modeling.

L55-57, “…Siberia in the absence of pyroCb convection as spaceborne lidar observations indicated (CALIPSO, 2022).”: How can CALIOP data be used to show there was no pyroCb action? The cited reference gives no help. In this Introduction section, it is presumed that any cited material will support the specific statement made. The reference here is a generic identifier of the CALIPSO data repository. In point of fact, there were 5 pyroCbs in the Sakha region of Siberia in July and August, 2021. PyroCb alerts and follow-up discussion in 2021 were shared in near real time on the Worldwide pyroCb Information Exchange (https://groups.io/g/pyrocb), for the specific purpose of engaging researchers across the globe and documenting individual pyroCb eruptions. The authors are asked to review the communication on this open platform and revise their description of the 2021 boreal and Siberia pyroCb occurrence.

L57, “Part of the smoke again entered the stratosphere in August 2021.”: No doubt. One or more of the reported pyroCbs (see above) naturally injected smoke to or across the tropopause. Moreover, many other boreal pyroCbs were catalogued in 2021 (see the above link to the reporting medium). It is inappropriate for the Introduction to this paper to combine the claim of no pyroCbs and the supposed self-lofting from Siberia to the stratosphere.

L59-65, Discussion of smoke heating, buoyancy, self-lofting: It is important to treat the arguments of Boers et al., de Laat et al., Torres et al., and Yu et al. separately. Torres et al. and Yu et al. dealt explicitly with a puroCb-generated source term at the tropopause and primarily stratospheric transport whereas Boers et al. and de Laat et al. considered lofting of non-pyroCb-generated smoke in the lower to middle troposphere (analogous to this manuscript’s Siberia hypothesis). The distinction I call for is because stratospheric self lofting has been well characterized observationally (to which the Kablick, Khaykin, and other citations in this paragraph attest) whereas the FTTS pathway has not. It also must be noted that the Boers/de Laat pathway on Australia’s Black Saturday has been reinterpreted as a classic pyroCb event, with rapid injection to the tropopause (Fromm et al., 2021, https://doi.org/10.1029/2021JD034928). Please revise the discussion here to reflect the disparate levels of certainty regarding pyroCb vs. non-pyroCb smoke pathways.

L199, “Chemical aging is assumed to lead to a spherical shape…”: On what basis is this assumed? Please defend this statement. There is abundant literature showing that aged tropospheric smoke particles embody depol. ratios that are significantly greater than the limiting value for uniform spheres, e.g. Dahlkötter et al (2014; doi:10.5194/acp-14-6111-2014), Burton et al. (2015; doi:10.5194/acp-15-13453-2015), Hu et al. (2022; https://doi.org/10.5194/acp-22-5399-2022),
Liu et al. (2022, https://doi.org/10.1016/j.jqsrt.2022.108080). Hence, Table 2’s “2%” “spherical” characterization is apparently an inaccurate simplification of non-pyroCb, tropospheric smoke.

Please revise to reflect the literature. Since the weight of the above-mentioned papers rests on the idea that aged smoke is better represented by an aspherical model than monolithic spheres, the implications of changing Table 2 are momentous. The authors are asked to reconsider their assumptions for aged tropospheric smoke and the implications for the optical properties they observed in the stratosphere during MOSAiC.

Regarding the concern above regarding the authors' assumption of spherical, aged smoke in Siberia (and eventually in the stratosphere), it is important to note that the only other published work describing non-pyroCb, FTTS transport is de Laat et al., (2012). Even if it is maintained that their "solar escalator" was an accurate depiction, the resultant stratospheric smoke from Black Saturday had depolarization ratios weeks after onset so great that the CALIOP observations thereof triggered a feature classification of "ice" (https://acp.copernicus.org/preprints/acp-2021-117/acp-2021-117-RR1.pdf). Hence these aged smokes were wholly inconsistent with the assumptions shown in Table 2. The authors are asked to comment on this inconsistency and make the appropriate revisions to their current assumptions on smoke aging.

Section 4.4. This section is problematic. My concerns are many and deep. This part of the paper is seriously flawed. Below I list the issues individually.

L482, "The smoke at different heights was transported in the same air column.": Meteorologically, I cannot make sense of this. What does "same air column" mean? The trajectories take different paths and have very different endpoints. This implies considerable speed and directional shear, which by itself seems to suggest a variable air column. Please explain.

Discussion of Figures 17 and 18: How "old" are the smoke layers on 12 August 2019 and 4 August 2021? Without knowing that, the reader has no idea how all those smokes got to that start point. Almost assuredly they didn't all arrive in a single "air column," but no information is given. I.e. these seem like arbitrary start points and hence it is unclear how to assess transport after the start points in relation to transport to them.

L482, 483, "The trajectories analysis in Fig. 18a
indicated similar wind speed and directions at different heights in the middle troposphere.": I cannot reconcile Fig. 18a with this description. There are significant differences in both aspects. If the authors want to convince the reader that these trajectories are similar, they need to quantify that and show what dissimilar trajectories would look like.

L483, 484, "The smoke slowly ascended from
around 3-4 km height to 5-6 km height.": The trajectories themselves account for almost a 1-km altitude gain. I.e. resolvable meteorology is responsible, not diabatic lofting. The authors need to acknowledge this.

L484, 485, “The backward trajectories (not shown) indicate no direct link of the air mass at 5-6 km height with lower heights (and thus a direct fire smoke uptake from the sources).” For this to make sense, the trajectories must be shown. Even so, it is difficult to understand what the authors mean here. What is “direct…uptake”? What are “the sources”? Either remove this sentence or bolster it with displayed trajectories and precise terminology.

L489-494, Nearly identical trajectory discussion regarding the 2021 smoke: I have the same concerns as stated above. Please take the same corrective actions.

L494, “Initial smoke plumes were at around 3 km height on 4 August 2021.”: What is meant by “initial” when the previous sentence refers to smoke 10 days earlier. Please expand and clarify with more precise information.

L495, 496, “Fractions of this smoke plume must have reached the tropopause and later on the lowest stratosphere.”: What does this mean? Why “must” the smoke have behaved like that? Was there a stratospheric, Arctic aerosol plume in Fall 2021 on par with the Raikoke/MOSAiC episode of 2019? Given the suggestiveness of the CALIOP observations in 2021 as compared to 2019 (the plume altitude and AOD are greater), one might hypothesize that a similarly strong and long-lasting plume was in evidence in 2021/22. Can the authors confirm such a plume? If so, that would be a powerful set of circumstances. If not, it would tend to shed doubt on the smoke composition of the 2019 Arctic stratospheric plume. The authors are asked to quantify the Fall/winter 2021 stratospheric aerosol load in comparison to 2019 and draw appropriate conclusions.

Figure 17. Unless the authors explain the rationale or algorithm for marking the “plume center height” in each panel, it is reasonable to conclude that the choices are arbitrary. If they have just been eyeballed, one could make an independent set of marks justifying no altitude gain; there are aerosols from the boundary layer to nearly 10 km in most panels. Moreover, the authors show one selected CALIOP curtain each day and thus don’t account for the full synoptic view of the smoke on each date. A fuller synoptic view of the plumes on these dates, involving multiple CALIOP curtains per day and horizontal map views of the overall plume (using something like visible imagery, UV aerosol index, carbon monoxide) are essential for the reader to have confidence in the arguments brought here. And, as mentioned above, some traceability from 12 Aug 2019/4 Aug 2021 back to a definable source is also called for.

Figure 17a: The longitude “170” seems to be incorrect for this CALIPSO orbit. It should be ~150E.

L486, “The observation were taken over the East Siberian Sea north of Siberia (78°N, 160°E).”: What observation? Do the authors mean “observations”? If so, what observations? 78N, 160E is not near the CALIPSO track, as far as I can tell. The orbit shown in Fig 17a is much farther west. On that track, the lon at 78N is ~123E. The center of the thick smoke layer in 17a is north of 80N, but still far west of the trajectory start point. Please clear up the confusion.

Figure 18: Why are the trajectories initialized at 07 UTC? This is neither close to the night or day orbits of CALIPSO as far as I can tell. Why not initialize them at the time of 12 Aug 2019, 4 Aug 2021 CALIPSO observations? If they were initialized at the CALIPSO observation time, that should be clarified in the caption. Assuming I have found the correct CALIPSO orbits to go with the two start points, the forward trajectories launched from them are significantly different than those shown. Please clarify or explain.

Figure 17a caption: Why are these heights (3, 5, 7 km) chosen? The initial layer is said to be at 3 km. Secondly, the displayed map legend says 3, 4, 5 km. The trajectories conform to these values. Are the heights in the caption the ones intended for display, or is the caption incorrect?

L489-497, Discussion of the 2021 smoke: There were 5 pyroCbs in the Sakha region of Siberia between 26 July and 7 August. No accounting of pyroCbs in 2021 is given by the authors. In addition to the Siberia pyroCbs, there were numerous pyroCbs in the USA and Canada in July and August. There was even a possible pyroCb in Greece in early August. Hence, there were multiple direct inputs of smoke into the UTLS that could have been transported to the Siberia zone in the time frame under study. It is therefore possible that some of the elevated smoke in Figure 17b is from pyroCb or pyroCu injections (when a pyroCb event occurs, naturally and frequently they are accompanied by numbers of pyroCu, which efficiently pollute the free troposphere. See Figure 7 of Fromm et al. (2021; https://doi.org/10.1029/2021JD034928) and attendant discussion. The authors are asked to account for the added complexity and uncertainty regarding Figure 17 and associated analysis.

L487, “The black dots indicate the position of the smoke layer…”: The smoke in each panel is seemingly much wider than the dots represent. A clearer explanation of the dot location choice is needed.

Figure 17, star symbol: What is the significance of the star symbol versus the dot? The star symbol is not defined in the text or figure caption.

L511-513 (End of Section 4.4), “We can conclude that even tropospheric smoke can ascend significantly from lower tropospheric injection heights up to the tropopause level within a few days and even enter the lower stratosphere as demonstrated in Ohneiser et al. (2021).”: Once again, O21 did not “demonstrate” with observations that smoke self-lofted across the tropopause in amounts to justify their conclusions. They were only able to hypothesize this. In the current work, the observational aspect did not demonstrate the FTTS diabatic pathway, even if what they showed in Figure 17 was totally robust. No stepwise continuation of the smoke lofting to and through the tropopause was demonstrated. This should have been achievable given the enormous AODs and multi-season aerosol persistence O21 reported. Moreover, in all comparative respects, 2021 Siberian smoke equaled or exceeded 2019 in terms of AOD and height. Considering these two primary factors as inputs to the self-lofting model, Fall and winter 2021's boreal stratosphere should embody a smoke plume at least on par with 2019. If that condition did not occur, there was effectively no observational demonstration of the FTTS diabatic lofting in either season. The authors are encouraged to assess that logic, dispute it, or consider a reinterpretation of their hypothetical model.

Minor Issues

Figure 1 (and lines 38-41): The images are offset by one day from those stated in the caption. The actual dates are 3-5 January, not 2-4 January. The thickest smoke on 3 and 4 Jan over the Tasman Sea is lower to mid-tropospheric. The UTLS smoke from the December phase of ANYSO pyroCb event had already blown east of New Zealand. The only substantial UTLS smoke in Fig. 1 is on 5 Jan. The arrows point to the plume from the 4 Jan pyroCb phase.

L42-46, discussion of smoke over cloud: The radiative implications of smoke over cloud are very complex. Referring to them in the context of Fig. 1 snapshots seems to draw relations that are likely to be irrelevant. Smoke plumes blown by fast UTLS winds will be almost totally decoupled from most cloud systems. There won't be much time for a cloud, small or big, to have an impact on a plume blowing faster than and independently of that cloud. Making the case for cloud effect on self-lofting calls for a much more sophisticated discussion.

L43, "compact layers": Please define "compact."

L67-68, discussion of dust self-lofting: The Daerden et al. paper is on Martian dust. Applicability to the Earth is unclear and not mentioned. Please elaborate or remove this citation. It was not evident to me that Gasteiger et al. discussed anything but vertical mixing, which is not synonymous with self-lofting. I could not find observational evidence therein of dust diabatic lofting. Please point it out if I missed it.

L187, "Siberian fire smoke reached the stratosphere via the self-lofting process.": As in other places in this manuscript, this is stated as fact. The authors hypothesized this in O21, and make statements elsewhere herein to the effect of "it must have happened," but definitive observational proof does not predate this paper. The authors are asked to modify the wording here and throughout to reflect the uncertainty of the FTTS self-lofting pathway.

L187-192: Citations are needed for the Kuwait- and Canadian-plume BC fraction. Same goes for the fuel types in Canada, Siberia, and Australia.

L193-195: "more homogeneous": Please define "homogeneous" since it is used as a comparator to pyroCb plumes.

L196, 197, discussion of pyroCb-plume depolarization ratio: A citation is needed for the "20%" value. Also, these double-digit depolarization ratios last far longer than the "fresh" stage. Please reword accordingly.

L271, "coherent structures": Please define "coherent."

L312-319, discussion of Kuwaiti oil-rig fire plumes: One of the prime science topics centered on these plumes was a validation of the Nuclear Winter hypothesis. Because of the size and darkness of these smoke plumes, it was thought that this might have been a natural (or at least inadvertent) experiment in self-lofting. Yet there are no publications to my knowledge that revealed significant self lofting. Rather, the consensus seemed to be that in spite of the absorption and optical depth of the plumes, little or no diabatic lofting was observed (Hobbs and Radke, 1992; Larry Radke, personal communication, 2008). Hence the strategic value of this case study is brought into question. The authors are asked to reflect on the published findings of observed Kuwaiti smoke heights as a function of time and make suitable revisions to the manuscript.

L446-454, Discussion of Figure 16 and MODIS AOT: This is a very interesting result. But the method needs to be explained better. Just how are the MODIS AOT data evaluated? What is done when there are no MODIS AOT retrievals due to clouds or smoke flagged as cloud? Given that MODIS data are daytime values, were CALIOP daytime curtains used for MODIS associations? And what are the implications of the results? Are the CALIOP-based AOT's low biased? Are the MODIS data a better approximation for the stratospheric plume AOT?

L471-476: The importance of this paragraph is not clear. The authors seem to be saying that if several sources of uncertainty all align in one direction, the overall error in final plume height can be very large. If that is a correct interpretation, isn't that self-evident? Please clarify.

L481, "Almost coherent smoke plume structures…": Assuming "coherent" was already defined, what is meant by "almost coherent?"