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

Reviewer: Mike Fromm

Overview

F23 present a new satellite-observation-based analysis of the Australia New Year (ANY) 2019/20 fire/pyroCb event. Their aim is to quantify the stratospheric smoke burden and evolution, resolving the two separate contributions from the December and January phases of ANY. There are three distinctive and new contributions herein. 1. determination of smoke AOD decay via photolysis, 2. non-pyroCb-pathway stratospheric pollution by the January-phase plume, yet 3. dominant influence of the January-phase smoke on the overall ANY stratospheric perturbation. In this pursuit, F23 employ aerosol data from nadir-viewing OMPS absorbing aerosol index, limb-view OMPS-LP aerosol extinction, CALIOP backscatter, and H₂O data from MLS.

This is a pursuit worthy of study and natural for ACP. The ANY case, individually and by comparison to other stratospheric smoke and volcanic events, is still imperfectly understood and the subject of varying accounts regarding transport pathway, physical evolution, composition, and radiative impact. This study intends to reduce the uncertainties in at least two important ways.

However, F23 have taken on a very complicated scenario, involving continuous smoke generation in southeast Australia in late 2019 and early 2020, punctuated by episodes of pyrocumulus (pyroCu) and pyrocumulonimbus (pyroCb) activity, in a dynamical meteorological setting. I was not convinced by F23's core new finding—that the January-phase smoke was injected only into the troposphere and didn't rise into the stratosphere until ten days post event. F23 build their case with a subset of their data items that under-samples the downstream smoke to the detriment of accurate plume-height and transport-pathway characterization. F23's analysis of aerosol-layer stratification in their illustrations is vague. In addition, their considerable reliance on OMPS UVAI maps (Figure 1) to aid in plume distinction as the smoke blends together is problematic.

F23 comes on the heels of two other papers attributing ANY's unprecedented mass of stratospheric smoke to a non-pyroCb pathway. Hirsch & Koren (2021) found no pyroCbs, only random oceanic thunderstorms to be the pathway. Magaritz-Ronen, & Raveh-Rubin (2021) concluded that pyroCbs did not suffice, and that a single Pacific-Ocean synoptic-scale cyclone on ~2 January performed the task. Now F23 attribute the preponderance of ANY stratospheric smoke to tropospheric self-lofting as the pathway while briefly dismissing the above two explanations. Given that Peterson et al. (2021) demonstrated clusters of pyroCbs in both December and January that penetrated the tropopause, and unprecedented UVAI plumes immediately following each impulse, is it F23's position that neither the pyroCb nor the cyclone nor thunderstorm pathway can explain the long-lasting stratospheric smoke observations? If so,

their analysis of satellite-data, transport, and lofting needs to be much more exacting than what is presented herein.

My assessment is that if F23 can mount a convincing argument of a diabatic troposphere-to-stratosphere pathway for the January-phase plume, the balance of material could become defensible and thus merit publication. But as will be elucidated in my report, F23'sanalysis of the December and January ANY smoke impulses is unclearly developed and plausibly incorrect. Hence, I recommend major revisions of this manuscript after considering all the concerns I list below.

A major concern of mine is that F23 discounted a crucial element of the pyroCb "smokestack"

General Concerns

phenomenon. The ANY event, like all pyroCb cases, involves the direct injection of smoke, ice, and biomass-burning gases to the top of the pyroconvective column. Like all convective exhaust, these materials disburse at column-top altitudes. These injection heights are quantified in the case of ANY and several other pyroCb events (such as Black Saturday, PNE, Chisholm, etc.) by weather-radar reflectivity and/or infrared brightness temperature image data. In the case of each, it is known that the column-top estimates are generally conservative, i.e. low biased. This has been documented (Fromm et al., 2021, https://doi.org/10.1029/2021JD034928 and references therein). Hence the source term for any such pyroCb event is reliably known to the extent that the pyroCb clouds are thus characterized. In the case of ANY, Peterson et al. (2021) provided an exhaustive accounting of all of the December- and January-phase source terms. Both the December and January phases embodied plumes that topped out in the LMS. Detection of those plumes post-pyroCb are then subject to spotty sampling by any and all satellite instruments. It is regularly the case that pyroCb-plume case studies involve these imperfect satellite data items convolved with various time lags between injection and ideal sampling. Because these fire events also involve somewhat continuous emissions throughout a wide range of vertical transport that the downstream plume picture is embodied by thick and thin smoke plumes from the surface to the topmost injection heights. Incomplete/imperfect sampling of these plumes can be suggestive of a multitude of pathways and processes. In the case of ANY, we can state with certainty that the December and January pyroCbs created smoke plumes that topped out above the tropopause ambient on the active dates. In the case of the 4 January pyroCbs, echotops exceeded 16 km (and Θ exceeding 380 K). Consistent plume heights were observable and traceable to these pyroCbs, 2-3 days post pyroCb. But even if such downstream sampling was non-existent or woefully incomplete, it wouldn't change the underlying veracity of the "smokestack" pathway and endpoint. Therefore, the challenge for identifying any other contributing pathway to the stratosphere in the case where direct injection is clearly established is thereby heightened. F23's Figure 2 and 3 appear to show confirmation that both December and January ANY smoke matches the smokestack expectation. Additional support for this end-to-end connection is provided in the supplement at the bottom of this review. F23 are encouraged to weigh this argument and evidence, and consider how it informs their approach.

Figures 1 (daily UVAI maps for ~5 weeks), 2 and 3 (ensembles of CALIOP curtains) are used to make the case that the December-phase of smoke injections immediately reached the lowermost stratosphere (LMS) but the January-phase smoke did not. As one would expect, the smoke emissions over the weeklong pyroCb event generated plumes that were straightforward to separate for a brief period, then became indistinguishable based on the UVAI alone. F23 blended the UVAI data with selected nighttime CALIOP curtains to attribute a portion to December and the balance to January. They discuss how they evaluated plume transport to connect these elements with the two phases, but that transport was only inferred via the dayto-day change in the UVAI plume positions. I could not find any additional tool for sourcereceptor connection such as Lagrangian trajectories. By its nature, the UVAI depends largely on AOD and altitude such that a low, dense layer's UVAI could be indistinguishable from a high, optically thin layer. It is obvious from F23's Figure 2, 3 that multiple layers, from the free troposphere to the LS, were the rule downstream of ANY. The flat color gradation used in the UVAI maps limits any meaningful discernment of low/high, dense/thin features. And as mentioned above, after the first week of January, the previously distinct UVAI plumes become inseparable. F23 recognize this and qualify their analysis with their selection of CALIOP curtains matched to the UVAI features. However, every CALIOP curtain used by F23 is from a nighttime orbit segment. These are systematically ½ day offset from the UVAI-measurement time. This might be OK if there is only one layer at stake and that layer is governed by very light winds. But given the true complexity of the smoke layering and natural wind-shear involvement, the association of the CALIOP features with the UVAI is wholly uncertain. F23 might have invoked other imagery-based retrievals available at night, such as IR-based CO, but they did not. F23 might have invoked daytime CALIOP curtains, but they did not. Hence the reader is inadequately informed about the true association of the CALIOP and UVAI features. On its merits, this complicated ANY smoke event requires a more rigorous and precise accounting between the CALIOP/UVAI and the two ANY injection phases.

For defensible reasons, F23 do not employ OMPS-LP aerosol extinction data as they do CALIOP for the assessment of the nascent plume altitude. However, this may have been a missed opportunity, given the limb-view data's coincidence with the OMPS-NM UVAI. While acknowledging F23's cited concerns about OMPS-LP utility in the presence of optically thick plumes, its natural combination with the UVAI can and does allow for a confident characterization of LMS plume-top. Moreover, it will be shown that such a combination reveals a finding of the LMS position of the young January-phase plume days before F23's conclusion.

F23's main analysis leading them to conclude that the January-phase pyroCb smoke was almost totally relegated to the troposphere (until it diabatically lofted across the tropopause ~13 January) exploits the CALIOP curtains shown in Figure 3 and selected point observations in Figure 8. Figure 3 has 12 panels, curtains from 5-10 Jan. Most panels show dense smoke straddling or above the tropopause. Presumably F23 do not attribute these to 4 January. But it is unclear how they interpret Figure 3. The text discussion calls out Figure 3 but does not offer any in-depth explanation of the various aerosol features. On its face then, Figure 3 seems to

contradict F23's premise. A sufficiently detailed discussion, commensurate with the many Figure 3 panels and the multitude of aerosol layers therein, is essential. By the way, this recommendation also applies to Figure 2 and attendant analysis.

F23 exploit CALIOP depolarization ratio in an interpretation of particle morphology. Two separate threads are presented. 1. They evaluate a temporal increase of depolarization in the stratospheric smoke, in combination with AOD decay, as evidence of photolysis-imposed loss of organic mass fraction. 2. Small depolarization of the January-phase tropospheric smoke as a sign of aging in a humid environment with a transformation to more spherical particles.

Regarding point 1, two questions arise. First, if the particles are losing mass, one might expect to see a change in CALIOP color ratio commensurate with particle-size reduction. F23 introduce CALIOP attenuated color ratio in Figures 2 and 3, so it is natural to ask if they analyzed the temporal variation of that quantity in relation to depolarization ratio. If so, those results would be to F23's advantage to report. If not, it is reasonable to suggest that F23 perform this analysis to more fully support their line of argument. Also, Baars et al. (2019; https://doi.org/10.5194/acp-19-15183-2019) showed that the 2017 Pacific Northwest stratospheric pyroCb plume decreased in depolarization over the course of ~3 months. Might F23 cite that work and comment on the implications of such transformation? Would that be consistent with a resupply of organics or other shell material on the BC core? Is there evidence of CALIOP depolarization ratio decline after the flattening shown in Figure 8?

Regarding point 2, is it reasonable to speculate on how particles that have a collapsed, nearly spherical BC core would become less spherical during the aging in the stratosphere? What is the evolution of the depolarization ratio of the tropospheric smoke? Is there a perceptible decline in time, consistent with the proposed aging? A more concrete concern regarding this point is that F23 generalize the ambient condition of the tropospheric January-phase smoke as "humid." While it is indisputable that the stratosphere is dryer than the troposphere, the speculation here about the ambient conditions implies (to me) that the smoke particles are far below the relatively dry UT. Is it F23's expectation that the January plume was largely lower than the UT (as several of the lower, dense layers in Figure 3 are) and thus lofted by several km before entering the stratosphere? See my earlier comment about the need for a much more precise treatment of the features in Figure 3.

I was confused by the general treatment of the December and January plume discussion. It appears that F23 are dealing separately with the vortical, rising plume elements and the overall ANY stratospheric smoke. Given that the two compact plume sub-elements represent a minor contribution to the overall AOD of the ANY smoke, the discussion of the global AOD evolution of the two phases seems to be unrelated to the compact-feature evolution. Perhaps I did not catch on to the interplay between the two themes. Please point out to me what I am missing, or otherwise add some discussion material to help the reader appreciate the two seemingly independent themes.

F23 claim that the January-phase smoke plume ascended gradually from the troposphere to the stratosphere over the period of 4-14 January. Figure 3, which chronicles the perceived 4 January smoke via CALIOP imagery terminates on 10 January. There are no additional granular CALIOP curtains filling the gap to 14 January. Presumably F23's suggested rise of the smoke to the tropopause would be discernable beyond 10 January (Figure 3). The reader should have the access to the full interpretation of the smoke evolution to be convinced of F23's evidence. The 4-day gap in this timeline stands as a barrier to this understanding. It is essential for F23 to present this entire timeline or explain how the evidence they show in Figure 3 is sufficient to secure their claim.

Section 3.8, "Smoke transport to the stratosphere" is perhaps the most consequential section of this paper. Yet it is populated by weak, confusing, and misleading points. Next, these are shown in bold followed by my reaction in plain text. "The NA wildfires in 2017 showed that self-lofting by radiative heating of the dense smoke layers caused smoke to rise from the UT into the LMS (e.g. Khaykin et al., 2018; Peterson et al., 2018)." This is an apparent mischaracterization of these two cited papers. Neither made that argument, or even hinted at it. The diabatic lofting observations they showed all started at stratospheric altitudes. If I missed these points in the two cited papers, please point them out to the reader. "Ohneiser et al. (2021) suggested self-lofting of smoke from the midtroposphere as cause of extensive smoke layers in the Arctic stratosphere during in the end of 2019 and beginning of 2020." F23 might consider balancing this statement by citing Boone et al. 2022; https://doi.org/10.1029/2022JD036600), who categorically refuted Ohneiser's characterization of extensive stratospheric smoke at that time. "Hirsch and Koren (2021) argued that smoke injections to the stratosphere may have occurred in the first week of January via crosstropopause transport by convective clouds south of the fire region (38°S), where the tropopause height is lower. However, the temporal evolution in the UVAI (Fig. 11a) indicates that most of the smoke remained north of 40°S, and only a minor portion was located south of 45°S." But Figures 1 and 2 show that there was abundant smoke south of 45S in late December and the first week of January. This comment is in no way an endorsement of Hirsch & Koren's claim; it is meant to point out that F23 show data that are in apparent direct conflict with this statement. The 2nd event fires (Jan 4) positioned dense smoke layers in the mid and upper troposphere (Fig. 3). Figure 3 shows tropopause-level and/or LMS smoke in 9 of the 12 panels. As I commented above, F23 do not examine Figure 3 (or 2) in granular detail or perform an explicit feature-by-feature source attribution. The reader is given no framework on which to match the above statement with the illustrations. We see evidence of vertical transport during the following week. What is this evidence? If this is referring to Figure 3, the evidence is not at all clear to me. For the reader to see F23's evidence, a much more detailed analysis of Figure 3 is required. The generally low depolarization ratios (<0.10) are not indicative of cloud formation. What is the relevance of this statement? Since this is referring to Figure 3, it is apparently meant to compliment F23's argument that this is smoke from 4 January Australia. Moreover, there are several large non-cloud features in Figure 3 that have depolarization ratio >>10%. Hence, this statement requires expansion toward a full characterization of the various scenes provided. "...we suggest self-lofting by radiation heating and isentropic crosstropopause transport as the cause of transport to the stratosphere for smoke from the 2nd

event, thus following the rising trend in the stratosphere, as demonstrated in Fig. 8b, also in the upper troposphere." "We suggest" implies a considerable amount of uncertainty (as opposed to, e.g. "we find" or "we demonstrated"). Prior to this statement F23 claim to see "evidence" of self-lofting in the troposphere. So, the reader needs to know if their evidence is inconclusive, which would leave F23 in the position of just "suggesting" it. If their evidence is shown to be conclusive, then their presumed position would be that tropospheric self-lofting was demonstrated. Please explain the claimed evidence and discuss their confidence level in the interpretation. This statement also mentions isentropic cross-tropopause transport as a pathway to the stratosphere. They made no prior claim of finding evidence of this for the January-phase smoke. Their only treatment of this pathway was in reference to and partial dismissal of Magaritz-Rohnen & Raveh-Rubin (2021). Hence, it is unclear what justification there is for the suggestiveness of isentropic cross-tropopause transport. Please elaborate, or consider dispensing with this suggestion.

Targeted Concerns

L198-200: "The 2nd fire event occurred on January the 4th, but smoke from this event showed only little immediate stratospheric influence (Fig. 3) in line with observations by Peterson et al. (2021)." What are the observations from Peterson et al. that support this statement?

Section 2.3: In this introduction of the MLS water vapor, it is called for F23 to cite Schwartz et al. 2020; 10.1029/2020GL090831) who did an in-depth analysis of the ANY MLS H2O data.

Figure 1: How is the AI color scaled? What are the ascending orbits? Why are they shown? All the panels have grainy resolution. It is very difficult to discern all the additional layer features.

Figure 1. I was not able to find a thorough description of how the green-line demarcation between December and January phases is justified. Please explain in a way that applies to every figure panel.

Figures 2 and 3: The 532 nm attenuated backscatter panels all have the meteorology-data overlay. These are not described in the captions or main text. Moreover, many of the panels are cropped such that the isoline labels are not displayed. If the meteorology data are necessary, they should be appropriately labeled and exploited in the main text. If they are non-essential, please consider reducing clutter by showing just the lidar data. Define "main layer" and "minor layer".

Figure 8b: The earliest triangle is ~10 days after the fire event. But the earliest triangle on the map is not where the phase-2 plume was on 14 Jan (10 days post event). This point appears to

be close, geographically, to the position on or about 7 Jan when Khaykin et al. started following this plume element. What is the date of this earliest triangle? If it is indeed ~7 January, then the triangles in Figure 8b are offset incorrectly. If instead the x-axis is relative to the December phase alone, this should be explained.

L287-288. "However, the depolarization ratios during the first week of January does not indicate any frequent cloud formation connected to the smoke layers." Meteorologically, what is meant by "frequent cloud formation?" What are the smoke layers to which F23 are paying attention? The reader has no basis on which to accept this assertion. Please bolster it with a callout to and detailed analysis of Figure 3, or providing an additional figure in support.

Conclusions, L322. "Smoke was injected to the stratosphere from two events." The first event was attributed by F23 to the December pyroCbs. What was the second stratospheric injection event? If F23 maintain the claim that the January impulse was strictly tropospheric, should this sentence be amended? If the second stratospheric injection event was from the January pyroCbs, this contradicts the arguments that are the basis of F23's findings. Please clarify.

Technical Points

Figure 2. "Four days CALIOP curtains...": Insert "of" before "CALIOP."

Figure 2 caption. "The day of the fire...": What is meant by this? Should it be "day of the pyroCb? Considering that this was a multi-day pyroCb phase, it would be advisable to revise this terminology.

Figure 2 caption. "malfunction": A description of the malfunction is called for, along with a citation so the reader knows what the anomaly is.

Figure 2, and throughout the document. "29 Dec.": The first phase of the ANY pyroCb event was a multi-day affair, 29-31 December. Please consider describing this phase in that way.

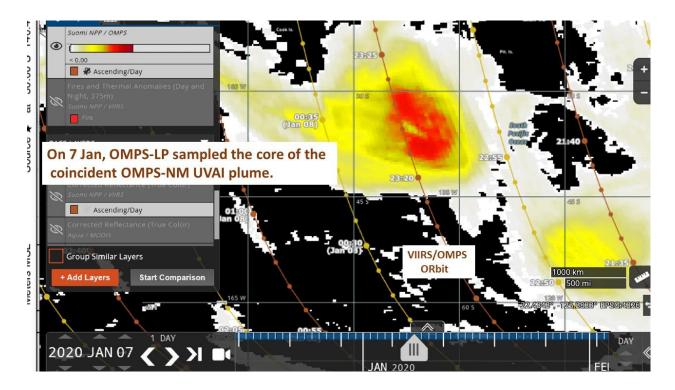
Figure 3 caption. "Same as figure...": Missing Figure number.

Figure 9a: A scaling factor for the extinction coefficient is missing.

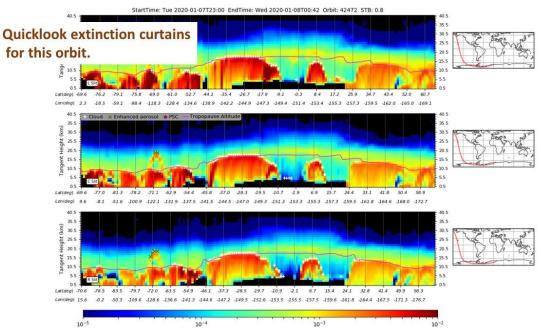
Figure 9c: "1.0" on the color bar: Should be "0.10"

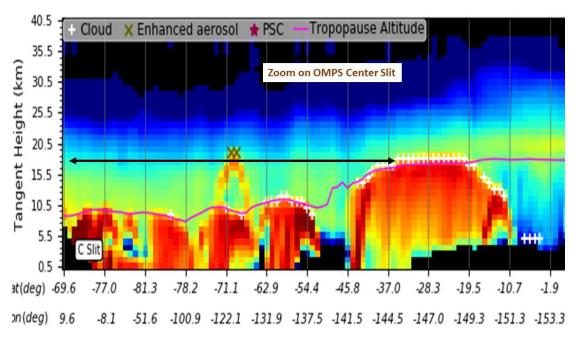
3.2 Compared to volcanism: Please state what is being compared to volcanism in this section heading.

<u>Supplement</u>



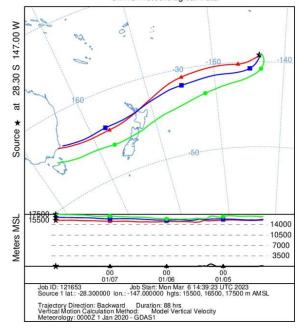


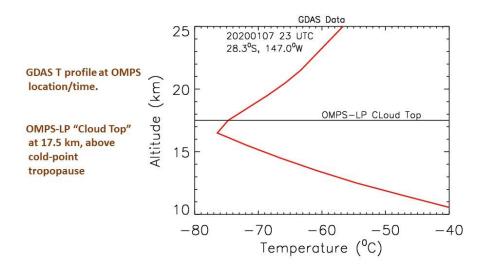


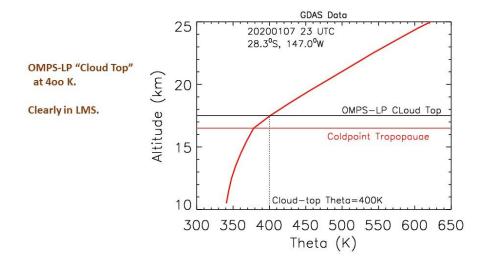


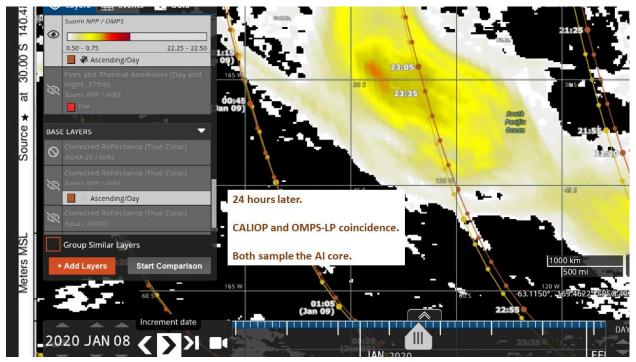
NOAA HYSPLIT MODEL Backward trajectories ending at 2300 UTC 07 Jan 20 GDAS Meteorological Data











Aerosol Extinction Values Retrieved at 997 nm(km⁻¹)

