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Measurement Report: Lidar measurements of stratospheric aerosol following the Raikoke and Ulawun volcanic eruptions

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Abstract. On 22 June 2019 the Raikoke volcano in the Kuril islands erupted, sending a plume of ash and sulphur dioxide into the stratosphere. A Raman lidar system at Capel Dewi Atmospheric Observatory, UK (52.4°N, 4.1°W) has been used to measure the extent and optical depth of the stratospheric aerosol layer following the eruption. The elastic channel allowed measurements up to 25 km, but the Raman channel was only sensitive to the troposphere. Therefore, backscatter ratio profiles were derived by comparison with aerosol-free profiles derived from nearby radiosondes, corrected for aerosol extinction with a lidar ratio of 40-50 sr. Small amounts of aerosol were measured prior to the arrival of the volcanic cloud, probably from pyroconvection over Canada. Volcanic ash may have first arrived as a thin layer at 14 km late on 3 July, and was certainly detected from 13 July onwards, eventually extending up to 20.5 km. Aerosol optical depths reached around 0.05 by early August, decaying thereafter to around 0.01 by the end of 2019 and remaining around that level until May 2020. The location of peak backscatter varied considerably but was generally around 15 km. However, on one notable occasion on 25 August, a layer around 300 m thick with peak lidar backscatter ratio around 1.5 was observed as high as 21 km.

1 Introduction

On 22 June 2019 the Raikoke volcano in the Kuril Islands (48.29°N, 153.25°E) erupted, sending a plume of ash and sulphur dioxide into the stratosphere (NASA Earth Observatory, 2019). With an estimated 1.5 Tg of SO_2 (Mann, G. and Vernier, J.-P., 2019), it was one of the largest injections of volcanic aerosol into the stratosphere since the Pinatubo eruption in 1991 and created vivid sunsets around the northern hemisphere (Science, 2019). Sulphur dioxide was measured from 11 to 20 km by the TROPOMI instrument on the SENTINEL-5 satellite on 24 June, with ash detected by the CALIOP/CALIPSO spaceborne lidar at 17 km on 22 June and around 13 km on 23 and 24 June (Hedelt et al., 2019). Lidar measurements from Hawaii measured a layer of aerosol around 1 km thick at 26 km on 24 September 2019 (Chouza et al., 2020); these authors also found using CALIOP data that the layer had ascended from around 19 km in the previous 2 months. During the months following the initial

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eruption the aerosol evolved in both height, depth and optical thickness, merging with aerosol from the eruption of Ulawun in Papua New Guinea (5.05°S, 151.3292°E) that occurred on 26 June 2019 (Chouza et al., 2020).

Measurements are presented here from a Raman lidar system based at the Capel Dewi Atmospheric Observatory, UK (52.4°N, 4.1°W), beginning from June 2019 and continuing until the Spring of 2020, showing how the aerosol cloud evolved over the lidar site.

2 Method

The Capel Dewi Raman lidar system (Vaughan et al., 2018) operates in the ultraviolet at 355 nm using a Continuum Powerlite 9030 laser with a pulse energy of 300 mJ and a pulse repetition frequency of 30 Hz. The receiver system is usually optimised for measuring signals above 2 km and has three interference filters to measure the elastic backscatter (355 nm), nitrogen Raman scattering (387 nm) and water vapour scattering (408 nm). Photon-counting electronics are used with range gates of 100 ns, providing a range resolution of 15 m. To enhance sensitivity in the elastic channel, a neutral density filter was removed, which extended the measurement range to around 25 km in the lower stratosphere, but raising the lower limit to around 7 km. All the data presented here were collected during night-time operation.

Raw data was collected with a time resolution of 10 minutes on most nights, and the files combined to whole-night averages for further analysis. Filters were applied during averaging to remove files affected by low cloud to guard against signal-induced noise problems. Despite this, the faint signals on the N_2 Raman channel in the lower stratosphere on this channel meant that long runs of data had to be combined to accumulate enough signal for analysis. This was only possible for a few nights during the period under consideration here.

Therefore, for each night of measurement, a density profile from a nearby radiosonde ascent (chosen using the wind direction at 200 mb) was used to simulate an aerosol-free lidar profile, which was fitted to the elastic signal above the aerosol layer on that night (usually above 20 km). An onion-peeling retrieval with prescribed lidar ratio (ratio of aerosol extinction to backscatter coefficient) was then used to derive the lidar backscatter ratio down to the upper troposphere. As cirrus clouds were frequently observed near the tropopause, the algorithm used two layers with different lidar ratio: a stratospheric value above 12 km (or above the cirrus layer if this was higher) and a different, usually lower value, below this height. (During the pyroconvective period absorbing aerosol was found near the tropopause, necessitating a larger value of lidar ratio).

The choice of lidar ratio is to some extent arbitrary, since a wide range of values are given in the literature for aerosols of volcanic origin. For example, Mattis et al. (2010), using Raman lidar measurements for small volcanic plumes in 2008-9, found a range of values from 30 to 60 sr at 355 nm. Ash tends to increase the lidar ratio: Lopes et al. (2019) quoted 63 $\pm$ 21 sr for the Calbuco eruption plume of 2015; Chouza et al. (2020) quoted 64 $\pm$ 27 sr for the Nabro plume of 2011 and Hoffmann et al. (2010) quoted 63 $\pm$ 10 sr for the Kasatochi plume of 2008. Remarkably consistent though these results are, Mie scattering calculations suggest that the lidar ratio depends strongly on particle size (Korshunov and Zubachev, 2013) and therefore varies from eruption to eruption.

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For nights when the volcanic aerosol plume was bounded above and below, and there was no cirrus cloud in the troposphere, an appropriate value of lidar ratio could be found by requiring that the backscatter return to the molecular profile below the layer. For most of the period of this study this resulted in values around 40 sr, which has been adopted for most of the dataset for consistency. A cross-check on the lidar ratio was provided on a few nights where enough profiles could be combined to yield a useful signal on the Raman channel. This allowed an independent measure of the optical depth of the stratospheric aerosol layer, further discussed below.

One of the characteristics of the Capel Dewi lidar is that its receiver is only sensitive to signals whose polarisation is parallel to that of the incident laser beam. Thus, when non-spherical particles are present, the backscatter ratio is underestimated and the effective lidar ratio becomes artificially large.

3 The Raikoke eruption

Following the eruption of the Raikoke volcano on 22 June 2019, the ash and sulphur dioxide plume initially moved westward before being entrained in a cyclonic circulation over the North Pacific. The on-line NOAA HYSPLIT model (Draxler and Hess, 1998) provides a tool for calculating the dispersion of a volcanic plume and figure 1 shows the simulated ash cloud 72 hours after the eruption, confined to a region between Kamchatka and Alaska. Profiles from CALIOP (available from https://www-calipso.larc.nasa.gov/products/lidar/browse_images/std_v4_index.php, last access: 26 August 2020) confirm the location of the ash plume at this time, along an orbit between 56°N, 177.8°E and 61.5°N, 173.5°E, at altitude 12-14 km.

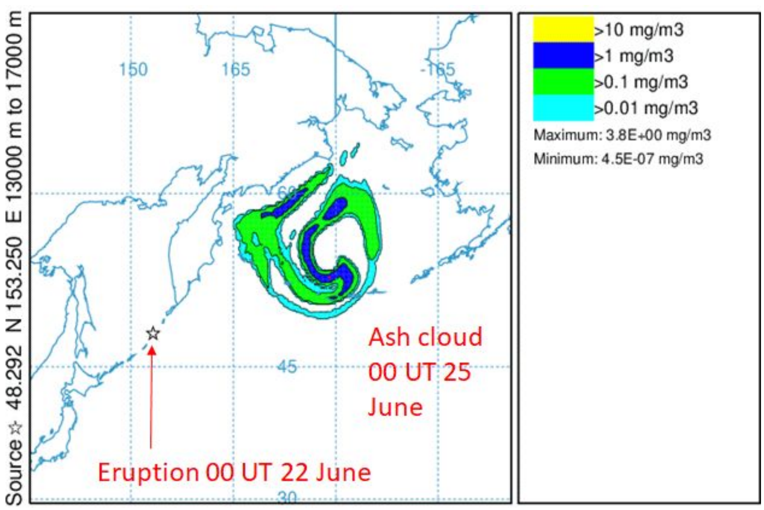

Figure 1. Dispersion of the volcanic ash cloud according to the Hysplit model 72 hours after the eruption. The model was initialised with a uniform ash injection between 13 and 17 km. Mass loadings are arbitrary and serve only to delineate the position of the cloud.

CALIOP profiles in the period 25-28 June show that the volcanic ash stayed broadly in the same region, becoming thin, patchy and generally confined below 15 km. The depolarisation ratio at 532 nm decreased from 30-40% on the 25th to below

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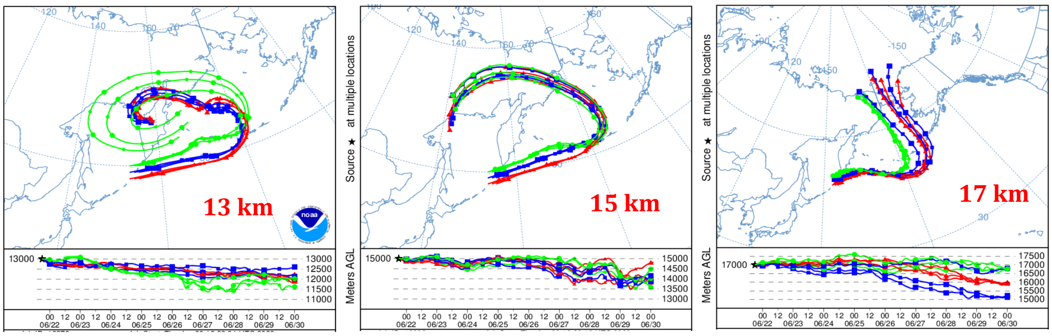

Figure 2. 8-day forward trajectories from nine points surrounding Raikoke, calculated using Hysplit and initialised 18 UT 21 June 2019, at three lower stratospheric heights

20% by the 28th. HYSPLIT trajectory calculations (figure 2) are consistent with the observations, suggesting little transport of material from the cyclonic circulation until the end of June; they are remarkably non-dispersive for 8-day trajectories. A further set of Hysplit trajectories (not shown), initialised from the end points of those in Figure 2, suggested the plume was confined until around 6 July, but these trajectories were more dispersive and cannot rule out a certain amount of transport westward in the first week of July.

During the latter half of June and in early July, pyroconvection over Canada injected layers of smoke and ice clouds into the lower stratosphere (similar to the case described by Vaughan et al. (2018)), making it difficult to distinguish the progression of volcanic ash remnants using the CALIOP profiles. We now turn to the lidar measurements at Capel Dewi and the arrival of the volcanic cloud in Europe.

4 Results

Lidar profiles at the end of June and the first few days of July 2019 showed numerous small aerosol layers in the lower stratosphere. An example, from the night of 1-2 July, is shown in figure 3a. Two aerosol layers are shown in this figure - one around 12 km, just above the tropopause, which seems from its optical properties to have been smoke (a lidar ratio of 100 sr was needed to account for the attenuation of the laser beam through the layer) and another between 13 and 14 km where a lidar ratio of 40 sr sufficed. A CALIPSO orbit around 3° east of the lidar at 0250 UT on 2 July measured an aerosol layer around 12-13 km between 53.7°N and 55.5°N with depolarisation ratio 10-20%, confirming the presence of non-spherical particles in the lower layer, but showing no trace of the upper layer.

A much more prominent aerosol layer was measured two days later (figure 3b); the maximum backscatter ratio was now 1.3 rather than 1.09 on 1 July. Again there are two layers, with the lower one more variable and more absorbing than the upper one. Both probably consisted of depolarising particles: CALIOP measured patches of depolarising aerosol ($\delta_v \approx 0.1 - 0.2$) at

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52.0°N, 4.4°E, both at 11 and 14 km, at 0220 UT on 4 July. These are most likely due to pyroconvection, but it is not possible to rule out the arrival of volcanic aerosol over Europe at this time. To emphasise the patchy nature of the observations in early July, there was no aerosol above the tropopause on the following night (4-5 July) and only a faint layer on 5-6 July (12 - 14 km, maximum backscatter ratio 1.03).

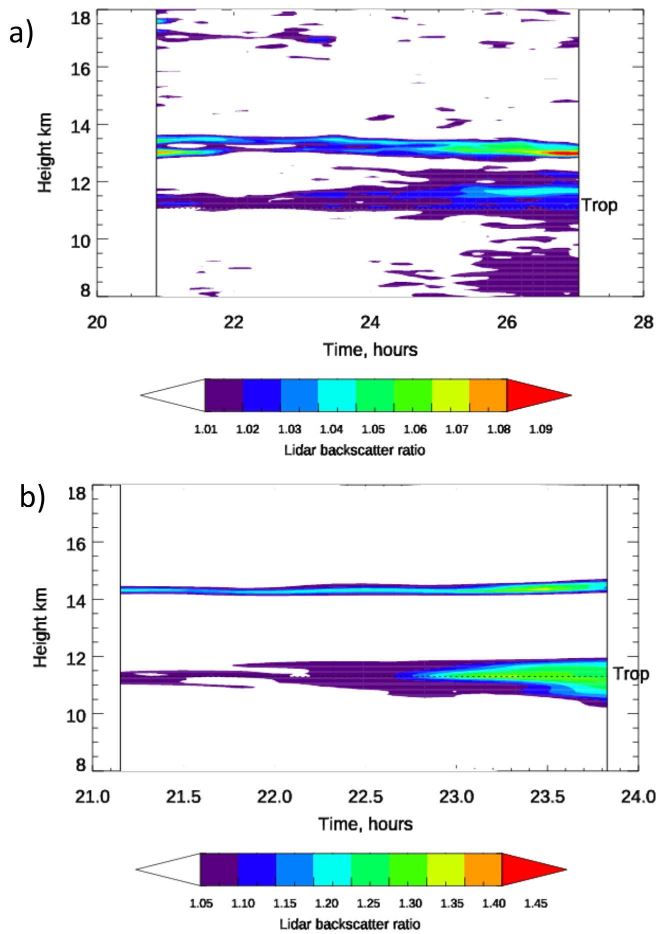

Figure 3. Lidar backscatter ratio measured at Capel Dewi during the nights of a) 1 July and b) 3 July 2019. Dotted black line denotes the tropopause height from the radiosonde at Valentia Observatory, Ireland (51.93°N, 10.25°W). Note the different colour scale on the two panels.

The first unambiguous measurement of the volcanic aerosol cloud was on the night of 13-14 July (figure 4a), when a prominent layer of aerosol with peak backscatter ratio 1.4 lay between 12.5 and 15 km. The figure shows the whole-night average (2130 - 0330 UTC); individual profiles showed multiple thin layers 200-300 m thick lasting about an hour (corresponding to around 60 km in length with the wind speed of 18 ms^{-1} measured by the sonde at 14 km). A CALIOP orbit passing to the east

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of the UK on this night measured stratospheric aerosol up to 15 km (figure 5), with very little depolarisation north of 47°N. This indicates that the aerosol consisted of spherical sulphuric acid droplets by this time, with little or no ash.

Measurements after 13 July are consistent with the continued presence of spherical sulphuric acid aerosol over the lidar site. The actual profiles were variable during the first 2-3 months after the eruption, with multiple layers in the height range 12–20 km. A notable example is shown in figure 4b, taken between 20:37 and 21:47 UTC on 25 August. This has a very prominent layer at 21 km - reminiscent of that seen by Chouza et al. (2020) at 26 km over Hawaii on 24 September, which they tracked back to the Kamchatka region. Although CALIOP profiles around 25-26 August showed numerous thin (< 1 km) aerosol layers below 19 km, they showed nothing above 20 km in the vicinity of the UK. The wind speed at 21 km according to the Herstmonceux radiosonde at 00 UTC on the 26th was 10 knots, so the layer in figure 4b could have been as small as 24 km in horizontal extent, perhaps explaining why CALIOP did not observe it.

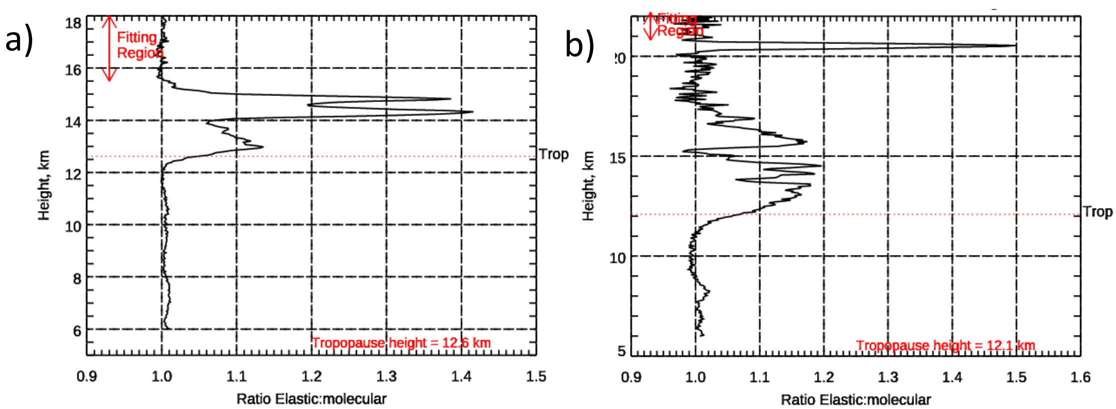

Figure 4. Average backscatter ratios for a) 13-14 July 2019 (21:31 - 03:22 UTC) and b) 25 August 2019 (20:37 - 21:47 UTC). The tropopause height as derived from the radiosonde is shown by the dotted red line.

For consistency in the analysis from 13 July onwards, a lidar ratio of 40 sr was adopted, except for two nights (7 and 13 Sept) when a value of 50 sr was needed to return the backscatter ratio to 1 in the troposphere, where the Raman measurements indicated no aerosol. For non-depolarising aerosol, this is the actual value of lidar ratio, which falls within the range of 30 - 60 sr reported by Mattis et al. (2010).

Figure 6a shows the evolution of the aerosol optical depth above 12 km between 27 June and 30 May 2020 as measured by the Capel Dewi lidar. The optical depths reached around 0.05 by mid-August 2019, declining slowly for the remainder of the autumn. Also shown on the figure are calculations of the aerosol optical depth above 12 km from the Raman channel, for nights clear enough to collect sufficient counts. In this analysis, an aerosol-free lidar profile was calculated from the radiosonde data and fitted to the Raman channel above the aerosol layer; the optical depth could then be derived from the ratio of the two profiles at 12 km. Even with long nights of data (and in the case of 4-6 February 2020, two nights combined data), the precision error bars on these estimates are large - but they are consistent with the estimates from the elastic channel, justifying the choice of lidar ratio. In contrast to the Raman estimates, where the precision error dominates, errors on the optical depth estimates

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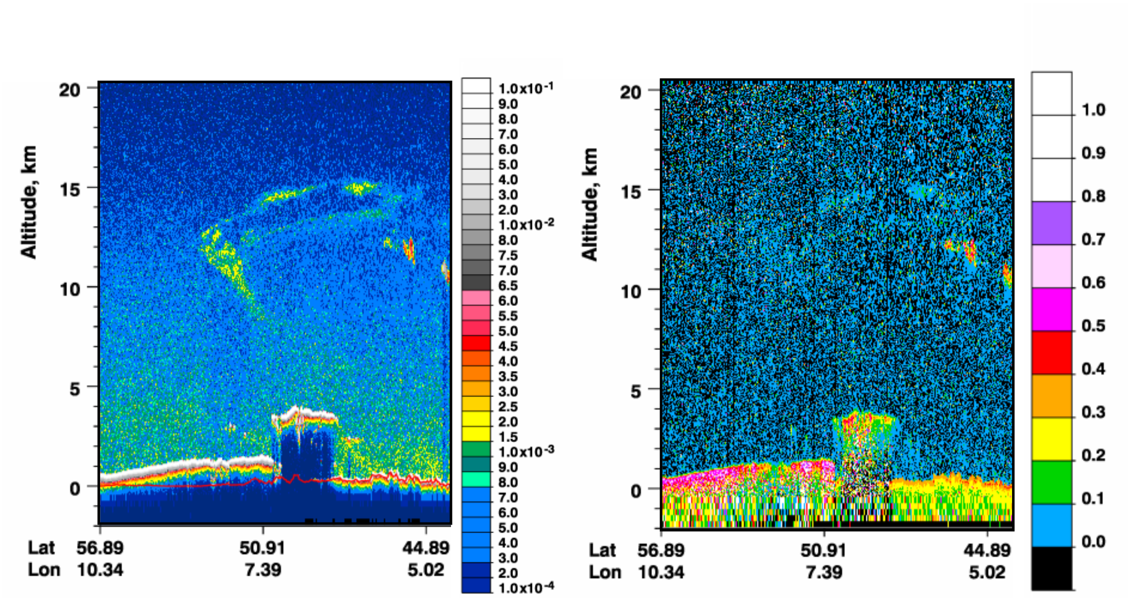

Figure 5. CALIPSO lidar measurements along an orbit to the east of the UK between 0210 and 0213 UTC on 14 July 2019. Left panel: Total attenuated backscatter at 532 nm, in sr^{-1} km^{-1}; right panel: depolarisation ratio. Image courtesy of NASA.

from the elastic channel are dominated by the systematic uncertainty in the lidar ratio, since the precision errors are very small (and not plotted on the figure). For this reason, many lidar groups prefer to present their results as integrated backscatter (e.g. Trickl et al. (2013); Zuev et al. (2017)), which does not depend directly on lidar ratio. Here we present the results as optical depth for comparison with the Raman measurements and because it is a more generally useful quantity, acknowledging that the systematic uncertainty is probably of a similar order to the precision uncertainty on the Raman estimates.

The peak optical depth of around 0.05 was reached at the beginning of August, declining to around 0.01 by the end of the year - an exponential decay time of around 3 months. The optical depth measurements reached a minimum of 0.008 on 4-5 February 2020, increasing slightly thereafter and reaching 0.014 on 20 May.

Figure 6b shows how the maximum measured backscatter ratio for each night increased to a peak of 1.6 on 1 August before decreasing sharply to an average value of 1.045 in 2020. Figure 6c shows the height of the highest extent of the aerosol layer, the peak backscatter ration and (for reference) the tropopause. The maximum height increased from around 15 km in mid-July to 20 km in September, remaining more or less constant thereafter at 20-21 km. The height of maximum backscatter ratio was more variable, with some outliers like that in 4b, but for the most part was around 15 km during 2019, with an apparent descent in 2020 to around 13 km.

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5 Conclusions

The eruption of Raikoke on 22 June 2019 introduced a cloud of ash and sulphur dioxide into the lower stratosphere. For the first couple of weeks after the eruption the cloud remained in the general region between Kamchatka and Alaska, with the SO_2 oxidising to sulphuric acid in the form of spherical droplets. At the end of June and beginning of July 2019, pyroconvection over North America introduced aerosol into the tropopause region, which is difficult to distinguish from this layers of volcanic origin. The first unambiguous observation of volcanic aerosol at Capel Dewi was therefore the night of 13-14 July. CALIPSO profiles in the vicinity measured low depolarisation indicating that the cloud mostly consisted of spherical sulphuric acid droplets.

The measurements show that the aerosol optical depth between 12 and 21 km reached 0.05 at the beginning of August, decaying to 0.01 by the end of 2019, and persisting up to May 2020 at around the same level. The maximum lidar backscatter ratio was 1.6 on 1 August 2019, with a sharp decrease reaching values < 1.1 from December onwards. It is likely that aerosol from the eruption of Ulawun in Papua New Guinea on 26 June 2019 mixed with the Raikoke aerosol over the months following the eruptions, so that the residual aerosol in 2020 contained contributions from both sources.

Data availability. http://dx.doi.org/10.17632/6j67sfwkjx.1

Author contributions. D. Wareing performed the measurements, G. Vaughan and H. Ricketts performed the analysis and all authors contributed to writing the paper.

Competing interests. No competing interests

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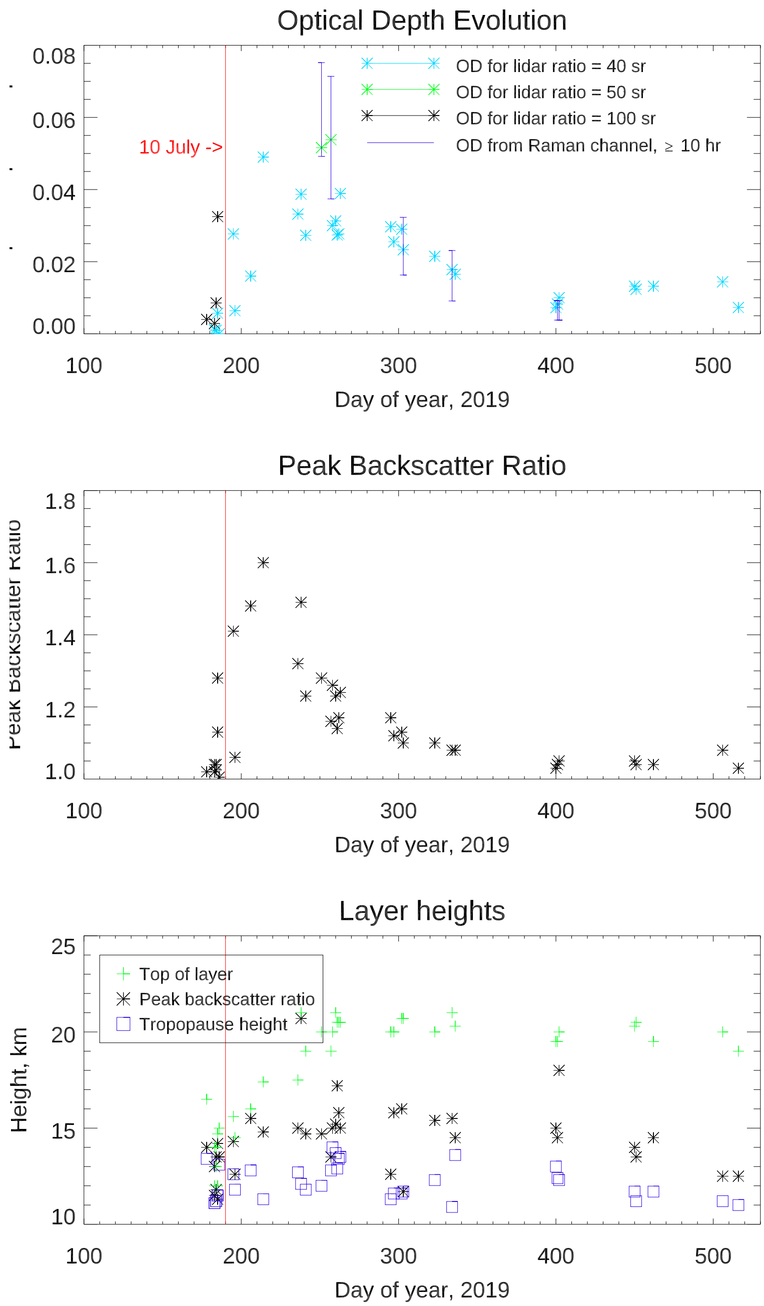

Figure 6. (a) Optical depth of the stratosphere between July 2019 and February 2020. Measurements after 10 July are considered to be volcanic aerosol, for which a lidar ratio of 40 or 50 sr was assumed (see text). Earlier measurements are of smoke layers, where the lidar ratio has to be artificially increased to account for the depolarising particles. Also shown are estimates of optical depth from the Raman channel where more than 10 hours data was measured; the bars denote $\pm 1\sigma$ limits. (b) The corresponding peak backscatter ratio for each night. (c) The height of peak backscatter ratio (black asterisks), the top of the aerosol layer (green crosses) and tropopause height as calculated from a nearby radiosonde (blue squares).