We would like to thank Kevin Ohneiser for taking the time to read and provide comments on our paper. We believe that changes made to the paper based on his and the reviewers' comments have improved the manuscript.

We have addressed his comments (in black) below in red.

Response to Kevin Ohneiser

The manuscript by Wells et al. shows model simulations of the Raikoke volcanic eruption in 2019 and comparisons with space-borne observations. They found that volcanic sulfate particles alone are not sufficient in order to explain the high AOT values that were observed during summer, autumn and winter 2019-2020. Including ash into their UKESM1 simulations, however, enhances the agreement between model results and observations. The manuscript is an important contribution to the literature as it shows comparisons of Raikoke modelling results including ash in the simulations vs. observations. There are, however, some questions and comments to parts of the manuscript. Some of the concerns are listed as follows:

Question 1: Why should the Raikoke event regarding the produced stratospheric aerosol be so different compared to Pinatubo, Sarychev and many others.

After these eruptions, the ash was removed quite quickly (within a few weeks) and the sulfate was then left as the only volcanic aerosol type? And now, the ash was present even after months (September –November 2019)?

Thank you for your question. The role of ash in the dispersion of volcanic aerosol after an eruption has not only been studied for the Raikoke eruption but also for Pinatubo (e.g. Shallcross et al., 2021; Stenchikov et al., 2021; Abdelkader et al., 2023). Ground-based and airborne lidar observations (Browell et al., 1993; Vaughan et al., 1994) suggested the base of the Mount Pinatubo volcanic plume contained ash particles coated in sulphuric acid for approximately 9 months after the eruption. More recently, after the Mt. Kelud eruption, observations suggested that ash-rich aerosol accounted for part of the aerosol plume 3 months following the eruption (Vernier et al., 2016).

It is also quite possible that the Sarychev Peak eruption also contained a significant amount of volcanic ash. If you consider the stratospheric AOD shown in Figure 5 of Haywood et al. (2010), it becomes immediately clear that the observations show a far longer e-folding time than the model. It is quite possible that including ash in simulations of Sarychev Peak would have led to an improved agreement with observations from the OSIRIS limb-sounding instrument. Definitively attribution of any improvement would be difficult because the simulations of Haywood et al. (2010) were performed with an older version of the model

(HadGEM2-ES) which utilised a different aerosol scheme and many other different processes and parameterisations.
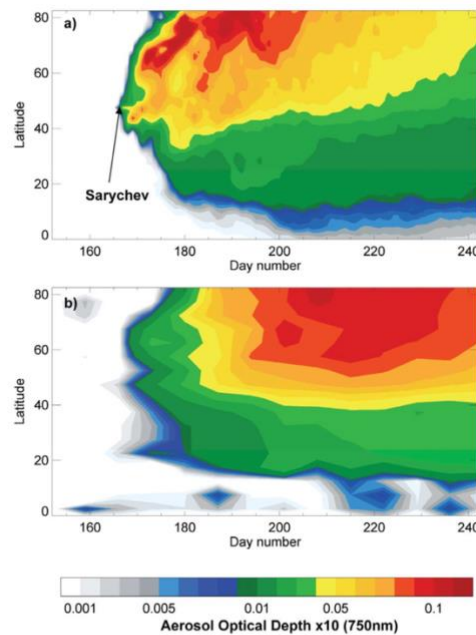

Figure 5. Time versus latitude plots of the aerosol optical depth at 750 nm from (a) HadGEM2 and (b) OSIRIS. The pre-eruption zonal mean AOD is removed from the OSIRIS data to give the perturbation to the AOD.

Question 2: If volcanic ash would be present in the northern hemispheric stratosphere in summer and autumn 2019 (authors write 0.4 – 1.8 Tg) in a comparable amount as sulfate (authors write 1.5Tg ± 0.2 Tg), one should find a lot of cases with enhanced particle depolarization ratios of the stratospheric aerosol layers, for example with CALIOP measurements. To our knowledge this was not the case.

Here are some examples that show low depolarization ratios only:

https://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_v4_detail.php?s=production&v=V4-10&browse_date=2019-07-18&orbit_time=06-56-08&page=4&granule_name=CAL_LID_L1-Standard-V4-10.2019-07-18T06-56-08ZD.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=04-54-59&page=4&granule_name=CAL_LID_L1-Standard-V4-10.2019-07-20T04-54-59ZD.hdf

With the greatest of respect, we present considerable evidence that the size distribution is perturbed which can only be through the presence of volcanic ash. We utilise the OMPS-LP Angstrom exponent for the first 30 – 40 days (see Figure 8 of the manuscript).
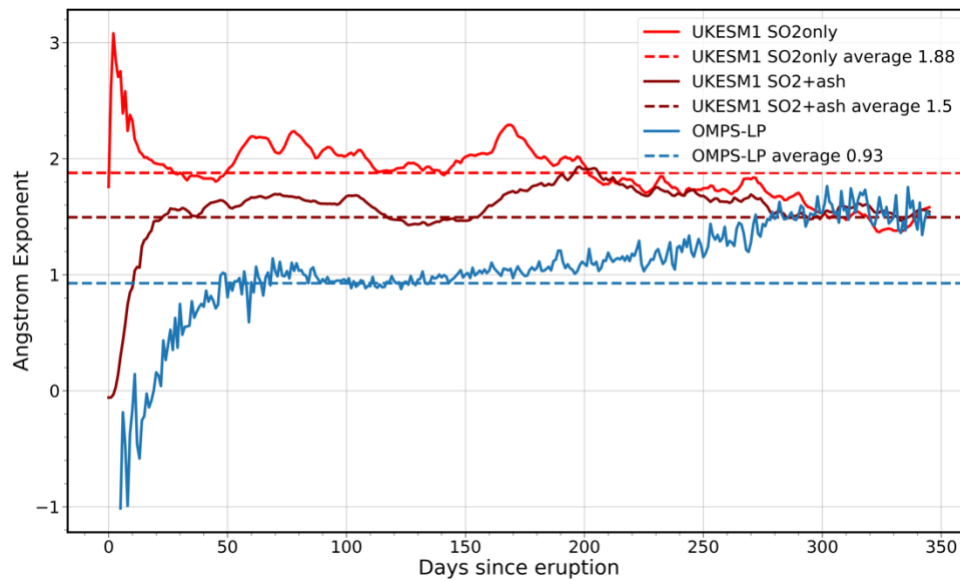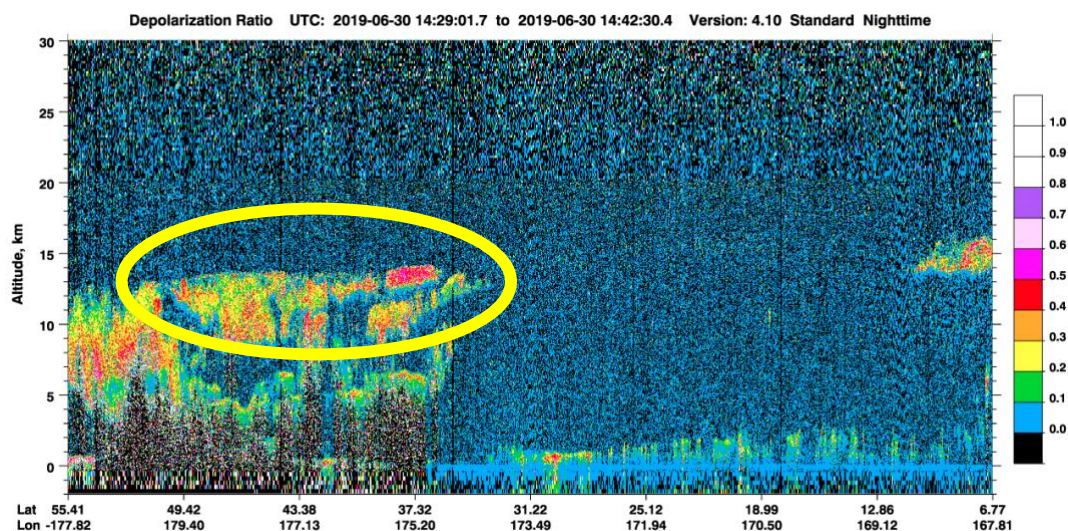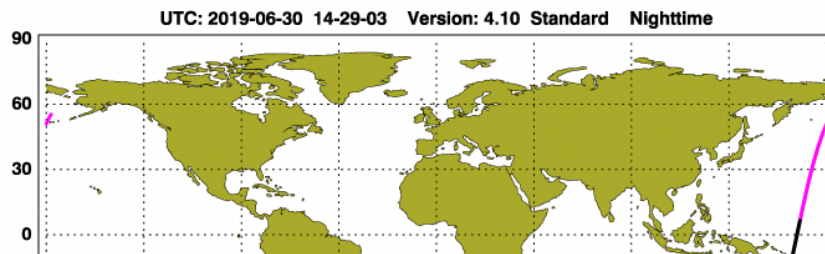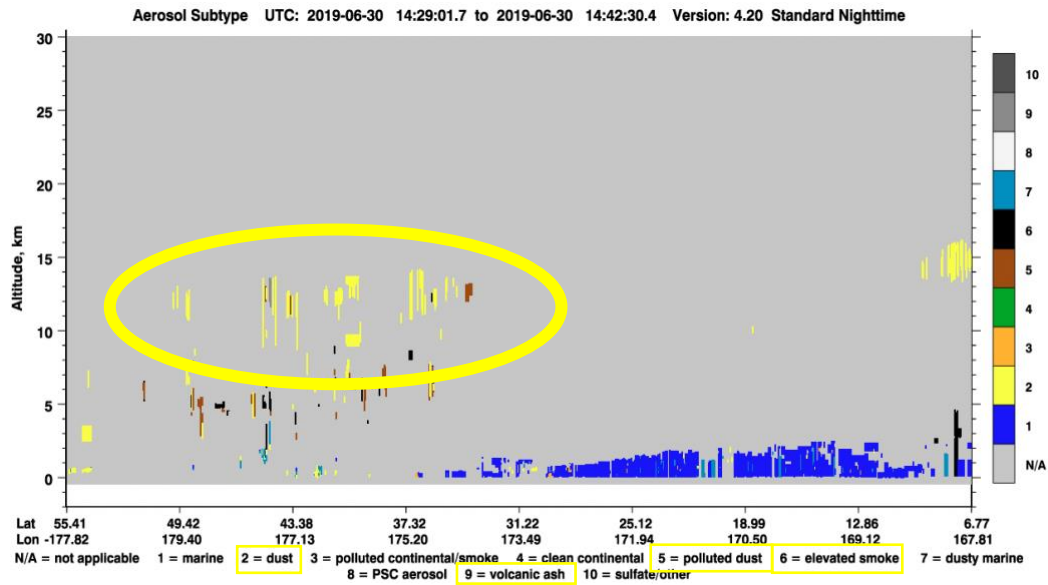
Figure 8: Daily evolution of Ångström Exponent for OMPS-LP (blue), UKESM1 SO2only (red) and UKESM1 SO2+ash (dark red). Calculated using area weighted sAOD between 30–90° N (same as Fig. 7) using 510nm and 869nm wavelengths.

We note that the observations do not suggest an extended influence in 'Autumn', i.e. September, October, November. The e-folding time for the Angstrom exponent suggest that the depolarising ash has been removed from the atmosphere within the first 40 days, while the model suggest that it is removed over a shorter period (about 20days). Note that the values of Angstrom exponent from around days 50 – 300 may be smaller than that after day 300 because sulfate aerosol under volcanic eruptions will exhibit larger sizes (and hence smaller Angstrom exponent) than background sulfate because of enhanced coagulation. This has been known from measurements during quiescent and eruption influenced measurements from balloon-borne optical particle counters for several decades (e.g. SPARC report; SPARC Report No.4 | SPARC (sparc-climate.org), their Figure 1.2, where the accumulation mode gradually decreases in number relative to the Aitken mode).
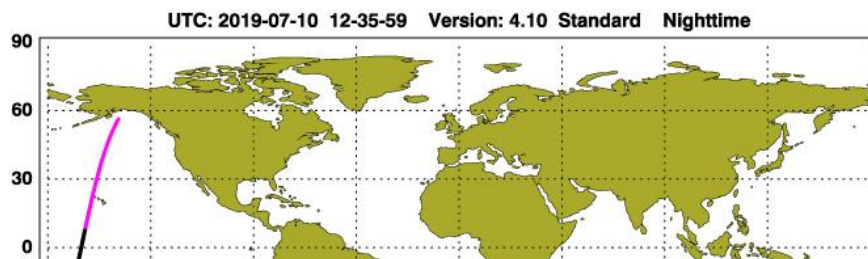
The OMPS-LP data suggest that super-micron ash is present in the stratosphere for around 40 days. We present below examples of CALIOP stratospheric aerosol layers with enhanced depolarization ratios. The first is from 30th June, nearly 10 days after the eruption.

Aerosol Subtype   UTC: 2019-06-30 14:29:01.7 to 2019-06-30 14:42:30.4   Version: 4.20 Standard Nighttime

| Lat | 55.41 | 49.42 | 43.38 | 37.32 | 31.22 | 25.12 | 18.99 | 12.86 | 6.77 |
| Lon | -177.82 | 179.40 | 177.13 | 175.20 | 173.49 | 171.94 | 170.50 | 169.12 | 167.81 |

N/A = not applicable   1 = marine   2 = dust   3 = polluted continental/smoke   4 = clean continental   5 = polluted dust   6 = elevated smoke   7 = dusty marine
8 = PSC aerosol   9 = volcanic ash   10 = sulfate/other

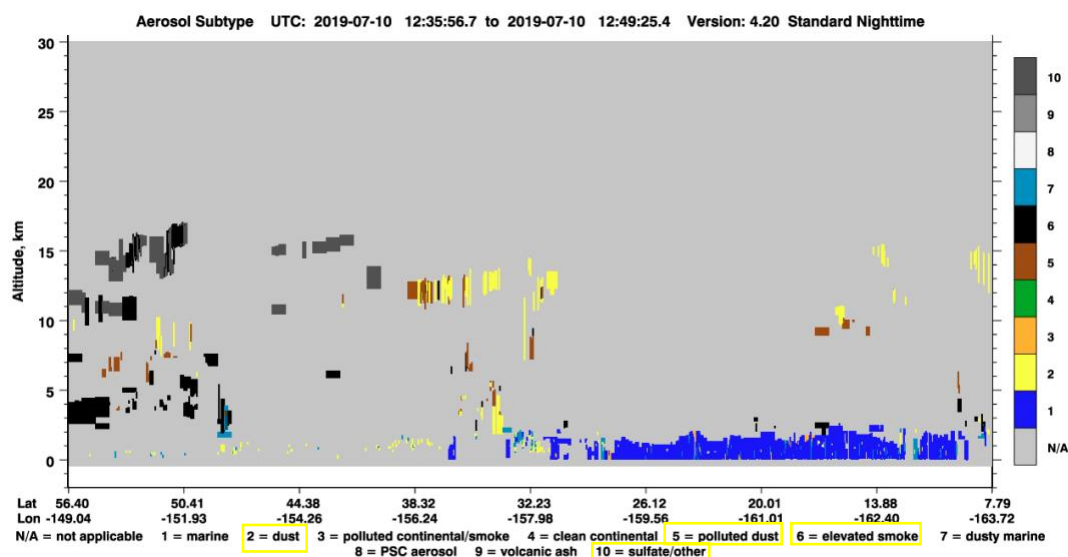UTC: 2019-06-30 14-29-03   Version: 4.10 Standard   Nighttime

This aerosol has come from the volcanic eruption (Figure 1) due to the timing and location that we have chosen. It has certainly not come from the Siberian wildfires as they didn't begin until 19th July. CALIOP identifies this mostly as dust but also includes other aerosol subtypes including polluted dust, elevated smoke and volcanic ash.

Almost 20 days after the eruption we present another CALIOP swath across the north pacific. We again see a combination of aerosol subtypes identified here including dust, polluted dust, elevated smoke and sulfate.

UTC: 2019-07-10 12-35-59   Version: 4.10 Standard   Nighttime

Aerosol Subtype UTC: 2019-07-10 12:35:56.7 to 2019-07-10 12:49:25.4 Version: 4.20 Standard Nighttime

| Lat | 56.40 | 50.41 | 44.38 | 38.32 | 32.23 | 26.12 | 20.01 | 13.88 | 7.79 |
| Lon | -149.04 | -151.93 | -154.26 | -156.24 | -157.98 | -159.56 | -161.01 | -162.40 | -163.72 |

N/A = not applicable   1 = marine   2 = dust   3 = polluted continental/smoke   4 = clean continental   5 = polluted dust   6 = elevated smoke   7 = dusty marine
8 = PSC aerosol   9 = volcanic ash   10 = sulfate/other

We further present a final CALIOP swath almost 30 days after the eruption. Even after 30 days there are still a combination of aerosol subtypes identified by CALIOP including dust, elevated smoke, volcanic ash and sulfate.

UTC: 2019-07-18 00-22-02   Version: 4.10 Standard   Daytime

Aerosol Subtype UTC: 2019-07-18 01:02:27.6 to 2019-07-18 01:14:09.2 Version: 4.20 Standard Daytime

| Lat | 76.27 | 80.47 | 81.77 | 79.06 | 74.30 | 68.83 | 63.07 |
| Lon | 156.09 | 132.98 | 92.80 | 58.47 | 40.88 | 31.53 | 25.78 |

N/A = not applicable   1 = marine   2 = dust   3 = polluted continental/smoke   4 = clean continental   5 = polluted dust   6 = elevated smoke   7 = dusty marine
8 = PSC aerosol   9 = volcanic ash   10 = sulfate/other

We conclude that, as evidenced from the OMPS-LP, super-micron dust/ash is present in significant quantities for the first 30 – 40 days after the eruption. In each of the example's dust (yellow) was identified by CALIOP due to larger depolarizing ratios.

Question 3: Why is the potential impact of the record-breaking Siberian fires on the UTLS aerosol load totally ignored (for the period from August 2019 to December 2019)? The smoke certainly influenced the aerosol in the 8-15 km height range for latitudes from 65-90°N. The Siberian smoke is discussed by Ohneiser et al. (ACP, 2021) and by Ansmann et al. (Frontiers, 2021).

We hypothesize that simulations with sulfate and smoke (instead of sulfate and ash) may even explain better all the OMPS-LP and CALIOP observations.

Thank you for highlighting this. The manuscript has been updated to include information about the Siberian and Canadian forest fires. As you say, this is a hypothesis, but far from proven at present. For example, the study of Boone et al (2022) conclude, "Contrary to previous reports, the aerosol blanket was not comprised of smoke particles". A sensitivity study including sulfate and smoke would be interesting but something for future work.

Question 4: How is the AOD computed in the case of the CALIOP observations. Is it computed from the backscatter profile multiplied by a sulfate lidar ratio (of 40-50sr)? ...and then integrated from the tropopause to 20 km height?

Since we believe that the smoke fraction was more than 80-90% (and the sulfate fraction 10- 20%, and the ash fraction 0%) at high northern latitudes in the UTLS height range then the appropriate lidar ratio is 80-90 sr according to Ohneiser et al. (2021) and then the CALIOP AOD would be even a factor of 2 larger then shown since September 2019.

The AOD is calculated using the CALIPSO L2 aerosol extinction coefficient at 532nm. We use the observed tropopause height and integrate above this to calculate the stratospheric aerosol optical depth. We have acknowledged the limitations of using this data in the updated manuscript.

Question 5: Are you sure that all the spaceborne passive remote sensing techniques work properly during the maximum of the stratospheric perturbation in August 2019 and September 2019? Maybe the channels were almost, partly or totally saturated or almost saturated, at least strongly biased?

We acknowledge in the text that there are limitations to using a limb-profiler for the months immediately following the eruption. This is taken into account when we create the combined observational dataset in section 4.3.

Table 1 is not useful, AERONET data are biased by the tropospheric impact, measurements at 19-20°N were certainly influenced by the Ulawun eruption. It is at least impossible to state the Ulawun eruption had no effect. What about an MLO lidar? If there is a lidar, what depolarization ratio was measured, what about the observed AOD?

Since the AOD is elevated for three months at 95% confidence we disagree that Table 1 (now Table 3) is not useful. The likelihood of this happening by change (assuming that each month is independent) would be approximately 0.05^3. We acknowledge the impact of the Ulawun eruption on the Mauna Loa observations in the text. We also compared the two UKESM1 model simulations (SO2only and SO2+ash) to those performed as Raikoke-only (removing the Ulawun injections) and noted a negligible influence ($<0.3 \times 10^{-3}$) from the eruption in the MLO region.

Is the Brewer-Dobson Circulation already so strong in August and September to explain such a strong transport of aerosol towards the North Pole?

The transport of the plume and therefore the impact on the climate can be dependent on a multitude of parameters, including the location of the volcano and the local meteorological variability (e.g. Jones et al., 2017). We can also see from the observations of sulfate aerosol in the third CALIOP swath which we provide in this document, that the plume has travelled northwards of 60N by 18th July.

Question 6: According to Fig. 9, the initial aerosol optical thickness in summer would be overestimated when assuming a sulfate + ash mixture. However, already from October onwards the high observed AOT values cannot be explained with sulfate + ash only; and of course, not at all with sulfate only. The decay times of sulfate and sulfate + ash seem to be way to short in order to describe the high AOTs in late 2019 and early 2020. The decay time of wildfire smoke is longer and the authors should mention that especially the Arctic aerosol situation was more influenced by wildfire smoke particles. A significant amount, especially north of 65-70°N the dominating amount of the stratospheric aerosol type must have been Siberian wildfire smoke particles, as Ohneiser et al. 2021, ACP and Ohneiser et al. 2022,ACPD write.

Thank you for highlighting this. Discussion around the influence of the Siberian and Canadian wildfires has been included in the revised manuscript. However, as there is conflicting scientific evidence e.g. the analysis by Boone et al. (2022) of ACE data, we address both sides of the argument surrounding the impact of the Siberian and Canadian wildfires.

Figure 9 is confusing. A mean tropopause for the latitudes from 30-90°N makes no sense.... A UTLS layer, one half in the upper troposphere and one half in the stratosphere, is not what is expected after a moderate eruption such as the Raikoke eruption. One expects that the layer is fully and clearly above the tropopause. And all the aerosol in the upper troposphere (at latitudes below about 55°N) should have been be efficiently removed by cirrus clouds. In addition, we speculate, nobody knows the exact SO2 injection heights? Thus, all the simulations seem to be just playing around with possibilities.

We agree that it is difficult to identify the transport of the aerosol under an area average. Therefore we have included a new figure which shows the monthly aerosol extinction coefficient as a function of latitude and altitude. This gets round the problem of showing a single line for the height of the tropopause (albeit with the variability also indicated). In this figure we do see some transfer of aerosol from the stratosphere to the troposphere, mostly at the midlatitudes. This section has been rewritten to include the new analysis.

References:

Abdelkader, M., Stenchikov, G., Pozzer, A., Tost, H., and Lelieveld, J.: The effect of ash, water vapor, and heterogeneous chemistry on the evolution of a Pinatubo-size volcanic cloud, Atmos. Chem. Phys., 23, 471-500, 10.5194/acp-23-471-2023, 2023.

Boone, C. D., Bernath, P. F., Labelle, K., and Crouse, J.: Stratospheric Aerosol Composition Observed by the Atmospheric Chemistry Experiment Following the 2019 Raikoke Eruption, Journal of Geophysical Research: Atmospheres, 127, e2022JD036600, https://doi.org/10.1029/2022JD036600, 2022.

Browell, Edward V., Carolyn F. Butler, Marta A. Fenn, William B. Grant, Syed Ismail, Mark R. Schoeberl, Owen B. Toon, Max Loewenstein, and James R. Podolske. "Ozone and aerosol changes during the 1991-1992 Airborne Arctic Stratospheric Expedition." Science 261, no. 5125 (1993): 1155-1158.

Haywood, J. M., Jones, A., Clarisse, L., Bourassa, A., Barnes, J., Telford, P., Bellouin, N., Boucher, O., Agnew, P., Clerbaux, C., Coheur, P., Degenstein, D., and Braesicke, P.: Observations of the eruption of the Sarychev volcano and simulations using the HadGEM2 climate model, Journal of Geophysical Research, 115, 10.1029/2010jd014447, 2010.

Jones, A. C., Haywood, J. M., Dunstone, N., Emanuel, K., Hawcroft, M. K., Hodges, K. I., and Jones, A.: Impacts of hemispheric solar geoengineering on tropical cyclone frequency, Nat Commun, 8, 1382, 10.1038/s41467-017-01606-0, 2017.

Shallcross, S., Mann, G., Schmidt, A., Haywood, J., Beckett, F., Jones, A., Neely, R., Vaughan, G., and Dhomse, S.: Long-lived ultra-fine ash particles within the Pinatubo volcanic aerosol cloud and their potential impact on its global dispersion and radiative forcings, April 01, 2021, 10.5194/egusphere-egu21-16034, 2021.

Stenchikov, G., Ukhov, A., Osipov, S., Ahmadov, R., Grell, G., Cady-Pereira, K., Mlawer, E., and Iacono, M.: How Does a Pinatubo-Size Volcanic Cloud Reach the Middle Stratosphere?, Journal of Geophysical Research: Atmospheres, 126, e2020JD033829, https://doi.org/10.1029/2020JD033829, 2021.

Vaughan, G., D. P. Wareing, S. B. Jones, L. Thomas, and N. Larsen. "Lidar measurements of Mt. Pinatubo aerosols at Aberystwyth from August 1991 through March 1992." Geophysical research letters 21, no. 13 (1994): 1315-1318.

Vernier, Jean-Paul, T. Duncan Fairlie, Terry Deshler, Murali Natarajan, Travis Knepp, Katie Foster, Frank G. Wienhold, Kristopher M. Bedka, Larry Thomason, and Charles Trepte. "In situ and space-based observations of the Kelud volcanic plume: The persistence of ash in the lower stratosphere." Journal of Geophysical Research: Atmospheres 121, no. 18 (2016): 11-104.