

In this note I offer comments limited to Section 3 and 5 regarding the evidence for mixing of volcanic sulfuric acid with smoke, for the authors to consider. This manuscript makes an effort to suggest possible effects of mixing volcanic H₂SO₄ with smoke and thereby creating a “new black” particle linear depolarization ratio (PLDR). The example given is a volcanic SO₂ plume on 8 August 2017. In looking at the figures and text I see some apparent inconsistencies and possible misinterpretation.

REPLY: We thank very much Dr. Fromm for his careful reading, comments and suggestions, which we address in the following. We agree that at the moment the discussion on the possible physical explanation for the increased smoke PLDR is not complete. Thus we decided to remove this section from the revised manuscript. More efforts will be made to investigate this issue and re-assess the possible coexistence of smoke particles and particles of sulfuric nature and whether this could affect the former in such a way to form near-spherical particles.

Figure 15 shows a map that includes upper tropospheric SO₂, and attributes that to Shiveluch. (The bottom panel of Figure 15 is confusing in that it refers to Himawari-9 imagery but the analysis illustrated is something else.) Although an eruption on 8 August at Shiveluch was reported, it seems much more likely that the SO₂ came from a 7 August 2017 eruption of Bogoslof in the Aleutians. <https://www.adn.com/alaska-news/2017/08/07/alaskas-tiny-bogoslof-volcano-eruptsagain-sending-an-ash-cloud-miles-above-the-aleutians/> <https://volcano.si.edu/volcano.cfm?vn=311300>. It becomes apparent that the SO₂ derives from this eruption when one follows the OMPS SO₂ back in time. The plume starts on 7 Aug right near Bogoslof (credit NASA Worldview). Regardless of the source of the SO₂, the only evidence suggestive of volcanic influence is the map of 8 August SO₂ (Figure 15). The pyroconvection in Canada leading to the stratospheric smoke occurred 4 days later according to papers the authors cite. Presumably if volcanic sulfur was responsible for the months of double-digit PLDR “new black” one might expect to see a robust volcanic signature close to British Columbia on or much closer to 12 August. What can be shown in that regard?

REPLY: Thank you for bringing this information to our attention. Indeed, the two eruptions occurred one day apart. To determine whether the plume (which at the time we believed came from Shiveluch) was transported towards the area of British Columbia, we performed forward trajectory and dispersion analysis using the offline

coupled atmospheric and dispersion model FLEXPART-WRF (Brioude et al., 2013). Updated simulations with the FLEXPART showed that both plumes (from Shiveluch and Bogoslof) travelled towards the same direction, to the area of British Columbia. Nevertheless, we have deleted this part from our paper, for the reason mentioned above.

The paper refers to the 8 August CALIPSO measurement as "daytime" when it is in fact a night-time orbit segment. Consequently, the connection made between this CALIPSO measurement and the daytime 8 August MODIS image in Figure 4 is inaccurate. On a technical note, the red dashed lines in Figure 6 for the 15 August CALIPSO data are not where the text directs the reader: the 13 km smoke layer.

REPLY: Thank you for noticing this, we have corrected the figures.

The authors refer to a 12 August CALIPSO measurement (Page 6, line 23) but don't show any such measurement. It is apparent they meant 15 August but this needs to be clarified (if indeed they intend to show a 12 August measurement) or corrected.

REPLY: Thank you for noticing this, we have corrected it.

On page 6, line 16 the authors seem to state that between 8 and 15 August the stratospheric smoke plume had already blown to Europe: ". . .8 and 15 August 2017, when the smoke plume has already reached Europe" They do not present any data to support that and I believe there is no support for that claim. The leading edge of the plume on 15 August was still entirely over Canada.

REPLY: Thank you for this comment. Indeed, the plume was evident above Europe only after late August. We corrected this sentence in the manuscript.

In summary, I was confused by the material in Section 3 and 5 and thus was left unconvinced of any meaningful mingling of volcanic sulfates and pyroCb-injected stratospheric smoke. Presumably, if the transport pathway was what the authors claim—pyroconvection—the sulfates would have to have been in large concentration in the vicinity of the pyroCbs and in the inflow part of the atmosphere (i.e. lower troposphere). If the two mingled by virtue of UTLS sulfates in high concentration encountering the pyroCb outflow, one

might expect that the sulfates would be detectable leading up to the pyroCb injection. This might be an avenue for the authors to explore because there is good CALPSO coverage of the Canadian and upstream environments on all the days between 8 and 12 August. It is becoming increasingly evident that double-digit PLDR is quite common for stratospheric smoke. In personal communication with one of the coauthors, I discussed a similar phenomenon in northern summer 2014. Here is an example of double-digit PLDR of stratospheric smoke over Scandinavia at that time (credit: <http://lidar.ssec.wisc.edu/>) http://hsrl.ssec.wisc.edu/by_site/18/2014/08/17/am/#MF2HSRL The Black Saturday (Australia, February 2009) pyroCb stratospheric smoke also had double-digit PLDR.

Here is an example of week-old smoke at that time (Credit: NASA):

https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_detail.php?s=production&v=V4-10&browse_date=2009-02-15&orbit_time=12-52-14&page=3&granule_name=CAL_LID_L1-Standard-V4-10.2009-02-15T12-52-14ZN.hdf

It is unlikely, or at least un-established, that precursor volcanic activity occurred in these 2009 and 2014 cases. Hence it would seem that there is another common bond, albeit still unresolved, embodied in this growing record of anomalously large PLDR in dry stratospheric smoke environments.