Review of Ohneiser et al. (2022), “Self-lofting of wildfire smoke in the troposphere and stratosphere: simulations and space lidar observations”

Reviewed by Mike Fromm

Note: “O22” is shorthand for the author group of this manuscript. “O21” is used to refer to Ohneiser et al. (ACP, 2021).

This manuscript is a first revision.

Assessment Overview

This review was hampered by the fact that the author tracked change (ATC) document is inaccurate. For example, the paragraph on lines 204-213 is not in the original manuscript. Yet it is un-highlighted in the ATC document. Hence, I abandoned my attempt to use the ATC document.

O22 have responded to both reviewers' comments and substantially modified the manuscript. My assessment is that their responses to my deepest concerns were inadequate and the revised manuscript is as flawed as the original. Their crucial new section, replacing the flawed original section, abounds with demonstrable inaccuracies, misinterpretations of satellite data, conflicting messaging, and unmet expectations from their new, season-long CALIOP analysis.

O22 have now made two attempts to justify the Ohneiser et al. (2021) hypothesis of tropospheric smoke self-lofting without success, in my assessment. Unless the scope of this paper is refined by eliminating the section on tropospheric self-lofting, it does not merit publication.

Since little was changed in sections other than the replaced one, I will limit my review to the major change they made.

<u>Major Concerns</u>

First, it must be stated that a core tenet of O22's thesis is that Raikoke AOT cannot explain any more than about 10% of the MOSAiC stratospheric aerosol. They established that argument in the published O21, arguing that Raikoke sulfate AOT did not exceed 0.025, citing Kloss et al. (2021). Therein O21 acknowledged that the 0.025 value is a "mean value for the latitudinal belt from 40–55N in August 2019" as compared with specific MOSAIC lidar measurements. This is an "apples-to-oranges" comparison. Even though this is now established in peer-reviewed form, it must be invoked anew and questioned because O22 maintain this assertion as fundamental to their argument that smoke dominates the MOSAIC stratosphere while inadvertently presenting and interpreting contradictory lidar data (details given below). In short, they show an Arctic CALIOP layer, ascribed to Raikoke sulfates, that has an AOT far exceeding 0.025, even 0.1. Thus, it is abundantly clear that an apples-to-apples comparison of native lidar data diminishes the published and maintained assertion that Raikoke cannot explain the MOSAIC AOT.

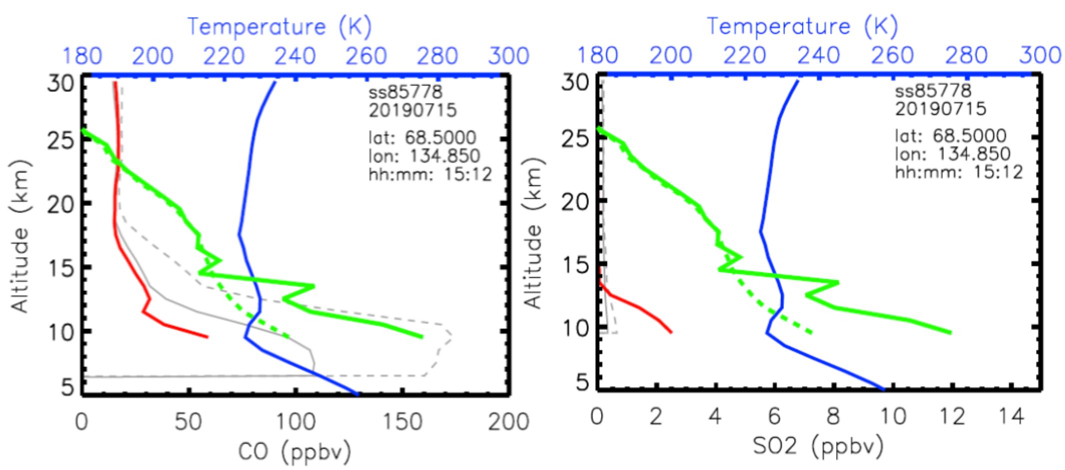
15 July 2019 CALIOP Analysis (Fig. 14a)

In O21 and again in O22, the Siberian tropospheric smoke buildup deemed to be the source of the self-lofting began about 20 July 2019 and reached a peak around 26 July. O22 now present a CALIOP tropopause-level aerosol observation on 15 July 2019 as their centerpiece "footprint" of the self-lofting pathway, "(downwind) of the main fire areas" (See Line 530-531). They emphasize that they have examined every relevant CALIOP high-latitude curtain between the Raikoke eruption date and early October. The 15 July aerosol layer is presented as the first signal of UTLS smoke that was lofted diabatically (and support that statement by declaring that it is "expected and predicted by the simulations (Fig. 7) as a consequence of the ascent rate profile with the minimum at the tropopause."). They do not identify any new "fire areas" that are upstream of this 15 July CALIOP observation. It is physically impossible to connect a 15 July aerosol observation to a smoke buildup that begins later. So O22 either failed to introduce a new fire area and smoke buildup prior to 15 July or they have made an illogical source-receptor connection. If there were to be a new fire area and smoke source, it would have to have been in place around the start of July in some unspecified burning area. This new source would make that of O21 (Siberia, late July into August) irrelevant or at most a secondary contributor.

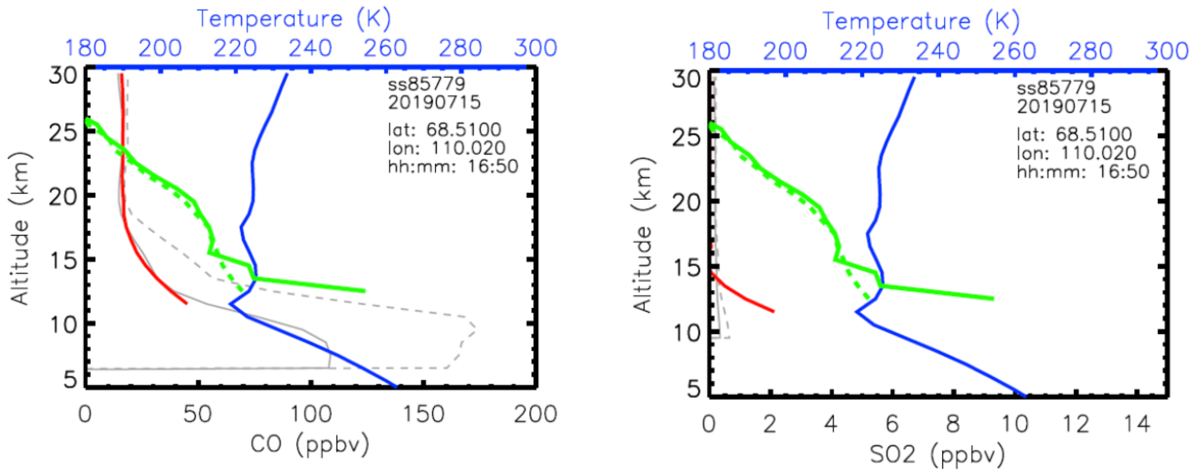
There is strong, independent evidence that the diffuse 15 July CALIOP layer O22 interpret as smoke (Figure 14a) is Raikoke sulfate. There are two coincidences with ACE-FTS and Imager occultations straddling the CALIPSO orbit. As shown below, the tropopause-level diffuse aerosol layer O22 attribute to non-pyroCb smoke is accompanied by SO_2 enhancement yet no CO enhancement. This is of course more supportive of the layer being a Raikoke sulfate layer than smoke. Hence this 15 July CALIOP curtain, showing a widespread high-latitude tropopause-level sulfate plume actually offers a rebuttal to the O22 argument that this, and other similar looking later layers, were smoke. It will be shown later that CALIOP/ACE coincidences between 20-26 July all show support for volcanic material over biomass burning aerosol.

ACE figure caption: Two panels. Each shows Imager 1 µm total extinction (green) and temperature (blue). Background extinction is also plotted (green dashed line), calculated as the average of May 2019 data north of 40°N. Left panel shows FTS SO2, right panel shows FTS CO. Each is plotted in red. Background average and avg. + 3-sigma are gray solid and dashed lines, respectively. Note: Extinction abscissa is not shown, to minimize clutter. Extinction is plotted on a log scale between 5e-5 and 5e-2/km. Note: CO and SO2 are plotted on a linear scale. SO2 background average and 3-sigma hover close to the x-scale origin but both are visible. Annotation gives occultation ID, date, time, latitude, longitude.

CALIOP/ACE figure caption: O22 Fig. 14a extended to cover Siberia, with ACE SO2 profile overlain. Vertical red arrow shows ACE latitude.

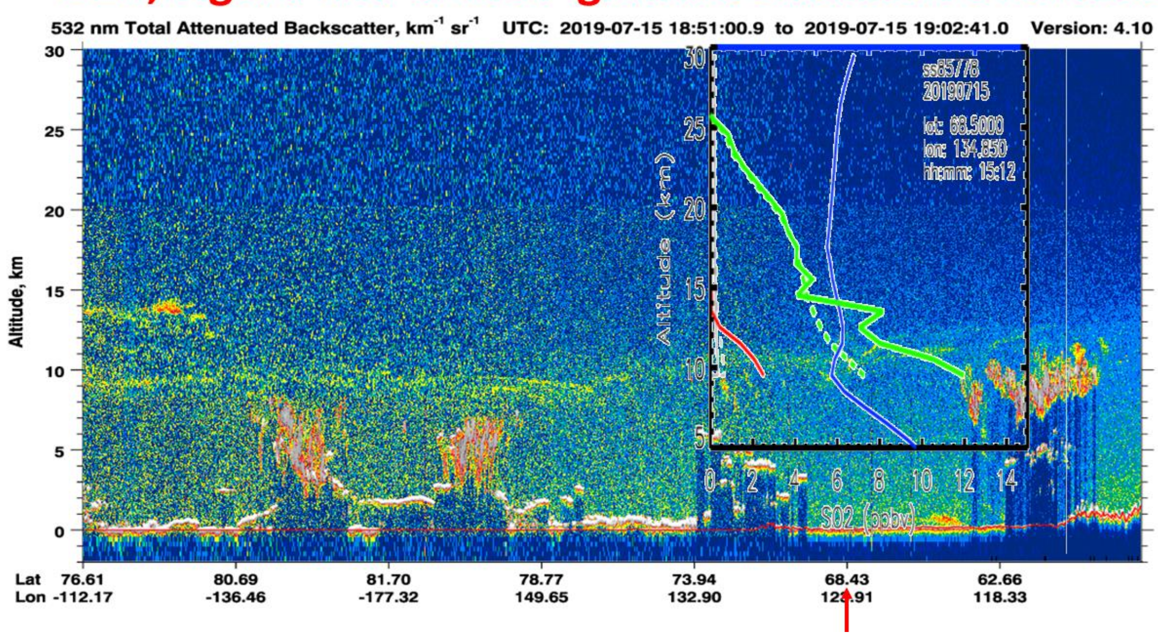


ACE occultation east of the CALIOP orbit.

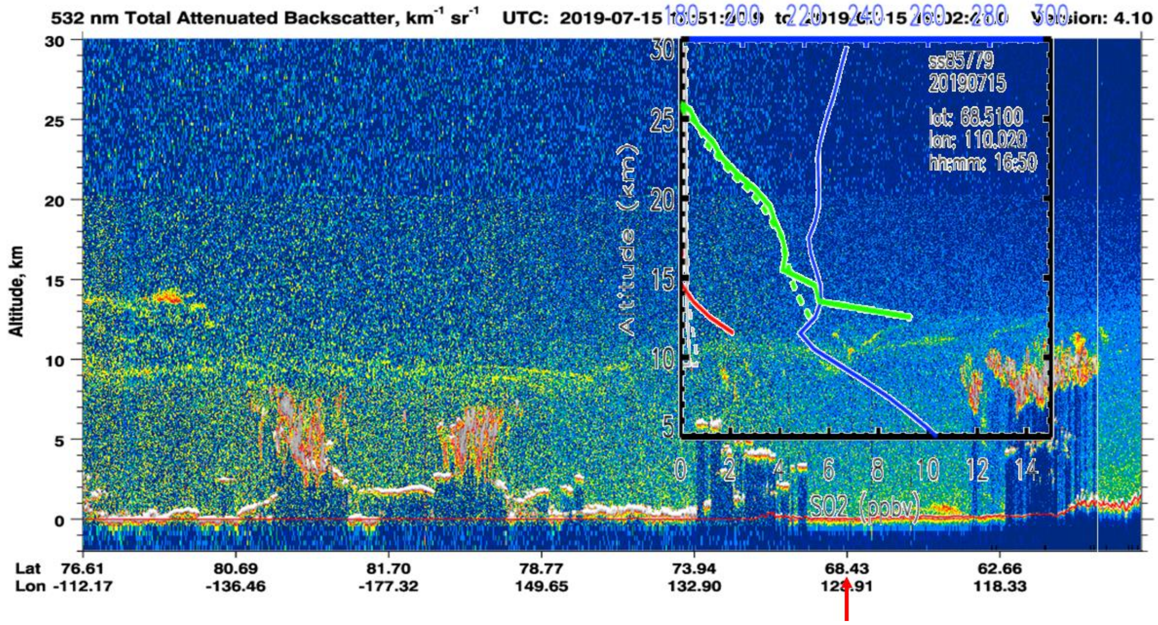


ACE occultation west of the CALIOP orbit.

O22, Figure 14a CALIOP granule extended over Siberia



O22, Figure 14a CALIOP granule extended over Siberia



As mentioned above, the 15 July onset of the tropopause-level smoke "footprint" is the centerpiece of their revised line of argumentation in support of the tropospheric self-lofting scenario. By itself it renders this section as thoroughly unconvincing. But O22 introduce two other new lines of argument that are equally weak. These will be covered in more detail below. Given that this is their second attempt to tease out tropospheric self-lofting observations, the essential importance of it to O22's overarching claim, and that Raikoke sulfates provide an alternative to smoke even for these stratospheric entry-level aerosols, this should be viewed as a closed case, in my assessment. The authors are encouraged to refute this conclusion or defend the new material in Section 5.

Figure 7b and discussion thereof:
Meteorologically I do not understand how the vertical gradient of potential temperature leads to a local minimum of lifting rate at the tropopause. Potential temperature increases monotonically throughout the tropopause and lower stratosphere. The naturally positive gradient is weak in the well mixed troposphere and larger in the stable stratosphere. The tropopause manifests as the transition from small to large positive gradient. What is it about the potential temperature gradient change that leads to the local minimum in lifting rate? Some more explanation would be beneficial.

Abstract, Lines 4-6, "The main goal of the study is to demonstrate that radiative heating of intense smoke plumes is capable of lofting them from the lower and middle free troposphere (injection heights) up to the tropopause without the need of pyrocumulonimbus (pyroCb) convection.: This has already been accomplished by Boers et al. (2010), who prescribed similar conditions involving super strong smoke AOT and little or no diffusion over several days. For this work to represent new information it would have to show observations in support of simulations like Boers et al. or this one. O22 state in the body of this work that this is essentially "impossible." Hence, a demonstration (beyond modeling) has not been shown. How does this affect O22's main goal?

Line 209-210, "…can complete the aging process and as a result get compact and spherical in shape. This manifestation of smoke aging was hypothesized by O21. Here it is taken as a given. In my first review I pointed out that several pubs showed aged tropospheric smoke retaining depolarization ratios outside the realm of pure spheres. O22 did not dispute the papers I cited. However, they did acknowledge that small departures from a perfect sphere will introduce a "significant jump" in depolarization. Consequently, the previously published reports of aspherical aged tropospheric smoke must either be disputed or else the O21 aged, pure spherical smoke hypothesis remains in dispute.

Line 220-222, "All observed pyroCb-related stratospheric smoke plumes, without any exception, show a high particle linear depolarization up to 0.2 at 532 nm…": This is incorrect. Siebert et al. (2000) and Fromm et al. (2008) show, for two major separate pyroCb events, smoke depolarization ratio that is in the aspherical regime but much less than the "large" values in the cited papers. Hence, even for undisputed pyroCb plumes, the depolarization ratio spans values from small to large. These publications should be mentioned along with the others and the implications discussed.

Line 253-254, "Only spherical particles are able to produce these rather low particle depolarization ratios of 0.02-0.03 as measured in the stratosphere in the summer 2019.": As acknowledged herein, O22 point out Raikoke sulfate observations using CALIOP and associate them with these near-zero depolarization ratios. From Line 521-523, "From end of June to mid July the number of spot-like layers with strong backscattering increased. Besides smoke layers, more and more volcanic sulfate plumes (indicated by a low depolarization ratio) appeared…" So in this regard, the authors have affirmed that sulfate typing can be inferred from depolarization ratio in isolation. Doesn't this complicate the CALIOP analysis performed herein?

Line 257-259, "A compact overview of the microphysical, chemical, optical and cloud-relevant properties of tropospheric and stratospheric smoke and changes of these properties during the aging process can be found in Ansmann et al. (2021b, 2022).": Fiebig et al. (2002; https://doi.org/10.1029/2000JD000192) conclude that the 9-day old free tropospheric smoke over Lindenberg in August 1998 was nonspherical based on lidar depolarization ratios between 6-11%. This is yet another published example of aged non-pyroCb smoke that has larger than spherical depolarization ratios. The authors are encouraged to include this reference and discuss the wider implications on their conclusions.

Line 527-529, "Very low wind speeds and weak horizontal air mass transport (stagnant conditions) favored the accumulation of smoke, the evolution of high AOTs on a regional scale, and thus self-lofting effects.": Indeed O21 showed AOT ramping up after 20 July. The AOT peak occurred on or about 26 July. Any "self-lofting effects" like accumulation of tropopause-level smoke would not begin until at least a few days after this AOT ramp-up, according to arguments made in this paper. Here O22 clearly stake their following arguments on the Siberia smoke build-up established by O21. This is problematic when considering the analysis that follows this statement. The authors should address this apparent problem.

Line 530, “On 15 July (Fig. 14a),…”: This is 5-10 days before the Siberia smoke started increasing. O22 say this smoke is downwind of the main fire areas, but there are no main fire areas until later in July. Moreover, the 9-10 km layer stretches from east longitudes to west longitudes, as far from Siberia as Hudson Bay. To state that this diffuse layer is downstream of Siberia is a stretch is seemingly in defiance of logic. Please explain.

Line 531-532, “This layer in the 8-10 km height range (not visible in the CALIOP data before 15 July) was…”: Here O22 clearly establish 15 July as the onset of their hypothesized, post-tropospheric-lofting smoke condition. It implies that something important started some days before. If so, there was no evidence presented showing that the Siberia fires were in action before 15 July. Neither did they introduce an earlier tropospheric smoke build-up anywhere in the northern latitudes. Please explain what precursor conditions existed, if any.

Line 532-533, "…well distinguishable from the plume-like pyroCb-related smoke layers and volcanic sulfate plumes at 13-15 km height. ": Which ones are smoke? Sulfate? All of them have nil depolarization ratio.
https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_detail.php?s=expedited&v=V3-30&browse_date=2019-07-15&orbit_time=18-00-00&page=3&granule_name=CAL_LID_L1_Exp-Prov-V3-40.2019-07-15T18-00-00Z.hdf
Note that AOT in the strongest plugs (gray backscatter) exceeds 0.3. Depolarization ratio there is nil. So, if these are Raikoke sulfates, O22 have shown that the volcanic sulfates have native AOT far exceeding 0.025.

Line 536-537, "The occurrence of such a diffuse layer around the tropopause was expected and predicted by the simulations (Fig. 7) as a consequence of the ascent rate profile with the minimum at the tropopause.": If this is expected and predicted by the model, then why did this layer just show up on 15 July at the tropopause? If it was the result of slow lofting in stagnant conditions, one would find this layer on earlier days at lower altitudes. Moreover, one would be able to trace it downward to very intense smoke layers. And that would have to have been earlier in July, when there was no reported "main fires" or smoke buildup. Please explain.

Line 548, "Shortly after the 26 July, Xian et al. (2022) report a strong increase of aerosol pollution over the High Arctic.": Xian et al. do not present any such data in 2019. Their case study is for August 2021. Please explain or remove this statement.

Line 548-550, "The area mean 550 nm AOT for the Arctic region from 70°-90°N increased from long-term mean values of 0.14 before 28-29 July 2019 to the record-breaking value of 0.4 on 10 August 2019. Never before such a High Arctic mean AOT was observed the authors [Xian et al.] **stated.":** Xian et al. do not present any data for 10 August for any year. I could find no place in that paper where they made a claim about any 10 August AOT being the largest ever recorded. How can the source for record breaking AOT in 2019 come from fires in 2021? This appears to be a misattribution of Xian et al. Please explain.

Line 551, "The source for the record-breaking Arctic aerosol can only be the Siberian fires in July and August

2019.": This is clearly at odds with their analysis of and importance ascribed to the CALIOP diffuse layer on 15 July. This line of reasoning is therefore problematic and needs to be revised.

Line 555-557, "Under these conditions with very large AOT values over extended Siberian and Arctic terrain one can assume that there were several subregions with AOTs>1.5 over days if not for more than a week so that the probability for self-lofting events was high in July and August 2019.": Here O22 unambiguously describe the conditions that are favorable for eventual lofting of smoke to the UT. It involves days of super large AOT in the lower troposphere. O21 claimed that these conditions ensued after ~21 July. So how can any of the CALIOP layers they discuss on 15, 25, and 26 July be the result of this mechanism? At the very least O22 are encouraged to abandon the O21 source term and find another high AOT event before 15 July (their first day of tropopause-level diffuse smoke attributed to this pathway).

Line 563-564, "For comparison, a Raikoke-related AOT of 0.025 was expected at 532 nm at high northern latitudes considering the emitted SO2 mass of around 1.5 Tg (Ohneiser et al., 2021).": AOT observations of Raikoke sulfates reported in this paper far exceed 0.025. This is at odds with O21. O21 did not argue the 0.025 limit based on SO2-sulfate conversion calculations, but rather other observations such as Kloss et al. As previously discussed in prior reviews, Kloss et al.'s AOT values were biased low with respect to point measurements such as those from lidar.

Line 576-577, "Such a high lidar ratio has never been observed for volcanic sulfate aerosol.": Perhaps until now. Given that O22 have perhaps inadvertently demonstrated the omni-presence of Raikoke sulfates, with small and large AOT, from the tropopause to lower stratosphere, days to weeks before smoke could have entered the UTLS in abundance, it may be reasonable to conclude that indeed some sulfates may have the optical properties that O22 relegate to smoke presence. Please comment on this and/or make suitable revisions.

Line 577-578, "The particle depolarization ratios were <0.03 at both wavelengths (as given in Fig. 15), a clear signature of perfect spherical particles, …": Agreed. And perfect spherical particles are not the norm for tropospheric smoke of this age. See my prior review and comments above. Please explain how the previously published reports of aged tropospheric and pyroCb smoke with depolarization ratios ~0.03-0.11 fit into O22's interpretation.

Line 579-580, "In cases of pyroCb-aided lofting, the depolarization ratios were always observed to be >0.1 during the first month after the pyroCb events.": This was not the case for Norman Wells (Siebert et al., 2000) or Chisholm (Fromm et al. 2008). This disparity must be recognized, acknowledged, and explained.

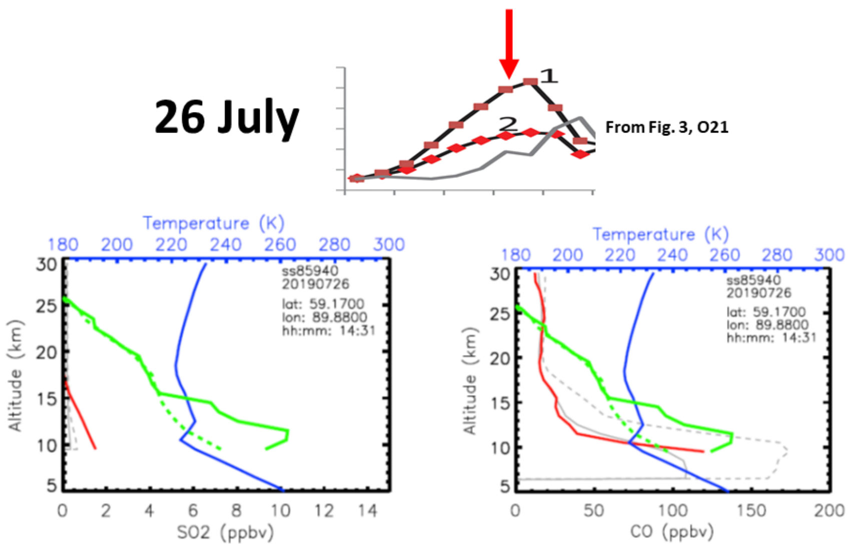
Line 585-586, "We therefore have our doubts that one can obtain a clear picture of the aerosol composition from infrared absorption spectra alone.": These doubts are well founded. It is prudent to doubt the full veracity of any composition determination based on any single remote sensing data item. That doubt applies equally to lidar backscatter, in this case the over reliance on spectral dependence of lidar ratio. Hence, doubts should be spread equally and the authors are advised to consider that.

Perhaps more importantly, Boone et al. (2022) did not rely solely on the IR spectra. A full interpretation of that paper must take into account the associated ACE Imager aerosol extinction and ACE-FTS SO2 and HCN measurements. These orthogonal indicators were all presented together by Boone et al., leading to their robust conclusion of sulfate dominance over smoke.

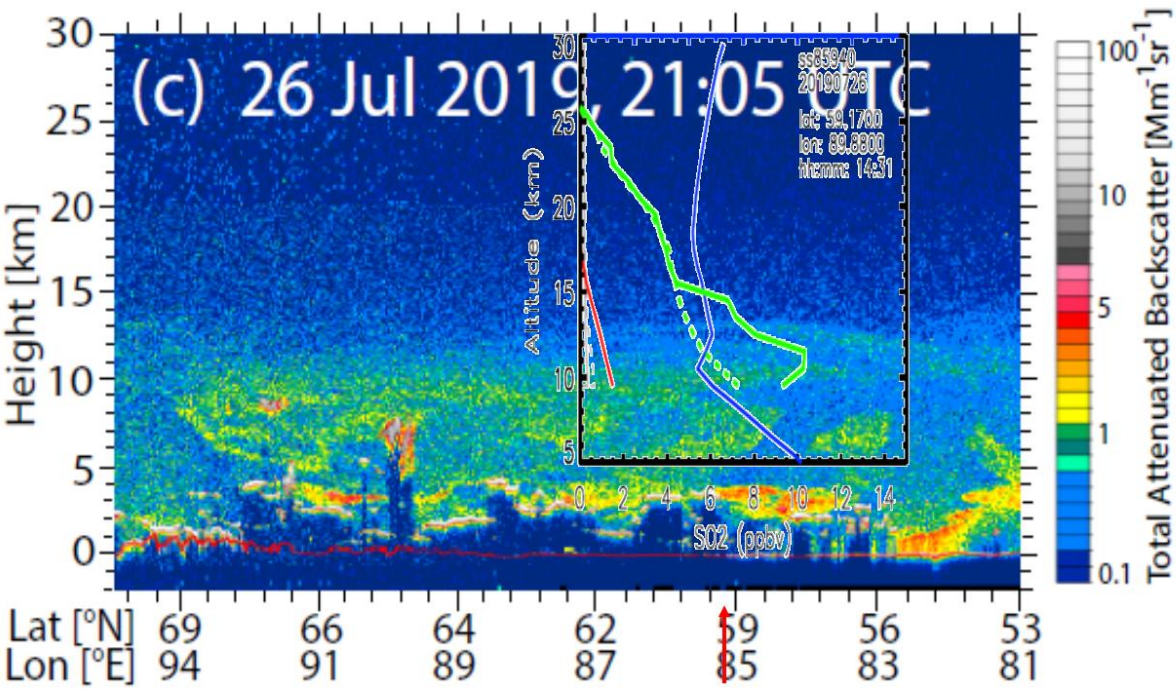
Line 587-593, discussion of AIRS CO: It is essential for O22 to show these results such that they can be evaluated and reproduced. Their claim here is brand new and of fundamental substance. The AIRS averaging kernels are such that the CO signal peaks between 300-600 hPa, so a strong signal at 100 hPa cannot be divorced from the total column amount. Moreover, a check that I performed on Siberia/Arctic AIRS August monthly CO showed no obvious enhancement at 100 hPa. More reason for O22 to fully lay out this analysis.

For additional benefit to the authors I append below an analysis similar to that shown above, combining CALIOP 532 nm attenuated backscatter coefficient and ACE data for daily coincidences over Siberia between 20-26 July 2019. Interpretations are given with each set of ACE plots and a wrap-up discussion follows all. In a nutshell, the

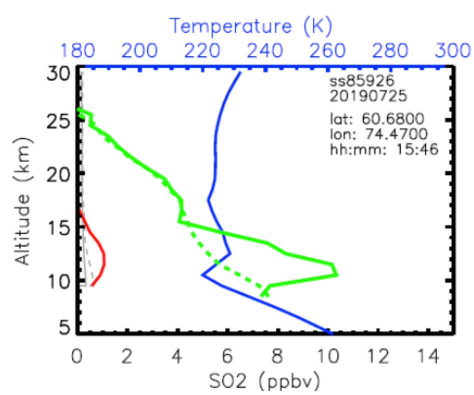
ACE data show SO2 enhancements with each coincidence and an absence of CO enhancement within any aerosol layer. Hence, over Siberia at the critical time of hypothesized self-lofting, all UTLS aerosols are combined with sulfur enhancement. One can compare the 25 and 26 July examples with O22 Fig. 14.

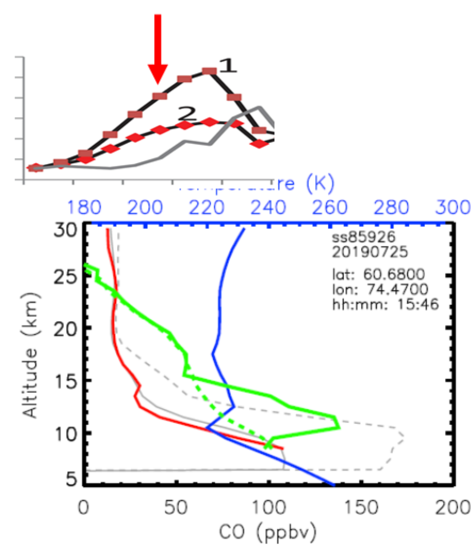


Aerosol and SO2 together. No CO enhancement in layer.

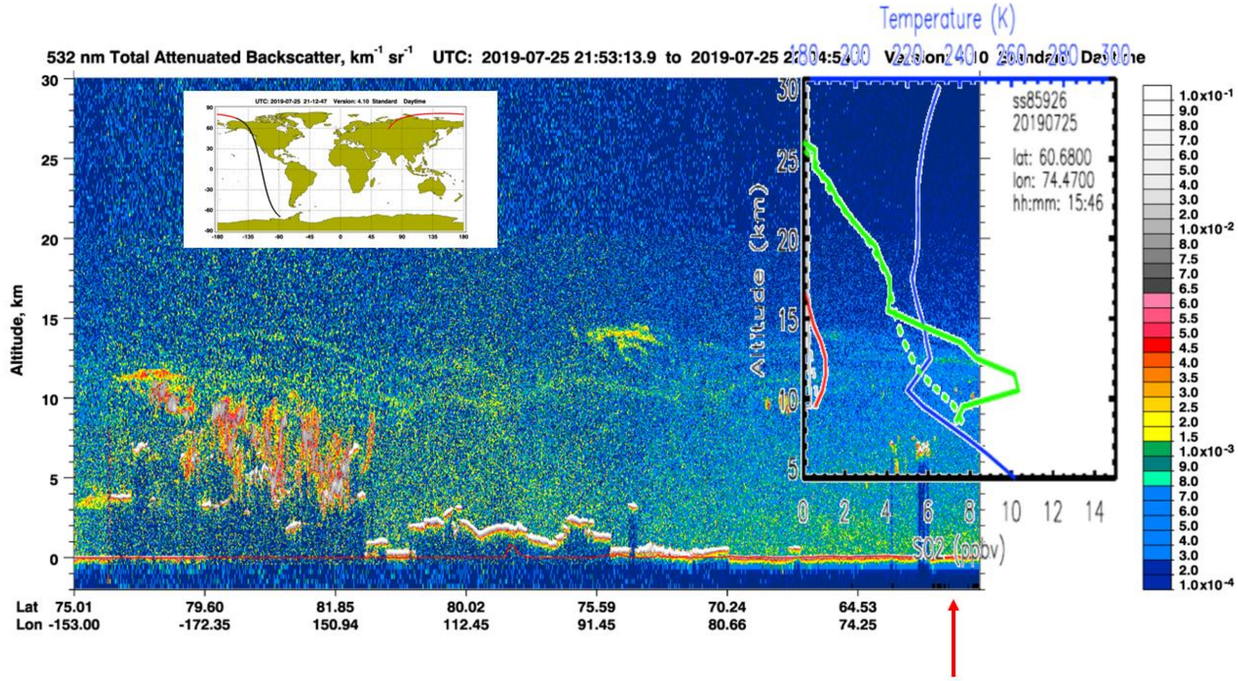


25 July

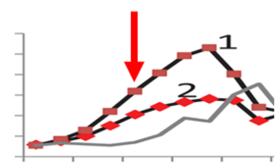


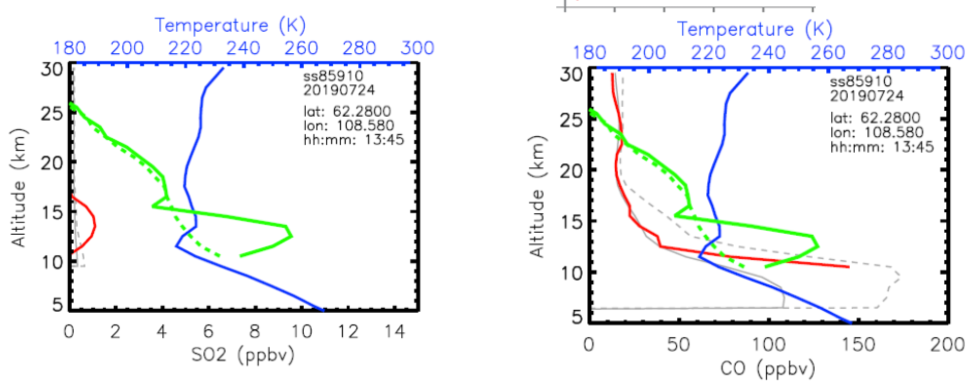


Aerosol and SO2 together. No CO enhancement in layer.

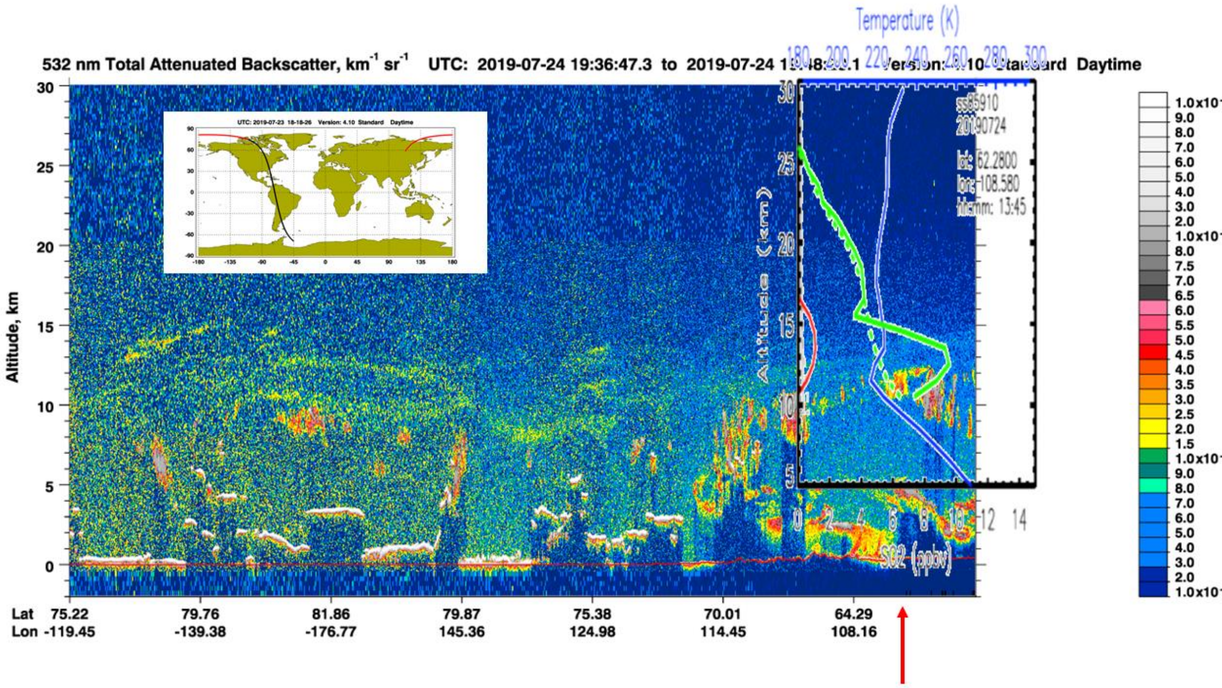


24 July

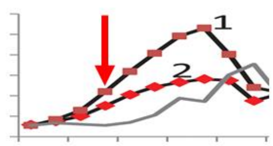


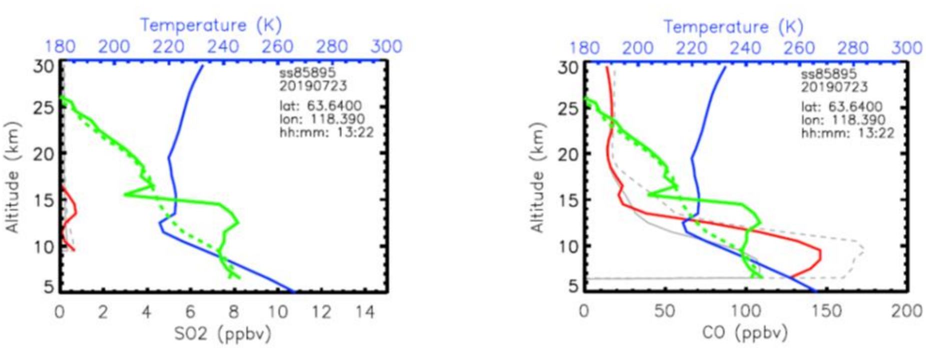


Aerosol and SO2 together. CO enhancement at bottom of layer.

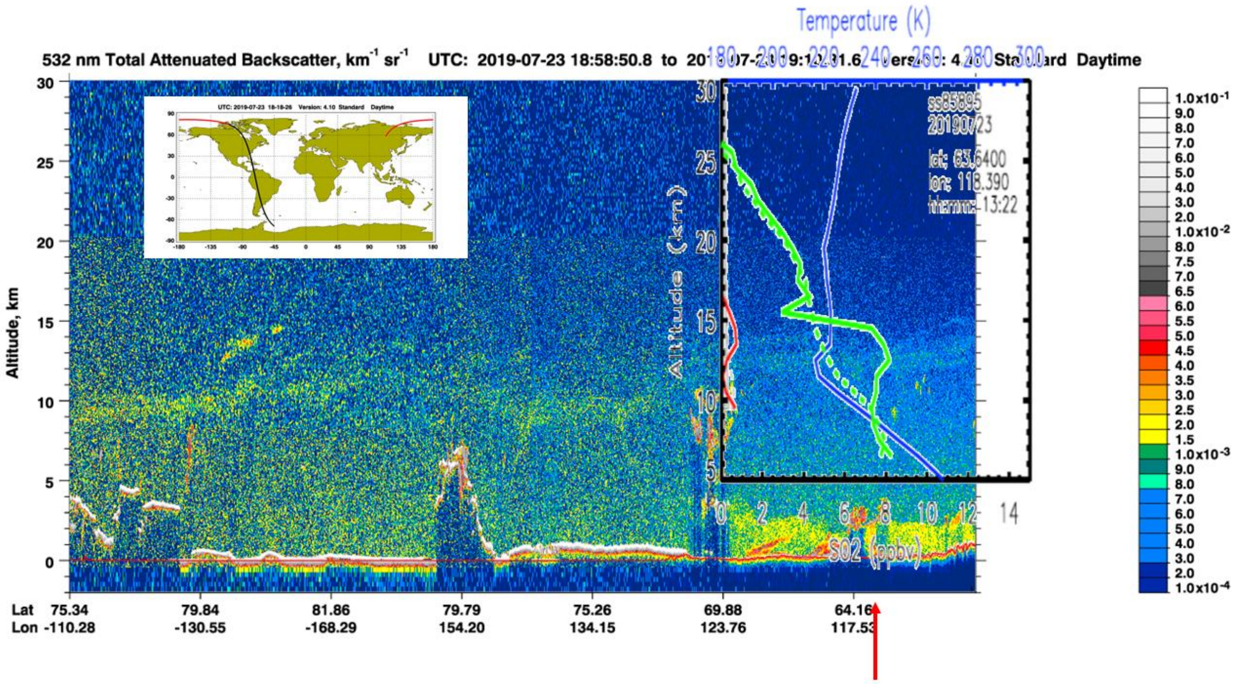


23 July

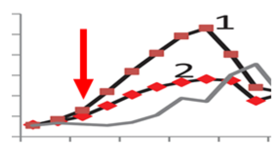


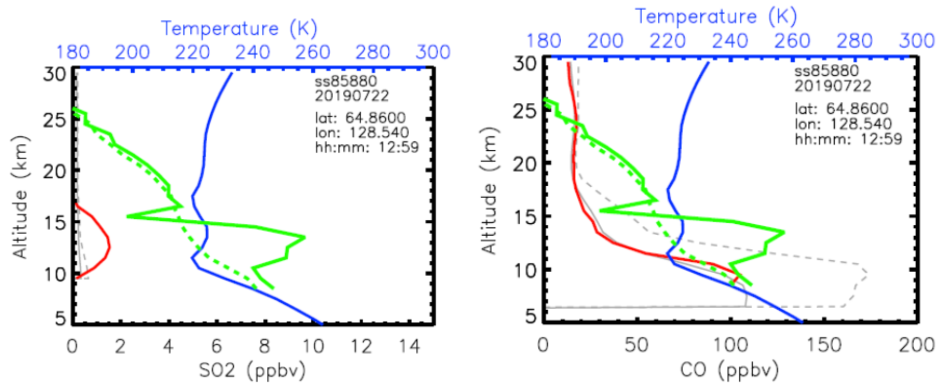


Aerosol and SO2 together. CO enhancement below main SO2 bump .

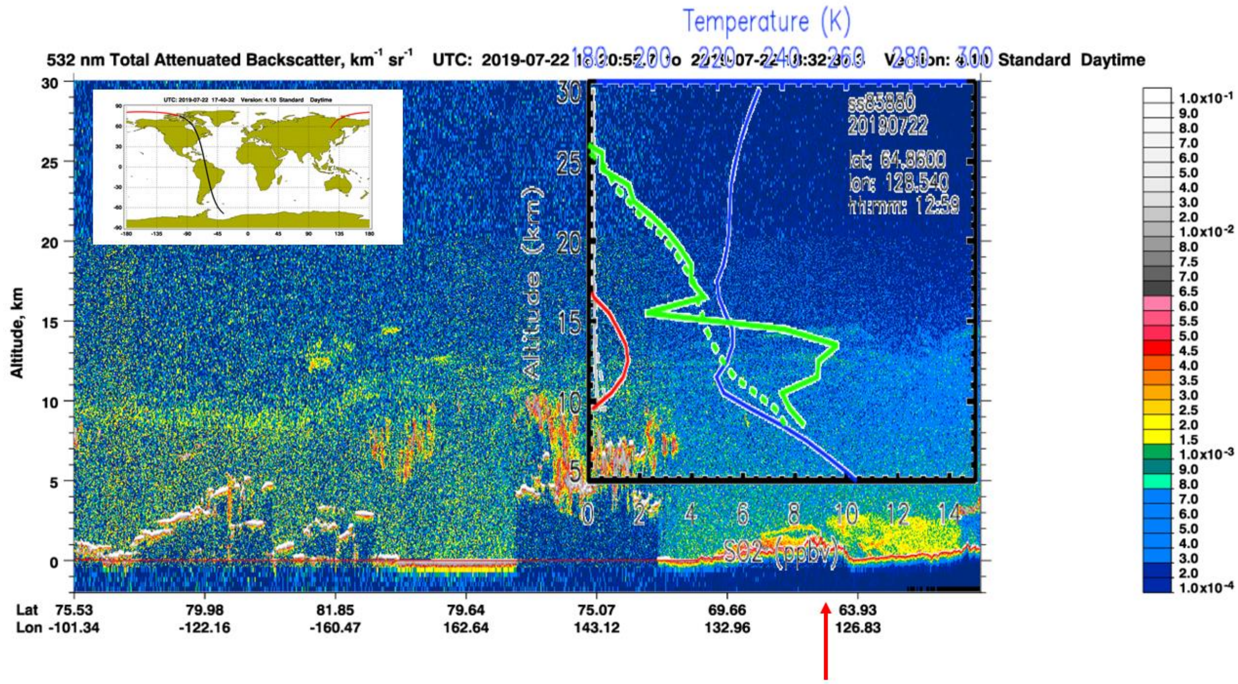


22 July

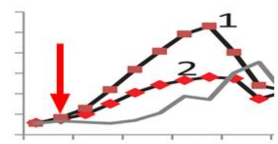


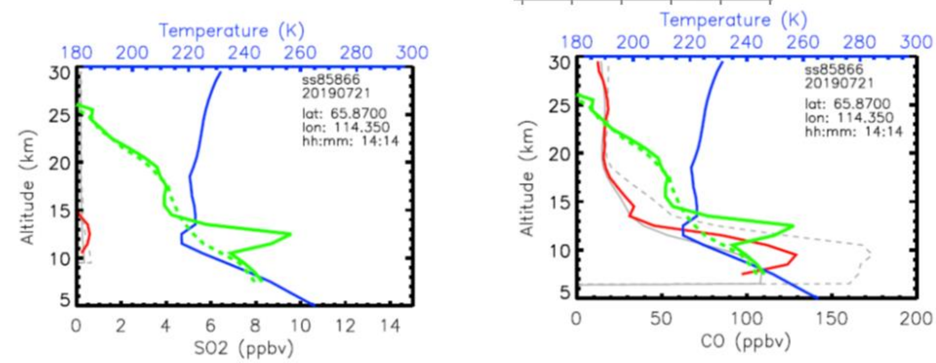


Aerosol and SO2 together. No CO enhancement in layer.

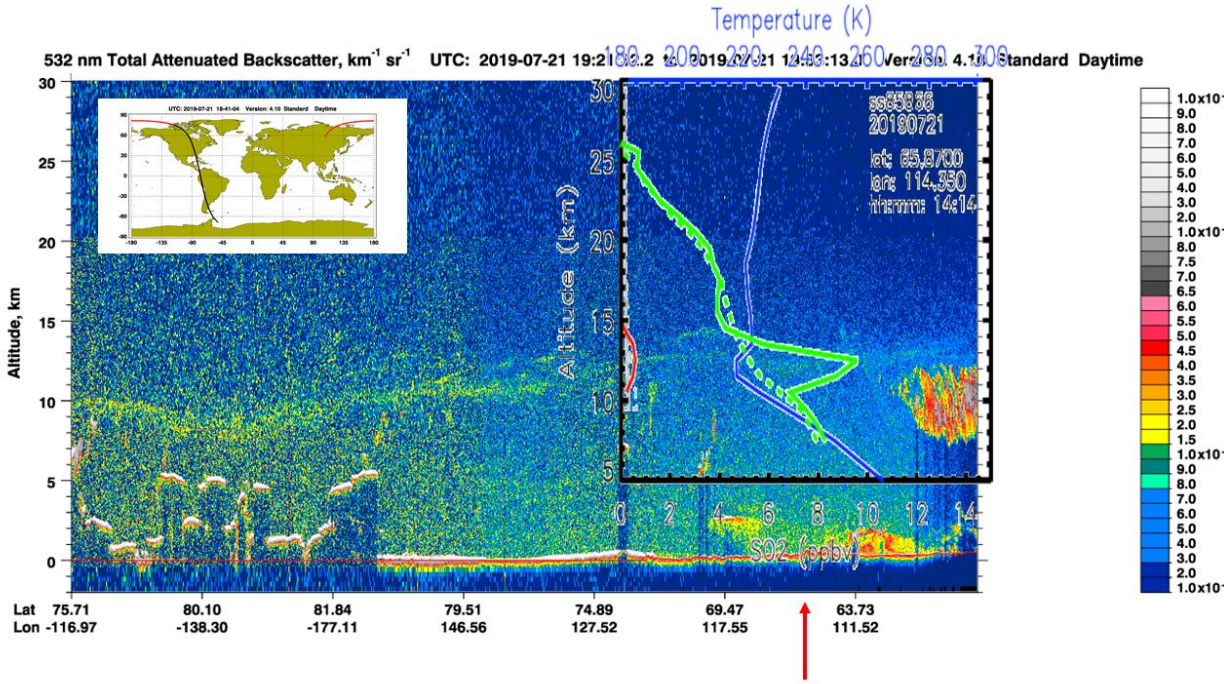


21 July

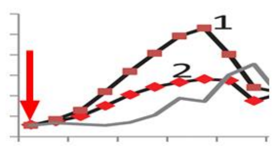


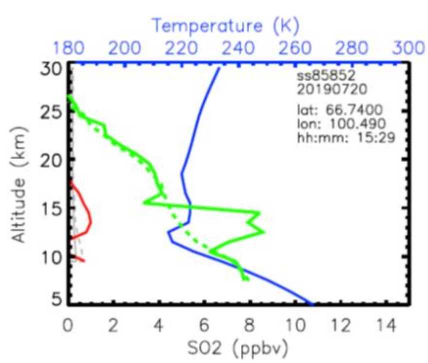


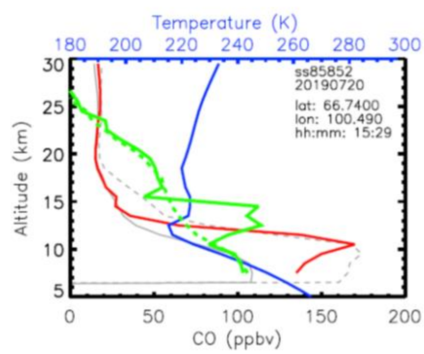
Aerosol and SO2 together. CO enhancement near bottom of and below layer.



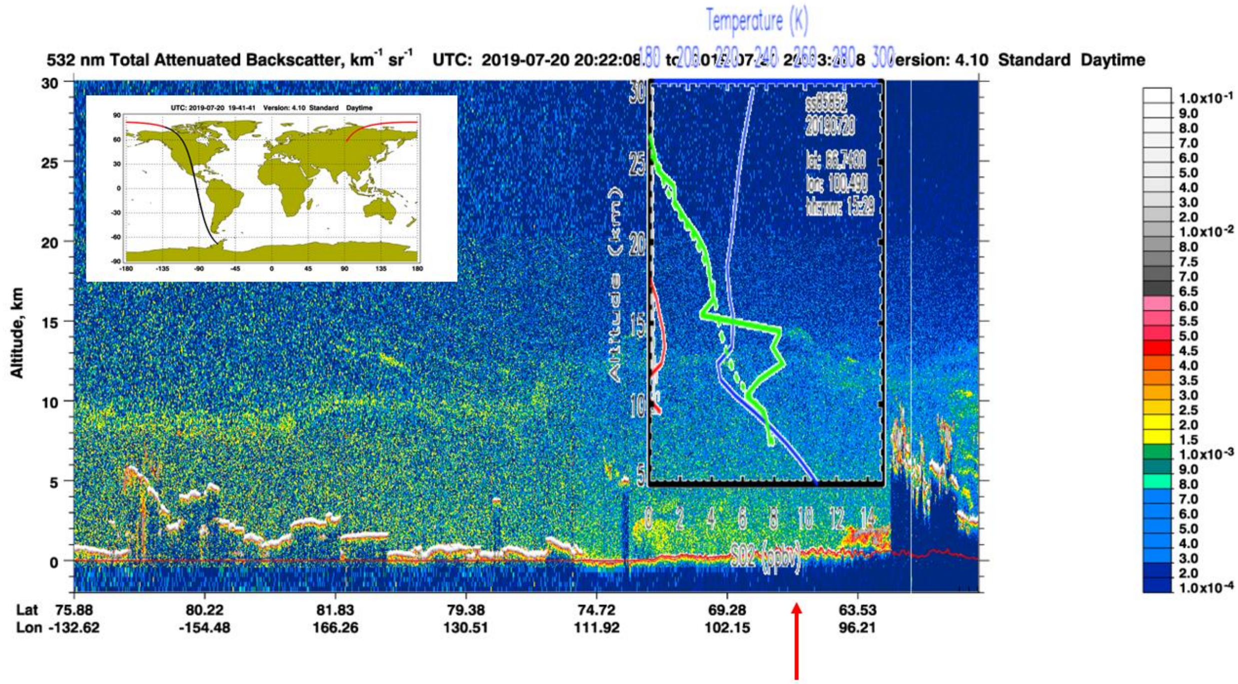
20 July







Aerosol and SO2 together. 3-sigma CO enhancement near bottom of and below layer.



Synthesis:

UTLS diffuse backscatter enhancements each and every day between 20-26 July over Siberia.

In every example, ACE SO2 was enhanced in the diffuse backscatter layer.

Excellent correspondence between ACE Imager aerosol enhancement and CALIOP backscatter.

On a couple occasions there was a CO enhancement in the lowermost portion of the most prominent ACE layer. These UT enhancements were at some points associated with no discernable Imager extinction enhancement.

There were no CO enhancements in the prime diffuse backscatter layers.

In the AOD onset period (20-22 July) there was no justification for locally lofted smoke to the UT. Hence, the UT CO enhancements were likely aged air from another source.

Conclusion: Raikoke sulfates clearly overwhelm any other explanation for UTLS aerosols over Siberia when the diabatic lofting of smoke was hypothesized to have started. The relatively strong but diffuse CALIOP backscatter is demonstrably sulfate in all the examples presented.