Ohneiser et al. (2021), hereafter abbreviated as "O21," is a provocative study of upper troposphere, lower stratosphere (UTLS) aerosols at high northern latitude between fall 2019 and spring 2020. It is provocative in that O21 observe UTLS aerosol nearly daily in that temporal span and conclude that the composition is wildfire smoke, the source is fires in a sector of Siberia in July 2019, and the transport pathway to the UTLS is diabatic heating/lofting. The manuscript is also provocative in that Raikoke volcano (Kuril Islands) erupted in late June 2019 and polluted the UTLS with a mass of SO₂ (https://twitter.com/simoncarn/status/

1142713198480482304) on par with or exceeding other eruptions that generated stratospheric clouds persisting for greater than a half year (E.g. Kasatochi, Sarychev Peak, Nabro, Kelut, Calbuco. See Solomon et al. (2016) for a tabulation.).

If O21's conclusions are borne out, it will be a new insight into the polar UTLS and smoke transport to the UTLS. However, there is overwhelming observational evidence that Arctic UTLS in the second half of 2019 and early months of 2020 was blanketed by Raikoke sulfates. Secondly, there is abundant evidence that the UTLS aerosol picture O21 describe over the July Siberia source sector is also dominated by Raikoke SO₂ and sulfates. Thirdly, the lidar data presented by O21 are more closely aligned with spherical sulfate droplets than smoke particles. These three points are elaborated on below.

Point 1

Kloss et al. (2021) show that the Raikoke volcanic cloud dominated the high-latitude northern hemisphere from eruption through the early months of 2020. I consulted Chris Boone, ACE-FTS Project Scientist and co-Principal Investigator, to help qualify the 2019/20 UTLS plume further. ACE not only delivers aerosol extinction profiles but profiles of SO₂ as well. These were combined by Cameron et al. (2021) in an examination of several UTLS volcanic events including Raikoke; the results further qualify those of Kloss et al. and show the strong presence of Raikoke SO₂ and sulfates at high northern latitude in summer and fall 2019. In addition, ACE IR spectra have been used to identify smoke aerosol in connection with ACE Imager extinction profiles (Boone et al. (2020)). The same technique was applied to high-latitude northern hemisphere 2019/20 ACE data while invoking published sulfate IR spectra for comparison. The findings are summarized as follows. In July 2019, the lower stratosphere in the latitude region near 60 degrees north is stuffed with sulfate aerosols from the Raikoke eruption. Identification of the composition is accomplished by looking at the infrared spectrum of the aerosols and noting the coincident enhancement of SO₂ and ACE Imager aerosol extinction layers. In September/October 2019, there is a blanket of aerosols in the lower stratosphere in the latitude region near 80 degrees north. The blanket appears to be composed of Raikoke sulfate aerosols. In February/March 2020, the aerosol blanket in the lower stratosphere near 80°N is still present. SO_2 has decayed. However, the spectra associated with the Imager aerosol layers

are consistent with sulfate. At no point did we find any evidence of biomass burning smoke playing a role in these stratospheric aerosols (Boone et al., 2020). Detailed support for these findings is available upon request.

Point 2

O21 hypothesize that their MOSAiC Arctic 2019/20 lidar signals are dominated by an impressive build-up of UTLS smoke that began between ~24-28 July 2019 in a zone within Siberia centered roughly at 60°N, 110°E (Figure 2). They present nadir satellite imagery combining fire hot-spot data, true-color imagery, and aerosol optical depth retrievals to show an intensification of burning and smoke concentration. They also display a single CALIOP curtain on 26 July to characterize the vertical structure and ascent of smoke layers (Figure 3). There are several concerns regarding Figures 2, 3 and their interpretation, listed below.

It is simply not possible to know from a single CALIOP curtain if ascent is taking place; there is insufficient information from such a quasi-instantaneous vertical snapshot. Additional information must be brought to bear.

There are technical issues with Figure 3 and its caption. The date of the CALIOP image is given as "2016." This is obviously an inconsequential typographical error. But more substantially, the latitude, longitude coordinates labeled along the bottom are wrong. The curtain displayed is actually situated west of the Figure 2 boxed focal point. It is west of the labeled coordinates by approximately 25° longitude. Compare Figure 3 with the actual orbital track and lidar data here:

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse date=2019-07-26&orbit time=21-04-20&page=1&granule name=CAL LID L1-Standard-V4-10.2019-07-26T21-04-20ZN.hdf

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse_date=2019-07-26&orbit_time=20-12-10&page=4&granule_name=CAL_LID_L1-Standard-V4-10.2019-07-26T20-12-10ZD.hdf

This is scientifically relevant for two reasons. One is that the vertical aerosol profile over the Siberia focus zone is unknown to the reader. Secondly, the aerosol profile on display is nominally upwind of the Siberia box, meaning that the history of those aerosols may be disconnected with processes occurring in the Siberia box.

Indeed, it appears from the CALIOP curtains linked above that the "Ascending smoke plumes (mostly in green)

are visible up to the tropopause at 10-11 km height as well as in the lower stratosphere..." [Quoted from Figure 3 caption] are mostly assigned as sulfate by the CALIOP version 4 aerosol subtype algorithm.

One can see from CALIOP curtains just upwind of the Figure 2 Siberia box, on days leading up to the O21 smoke AOD buildup, an assignment of sulfate subtype to "mostly in green" backscatter in the UTLS. Here is an example from 20 July:

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse date=2019-07-20&orbit time=20-33-51&page=1&granule name=CAL LID L1-Standard-V4-10.2019-07-20T20-33-51ZN.hdf

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse date=2019-07-20&orbit time=19-41-41&page=4&granule name=CAL LID L1-Standard-V4-10.2019-07-20T19-41-41ZD.hdf

Similar scenes are found each day thereafter leading up to the 24-28 July O21 period of focus (<u>https://www-calipso.larc.nasa.gov/products/lidar/browse_images/std_v4_index.php?d=2019</u>) This multi-day, broad, high-latitude swath of aerosols primarily defined as "sulfate/other" conforms to the findings of Cameron et al. and our deeper investigation into ACE July 2019 profiles near 60°N.

It stands to reason then, that if Raikoke sulfates are blanketing high northern latitudes at that time (Kloss et al., Cameron et al., and our investigation), they would also be evident over the Figure 2 Siberia box before, during, and after the hypothesized smoke uplift. The CALIOP example below shows aerosol subtype findings consistent with the prior examples on 20 July over the Figure 2 box:

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse date=2019-07-20&orbit time=18-55-21&page=1&granule name=CAL LID L1-Standard-V4-10.2019-07-20T18-55-21ZN.hdf

https://www-

calipso.larc.nasa.gov/products/lidar/browse images/show v4 detail.php?s=production&v=V4-10&browse date=2019-07-20&orbit time=18-03-11&page=4&granule name=CAL LID L1-Standard-V4-10.2019-07-20T18-03-11ZD.hdf

Hence, if the July 2019 Siberia-zone smoke is the initial condition for the O21 lidar observations in fall and winter, the presentation surrounding Figure 2 and 3 is insufficient to make that case.

Point 3

O21 acknowledge that the stratospheric aerosols detected by MOSAiC lidars were spherical according to aerosol depolarization measurements (Page 7, line 21). In principle, there seems to be less uncertainty as to the shape, composition, and depolarization of volcanic liquid sulfates in contrast to biomass burning smoke. Lidar measurements of tropospheric and stratospheric smoke converge on the idea that smoke is depolarizing (e.g. Burton et al., 2015; Fromm et al., 2008), suggesting some amount of asphericity. The first reported lidar observations of stratospheric smoke emphasized the unmistakable signal of depolarization (Siebert et al., 2000; Fromm et al., 2000). More recent stratospheric smoke papers, cited within O21, consistently report depolarization by smoke particles. Hence O21 would be establishing a new finding--stratospheric smoke with essentially no aerosol depolarization. Arguing for this peculiarity, O21 mention an aging process and a collapse of black carbon core. Neither process is described, and no publications are cited. Given the weight of evidence for the overarching presence of Raikoke UTLS liquid sulfates during the O21 reporting period and the MOSAiC depolarization results provided therein, the arguments for particle aging and black carbon coreshell collapse must be made more substantively.

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