Review of revised Friberg et al., "Short and long-term stratospheric impact of smoke from the 2019/2020 Australian wildfires." (Hereafter "F23").

Reviewer: Mike Fromm

F23 are to be commended for the efforts to update and improve the material they presented. Their track-change document is very helpful to the reviewer. These changes reduce, in many ways, the areas of uncertainty and confusion that I harbored regarding the initial submission.

However, these changes were not persuasive in terms of F23's assertion that 1. the January-phase plume had an insignificant initial stratospheric component and 2. that January-phase Australia smoke was transported from the troposphere to the stratosphere. Each of these is discussed in turn, in the next section.

My overarching concern with F23 is that they posit diabatic self-lofting from the troposphere to the stratosphere as a pathway leading to a stratospheric smoke pollution event greater than the direct pyroCb pathway acknowledged as having occurred five days earlier. This is an extraordinary claim, which requires extraordinary evidence in support. Given the acknowledged circumstances (the a priori existence of a major stratospheric smoke plume), this paper must show incontrovertible evidence of a slow, diabatic intrusion to the stratosphere that is as definite as the already published material on the Australia Black Summer pyroCb event. The manuscript does not meet that challenge, in my assessment. The ANY event still calls for explorations such as this. But the complexity of the atmospheric situation is not resolved or clarified by F23's analysis to my reading. Absent that outcome, I cannot recommend this publication. Major changes are called for.

General Concerns

Regarding point 1 above, F23's messaging is that their data and interpretation were supported by Peterson et al. (2021). But my assessment is that the data are still contradictory and the Peterson et al. results are mischaracterized. F23 downplay the January-phase stratospheric injection by citing Peterson et al.'s revelation that the December-phase injection mass was 2-8 times greater than that of the December phase. However, the January-phase injection mass was by itself on par with the 2017 Pacific Northwest pyroCb event (PNE), which was at that time unprecedented in the satellite record for stratospheric smoke perturbation. Hence it is reasonable to imagine an Australia event that consisted only of the January phase. The Peterson et al. stratospheric smoke-mass injection could logically have resulted in a lasting plume on par with PNE. Peterson et al. calculated the stratospheric smoke source term by two methods, one based solely on the UVAI. The UVAI maximum, combined with the stratospheric area of the UVAI plume, showed that the January-phase event was on par not only with PNE, but all other historical documented pyroCb events as well.

As useful and advantageous as CALIOP data are for the purpose of characterizing nascent pyroCb plumes, they are—like all other remote sensing data items--subject to the vagueries of optimal sampling. The nature of sampling and the naturally small footprint of a nascent smoke plume is that they are often missed or incompletely captured with satellite data like CALIOP. My intense experience with the study of the ANY event, no doubt the same as F23's, revealed that the January-phase smoke plume was missed by CALIOP such that ideal plume height and concentration was delayed for several days. There were missing complete orbits or orbit segments on some days, and other days where CALIOP was just unlucky. Our survey of CALIOP showed that CALIOP made some luckier samples of the nascent December plume, as shown in F23's figures. This goes to the heart of why I asked in the first review about daytime CALIOP curtains and OMPS-LP, even conceding the poorer quality of those data. F23 showed in their supplement several daytime CALIOP curtains, implying the recognition of their value toward a more complete characterization of the nascent smoke plume. The unfortunate lack of perfect CALIOP sampling should not be convolved in the interpretation of plume altitude and concentration. In the case of ANY and PNE, the UVAI-based estimates of the immediate stratospheric presence of smoke are an independent and robust marker of the stratospheric smoke source term. In short, there is a large, built-in uncertainty to the early sampling of CALIOP, even if all the CALIOP data are utilized. Unless F23 contend that the Peterson et al. January-phase stratospheric smoke mass is largely overestimated, the Peterson et al. results stand as an indication that the 4 January pyroCb event impregnated the stratosphere on par with almost all prior pyroCb events. If the veracity of the Peterson et al. estimates are not disputed, the challenge of showing additional smoke entering the stratosphere thereafter is considerable. F23 are encouraged to consider this argument and make whatever changes are called for in their line of analysis.

On the second point above, F23 made it much clearer in this revision how they accounted for the separation of December and January event smoke layers. Their CALIOP survey in Figures 1, 2, and Supplementary was more complete; their connection to phase 1, phase 2, and "other" was reasonable. From those data they built the central figures 7 and 11. This made it easier to understand these two important figures. But in my assessment, it did not adequately prove a diabatic pathway from the troposphere to the stratosphere, which is their central claim. Figure 7 contains only stratospheric observations. The CALIOP data assigned either to December or January are wholly or largely tied to two isolated, contained, and circulating smoke plugs followed by Allen et al. (2020 https://doi.org/10.1175/JAS-D-20-0131.1), Kablick et al. (2020), Khaykin et al. (2020), and Schwartz et al. (2020, **https://doi.org/10.1029/2020GL090831**). F23 expressly tied their December-phase data to one of these "isolated" entities. They did not directly tie their January-phase CALIOP data to an isolated smoke entity, but Figure 7a maps out the path of the plume in a way that compared almost perfectly with that of a smoke-vortex element illustrated in the above-cited papers. The important point is that the morphology of both of these smoke entities is wholly achievable by quasi-Lagrangian means due to their confinement and the absence of compromising uncertainties regarding smoke decay. These two contained smoke vortices perceptibly rose diabatically, in comparison to the extra-vortex ANY plumes (as hinted at by F23's "other" CALIOP data points in Figure 7).

F23's expressed and incidental following of these contained smoke vortices stands in contrast to the tropospheric CALIOP data they present. No attempt was expressed to follow these

CALIOP layers materially. F23 clearly outline the difficulty of characterizing tropospheric smoke morphology because of the various, dominant, ubiquitous processes such as wet deposition. Canonical application of these forces to tropospheric smoke would lead to a vertically stratified decay profile that could appear as identical to the evolution of the CALIOP tropospheric smoke observations in Figure 11. For this reason alone, the apparent slope upward in the troposphere in Figure 11 has more than one explanation, unlike the same slopes shown for the stratospheric subset.

The supplement plots by themselves cannot be used to infer upward transport. Static images 24 hours apart allow no such definitive statement. Moreover, I could not discern a systematic rise in the tropospheric smoke from these figures. If indeed there is such a signal, it would be essential for F23 to analyze and defend that scenario.

F23 should be expected to offer other potential explanations for the pattern seen in FIgure 11. Given that wide-scale tropospheric smoke pollution on par with that seen in this case is not uncommon, and other such occurrences were not followed by significant stratospheric pollution, alternate explanations should be given at least equal weight to diabatic lofting.

It is noteworthy that the smoke vortex associated with F23's January-phase CALIOP data was tracked by Schwartz et al. by virtue of MLS stratospheric water vapor enhancements. If this smoke entity had its origin in the troposphere, it would have been subject to the cold trap at the tropopause, a limiting factor on its water vapor content. The fact that all of the ANY contained plume elements shown by Schwartz et al. were defined by water vapor enhancement, and that they all were based in the lowermost stratosphere, indicates a commonality, that being a direct pathway via pyroconvection. If that is a defensible statement, the challenge for F23 is to show that slowly ascending air masses in the troposphere can deliver not only smoke through the cold-point tropopause, but water vapor plumes as well.

Figure 8 is composed of zonal averages. The range from 20-80S would embody tropopause heights generally above 14 km in the northern realm to lower in the southern realm. Since this analysis is done with an absolute altitude scale, it is to be expected that the data below 14 km is a blend of tropospheric and stratospheric aerosol. Since these data almost certainly represent some unknown blend of stratospheric air, it is uncertain as to how to assess these lowermost data points in relation to those above 14 km.

Both phases produced UTLS smoke with equally small depolarization ratio (depol for short). E.g., see the intense stratospheric layer in Figure S3b,c. The fact that the December phase produced such low depols at such high altitude prompts the question as to the true difference between the December and January nascent UTLS plumes' particle-shape populations. Could F23 comment on that?

F23 have convincingly shown that the January-phase stratospheric depol is somewhat less than the December phase. But what does that necessarily say about its origin altitude? Wouldn't we expect the photolytic process to drive both depol populations to the same eventual value? If so, what would account for the difference in the December and January aged plumes at stratospheric altitudes?

Peterson et al. showed that, by number, there were many more pyroCbs in the December phase that injected only to the troposphere than on 4 January. That may or may not have been evident in the CALIOP curtains shown in F23, but it is plainly evident in the full set of CALIOP curtains that there was abundant tropospheric smoke from 29 December onward. Given Peterson et al's accounting, doesn't it seem reasonable that there was much tropospheric smoke from the December phase that was in place by the time of the January phase?

Technically, F23 do not show diabatic transport from below the tropopause. They infer it. The stratospheric fraction of observations can be accepted as reflecting diabatic lofting largely on the strength of prior publications such as Khaykin et al;, Kablick et al. and Schwartz et al., as well as the several papers on the PNE pyroCb event. But the troposphere-to-stratosphere mechanism has still not been proven with observations.

In terms of the flow of argumentation in F23, I found several instances where they assert their conclusion about the diabatic lofting from the troposphere prior to any detailed analysis. In this way they seemed to put the cart before the horse. It is advised that they not only bolster their analysis proving the tropospheric diabatic lofting, but also withhold any conclusions/assertions until the reader sees the proof.

F23, on occasion, refer to the smoke-layer tops in the LMS as "minor" in relation to the bulk of the aerosol layer. This is reasonable from a descriptive perspective, but considering the strong vertical stratification of aerosol lifetime, this may be prejudicial. There is no doubt that typical pyroCb events distribute smoke throughout the troposphere up to the UTLS, leaving what may appear to be a minor portion at the topmost altitudes. But that "minor" part has a much greater potential to last than the eye-catching tropospheric parts. Moreover, sampling by CALIOP may give the strong appearance of the topmost smoke as being small in proportion to lower plumes. A telling example of this is shown in Figures 6 and 7 here:

https://doi.org/10.1029/2021JD034928. It shows an early view of the PNE smoke plume that captured a small footprint of the pyroCb smoke plume. This "minor" feature represented the most consequential early indication of the smoke that polluted the stratosphere. Hence F23 are asked to reflect on the "minor" indications of the January-phase LMS smoke as early as 6 January illustrated in Figure 11.

F23 responded to my initial review's question about exploiting CALIOP color ratio. They argued that the differential attenuation made that an insurmountable hurdle. However, they did exploit CALIOP color ratio in Martinsson et al. (2022) in a manner I was alluding to. Hence, I still wonder if one could discern systematics in the temporal evolution of the color ratio. Both the color ratio and the differential attenuation are associated with particle size. There might be qualitative as well as quantitative tactics to assess the temporal changes even while acknowledging the attenuation issue. Since this group met that challenge in Martinsson et al., they are encouraged to do the same here or explicitly address the reason why they avoid that in this work.

Katich et al. (2022, DOI: 10.1126/science.add3101) used in situ aircraft data to develop a "fingerprint" of pyroCb stratospheric smoke, in comparison to non-pyroCb smoke, in terms of an especially large coating thickness around the BC core. F23 may wish to review this paper and comment on any potential conflicts with their hypothesis of photolytic processes reducing OA material mass.

Targeted Comments/Concerns

Below, line numbers, figure numbers, and F23 quotes are in **bold**. My reaction is in plain text.

L30: The vertical definition of the LMS is not given herein. The term is used sometimes qualitativley, but also used as part of a targeted calculation of AOD. The details of that calculation need to be provided.

L53-54, "At least 38 PyroCbs injected smoke to the stratosphere during two events…": Please revisit Peterson et al. and revise this statement. According to Peterson, only a subset of the 38 pyroCb-pulse injections reached the stratosphere.

L59-62: Even with F23's revisions, the statements in this paragraph conflate two previous published conclusions under the banner of transport from the troposphere to the stratosphere. Only Ohneiser et al. fits within this pathway. The Peterson and Khaykin papers start the plume ascent in the lowermost stratosphere. Please add the proper nuance.

L73, "…aerosol stayed in the stratosphere for a year…(Ohneiser et al. (2022).": As far as I can tell from Ohneiser (2022), they only show PNE smoke for ~8 months. Did I miss it? If not, please provide another citation.

L125-126, "The depolarization ratios for smoke from the 2nd fire were clearly lower than those for smoke from the 1st fire…": While some systematic difference is visually apparent, there is a considerable overlap in depol between the two phases. The December phase generated some very low-depol layers above the tropopause, as did the January phase. There are several additional CALIOP curtains attributable to the December phase, not shown here, that reinforce the realization of overlap in the depol ratio between phases. In general, it appears the free tropospheric smoke depol has single-digit depol, stratospheric has decidedly double-digit depol., and tropopause-level smoke has a wide range, as manifested in both phases. Would the authors care to comment on that?

L125 paragraph and figures called out: It would be helpful to have marks such as arrows on the figures pointing to features the authors want to highlight to make their point.

L127: What property? F23 describe two populations of smoke depol, but not in contrast to other particle types. Please elaborate.

L231-232, "Peterson et al. (2021) reported much larger stratospheric impact from the 1st fire, based on studies of the fires' immediate impact.": Yes, but the stratospheric mass from Phase 2 was equivalent to PNE, according to Peterson's Figure 1. So, on its own merits, the Phase 2 plume was a major stratospheric presence.

L197 and elsewhere: "elevation" is regularly used to characterize an increase in AOD. This term also denotes changes in altitude. It might be advisable to choose another descriptor of the AOD amplitude change.

L232-233, "10 days after the PyroCb formations we start to see more stratospheric influence (Figure. 7).": Figure 7's first January-phase data point is on 14 January, ten days post event. But Figure 2 and especially supplementary figures show January-phase CALIOP curtains dating to 5 January (1 day post event). Moreover, Figure 11 starts on 5 January. But the reader is first introduced to the January-phase smoke by the callout to Figure 7. So, it seems to be misleading to support the above statement by this figure callout. Figure 11 shows stratospheric influence from the January phase being first detected by CALIOP on or about 6 January. The weight of Figure 2, 11, and the supplementary figures indicates that a re-characterization of this sentence is called for.

L233: It's not clear what **"stratospheric influence"** means. Figure 7 simply follows two smoke vortices. This doesn't represent the entirety of the smoke plume. Please consider rephrasing this.

L236, "Over time, more and more smoke…": This is not obvious from the CALIOP curtains in FIg 1,2, and Supplement. This is a conclusion stated before any proof is given.

L239, "…rose by at approximately the same rate as…": Again, Figure 7 simply follows two vortices, one of which was spawned by the January plume. So it is no surprise; the ascent of that vortex has already been documented by Khaylin, Kablick, and Schwartz.

L257-258, "Some of the smoke from the 1st event reached the UT (Figure. 1) and may have risen later along with smoke from the 2nd event contributing somewhat to the second AOD peak.": This is putting the cart before the horse. The reader has yet to see any analysis proving tropospheric lofting.

L259, "Smoke from the 1st event rose markedly in the stratosphere before smoke from the 2nd event entered the stratosphere (Figure 7b).": This conclusion cannot be drawn from Fig 7. F23 started the plots on 14 January for the January plume. The various CALIOP curtains prove that there was stratospheric smoke from the Jan phase many days earlier. See also Peterson et al.

L265, "…the 2nd event that ascended later into the stratosphere.": At this point the reader has not been shown proof that tropospheric smoke ascended into the stratosphere. The material that has been presented at this point does not perform that task.

L295-297, "This explains the low depolarization ratios for smoke from the 2nd event…": See my comments about the copious observations of low-depol. tropospheric smoke in the December plume. Note also the multiple CALIOP observations of low-depol, tropopause-level smoke from both events.

L326-327, "From the 1st event we do not see evidence of extensive crosstropopause transport beyond the initial PyroCb…": The reader does not get any information aligned with this conclusion. What analysis did F23 perform to elicit this finding?

L354-355, "A continuous crosstropopause transport over the course of several weeks also affects the AOD evolution.": This pathway is taken as a given here. Cross-tropopause transport is not quantified in a manner to support the claim that it occurred over several weeks.

L356-357, "Our study indicates that smoke from the 2nd event had larger long-term impact…(Figure 9.": Nowhere, to my reading, did F23 explain how Figure 9a was constructed. Elsewhere in the paper they described tracking the two phases of smoke only out to 4 February. How did the smoke-phase distinction over several months get accomplished?

Figure 5: Do F23 wish to opine on the secondary AOD increase in the winter months? This period is not a focal point of the paper but the feature of increased AOD is in stark contrast to the decay signal, thus potentially more consequential than the earlier AOD dip and rise on which F23 focus.

Figure 7 caption, "…smoke transport and chemical evolution…": "chemical" is not shown, only optical properties. Chemistry is inferred from simulations, which is discussed in the text. But the figure caption should describe what is shown.

Figure 9 caption, "Smoke decay in the dense isolated cloud from the 1st event..": This first sentence is confusing. The figure panel a shows both events. And panel a's description is "zonal means." How are events segregated while calculating zonal means?

Figure 11. The fit lines are not described in the caption.

Supplementary Material, "Some Caliop curtain plots were excluded from the analysis since the aerosol attenuation signals were too weak even though the UVAI indicate presence of smoke aerosol.": This is unclear. Since there is no one-to-one relation between night CALIOP and day UVAI, how did F23 determine the weakness threshold used for excluding CALIOP layers?

Technical Comments

Figure S1: Please separate the left and right columns. The continuous black background makes it difficult to see the east edge of the left column and west edge of the right column.

Throughout the text, use subscript notation in “SO2.”