Summary Statement

O21's defense of their Siberian smoke hypothesis is admirable yet unconvincing. Hinging all their conclusions on the position that one and only one conclusion can be drawn from a peculiar wavelength dependence of lidar ratio places this paper at odds with a mountain of evidence that something other than smoke was in the stratosphere in 2019 before, during, and after O21's hypothesized smoke incursion scenario. If one considers the combination of stratospheric aerosol abundance, omnipresence, and longevity O21 show, this event is thereby in a category with ultra-strong pyroCb events and big volcanic eruptions. It is imperative then for O21 to offer more than a hypothesis if this work is to merit publication in its present form.

The MOSAiC lidar data are an invaluable resource that will illuminate multiple, exciting findings. It is of course exciting to contemplate a new UTLS pathway for smoke to pollute the stratosphere in such a big way. But it would be equally exciting to learn that stratospheric smoke and volcanic sulfate might both give somewhat similar lidar-signal patterns. Given that there is overwhelming support for a hemispheric volcanic sulfate plume in 2019/20, the MOSAiC lidar-data analysis would be fundamentally improved if Raikoke influence was put on an equal footing (at least) with the hypothesized Siberian smoke explanation.

Major Concerns

O21 are standing by a concise claim that all of the MOSAiC UTLS lidar-detected aerosol they show is wildfire smoke. They give no quarter to other compositions (such as Raikoke sulfate). This is a huge challenge, which not only requires convincing the reader that only smoke can explain their measurements, but also to explain the fate of non-smoke particles in the UTLS that were undoubtably abundant at high latitudes in summer 2019.

The new Fig. 5 shows that all trajectory overpasses of Box 2 occurred before or at the onset the ramp-up of Siberian AOD (Fig. 3). The 7-km trajectory is centered in

Box 2 on 17 July, 4 days before the onset of the large-AOD episode declared by O21. The 9-km trajectory parcel moves rapidly over Box 2 on 20 July, when AOD is unremarkable. For the explanation of self-lofted smoke to be of consequence, conditions within Box 2 on 20 July would have to have been primed by large, low-altitude AOD some days prior. Per O21's analysis of Fig. 1 and 3, such a condition did not exist. Hence it is unclear why that CALIOP curtain is shown in support of their premise. Moreover, in my original review I explained that CALIOP curtains looked like the 26 July one every day before that for several days. Thus, there is a consistent picture of ubiquitous UTLS aerosol in place that cannot be attributed to Siberian smoke. Whether the ambient UTLS aerosol is smoke from previous, unrelated injections or Raikoke sulfate, it must be confronted in terms of what happened to it such that it apparently vanished and was replaced by self-lofted Siberian smoke.

O21 base their hypothesis (and an upcoming paper) on Boers et al. and de Laat et al. Boers et al. was a theoretical prelude to de Laat et al., laying the framework for the Solar Escalator paper. On its own, the Boers et al. paper stands as a still unproven mechanism for lofting smoke from the lower to upper troposphere.

de Laat et al.'s position, that pyroCbs did not occur on Black Saturday, has been contradicted by observations given in multiple publications (BOM, 2009; Cruz et al., 2012; Dowdy et al., 2017). Pumphrey et al. (2011) proved that stratospheric enhancements of Black Saturday emissions were detected on the day after the pyroCbs. If BOM, Cruz, Dowdy were in error, and the Boers/de Laat mechanism was solely responsible for the stratospheric smoke plume documented by Pumphrey et al. and Siddaway and Petelina (2012), it would be reasonable to predict that O21's hypothesized aging, and its impact on particle depolarization, would drive the post-Black Saturday lidar landscape. CALIOP measurements of the stratospheric plume would exhibit the same contradictory signals as claimed by O21. I.e. the Black Saturday stratospheric smoke would embody nil depolarization and thus be dominantly mis-classified as sulfate. This is not the case. A perusal of CALIOP backscatter curtains of ~1.5-month-old Black Saturday smoke reveals native measurements of enhanced depolarization. In fact, it is likely that the enhanced depolarization was a factor in the CALIOP version-4 feature classification scheme. The layers are regularly

labeled as "cirrus" in lock step with classification of the layer as cloud composed of ice. This is in spite of the fact that the layers are above 20 km altitude. An example of one such scene is given here:

https://tinyurl.com/caliopsmoke

O21 are encouraged to survey additional CALIOP aged Black Saturday smoke detections from March 2009. They reveal other spurious classifications (such as volcanic sulfate) mixed with cirrus. It is evident that the best explanation for these features is smoke from Black Saturday. (See Siddaway and Petelina and Pumphrey et a;. for maps of the advected Black Saturday plume in the tropics.). As with the boreal 2019/20 situation, the lesson is that no single remote-sensing instrument probing the UTLS is sufficient for unambiguous characterization of particulate composition. This is why total reliance on MOSAiC lidars for characterizing three seasons' worth of aerosol observations requires several complementary data items, and the context provided by publications such as Kloss et al. (2021) and Cameron et al. (2020) in addition to Johnson et at. (2021).

The argument made by O21 regarding the published sAOD (e.g. Kloss et al.) falling short of the MOSAiC 0.1 value is without much merit. There is little doubt that sAOD in Kloss et al. is probably biased low, in part for the reason given in O21—saturation. The sAOD values shown therein, peaking at about 0.025, are also an artifact of the broad aerial/temporal averaging applied. Hence they make a poor point of comparison with individual lidar profiles. That being said, it is straightforward to see in CALIOP data that the stratospheric aerosol at high latitude prior to the hypothesized Siberia incursion, far exceeds sAOD=0.025. Take for example a CALIOP curtain on 22 July, with an aerosol layer over North America with native level stratospheric backscatter exceeding .003/sr. Applying a conservative lidar ratio of 50 gives extinction exceeding 0.1. <u>https://tinyurl.com/gtdot1</u>

The MODIS AOD analysis in Fig. 2 is impressive but inconclusive. No accounting is given of any significant difference in peaks. Several additional peaks are also quite impressive. Might one conclude that in those years a similar, scalable impact on the stratosphere was predictable? Were any observed? It should be straightforward to do so with the available ground-based lidar and satellite remote sensing data sets. Another caveat is that MODIS AOD is severely low-biased in the

presence of high-concentration aerosol plumes (Figure 7; Fromm et al., 2008). It is akin to a saturation bias; thick aerosol is classified as cloud. The import here is that there is a huge unknown in any MODIS AOD analysis focused on extraordinary plumes. The Siberia smoke situation in July/August 2019 was indeed extreme, but the true quantifiable extreme here and in many other cases is unknowable based solely on MODIS AOD retrievals. Hence it is unclear how quantifiably unique the Siberia 2019 smoke situation was. That being said, if indeed the Siberia 2019 smoke was lofted to the UTLS, it should be elementarily possible to follow the lofted smoke plume with satellite data such as CALIOP. If O21 can show observations of day-to-day, stepwise escalation of optically dense smoke from its initial placement to the tropopause and beyond (in accord with their preliminary theoretical calculations), this could be a compelling argument to include in the present thesis.

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