Gorkavyi et al. (hereafter "Gteam") have three primary thrusts in this manuscript on the Raikoke volcano plume of 2019, to 1. characterize the stratospheric plume SO2 and sulfate evolution, 2. Introduce a limb-view retrieval artifact called the "arch effect," and 3. Follow a compact plume element termed a coherent circular cloud (CCC). These themes are carried out by invoking five different satellite-based measurement/retrieval products, three nadir-imaging and two vertical profiling.

The above themes, data, and methods are an appropriate fit for AMT. It is evident from the data presented by Gteam that the Raikoke volcanic cloud is scientifically important, even remarkable, and needs to be thoroughly characterized in the literature. Moreover, Gteam present a novel method for limb-view aerosol profile handling to deal with a known limitation in the limb approach to quantifying perturbations that are inhomogeneous and/or geographically small in the context of instrument resolution.

My summary assessment of GTeam is that each of the three thrusts need substantial revision to qualify as publishable. In each respect, the material has either been presented in prior literature and not fully recognized herein, or the current presentation lacks clarity, validity, or motivation. Details of these primary concerns are next. Following these is a list of minor concerns. Technical concerns and questions are handled

by comments and annotations to the manuscript, provided separately.

<u>Primary Concerns</u>

Gteam's foremost new content regards the "arch effect." Here they focus on a fundamental uncertainty with respect to limb-view spectroscopy, that being the accuracy of retrieving extinction and altitude of an object that is small with respect to the instrument's (OMPS-LP in this case) line-of-sight field of view (FOV). The problem common to all such stratospheric limb viewers is that they peer through a roughly 200 km atmospheric path length (~180 km quoted by Gteam for OMPS-LP) composed of a wide altitude range in the Earth-centric reference frame. Generally, the assumption is made that any object in the view path occupies the entire FOV. To the extent that any object (like a meteorological cloud or aerosol plume) is smaller (vertically or horizontally) than the instrument's FOV, one or both of the retrieved extinction and feature altitude will be biased low. In highlighting the "arch effect," Gteam seem to be suggesting that the Raikoke sulfate plume presents a source of systematic error in OMPS-LP extinction and plume-height results. Hence the motivation for their focus on an adjustment to mitigate the "arch effect."

The "arch effect" argument, to be persuasive, requires at least one additional data item independent of OMPS LP to characterize cloud- or plume-object height, vertical thickness,

and horizontal extent all within the OMPS-LP FOV. I.e. OMPS-LP on its own is under-constrained for such an assessment. Gteam present the arch effect argument by invoking only OMPS-LP profile data. Moreover, the illustrative example of the arch effect is for a scene that has a Raikoke plume object that is ~40 days old. Even though it is presumed to be compact with respect to the OMPS LP FOV, this (and most other) Raikoke plume elements have spread over distances greatly exceeding the OMPS LP FOV. With the data that Gteam present as an example of the arch effect—a consecutive sequence of OMPS LP aerosol profiles—it is not possible to know the true size of the Raikoke plume object precisely in the FOV. It is known, however, that the CCC the Gteam is following has horizontal dimensions as great as ~600 km, based on the CALIOP curtain one day later, shown in Figure 13. It is conceivable that the 31 August OMPS example given for the arch effect was a case of the limb view sampling a peripheral part of the CCC, presenting a much smaller horizontal distance. But insufficient independent information is provided to inform the reader.

Exploring complementary data sets for the Gteam 31 August 2019 case of the arch effect, it becomes apparent that indeed the OMPS center slit sampled CCC they are tracking. The GOES East visible reflectance image shown below illuminates the CCC over coastal northwest Africa at OMPS measurement time: ~13:50 UTC. The visible meridional extent of the CCC is ~3.4

deg. Latitude, approximately 380 km. This is roughly double the OMPS tangent path FOV.
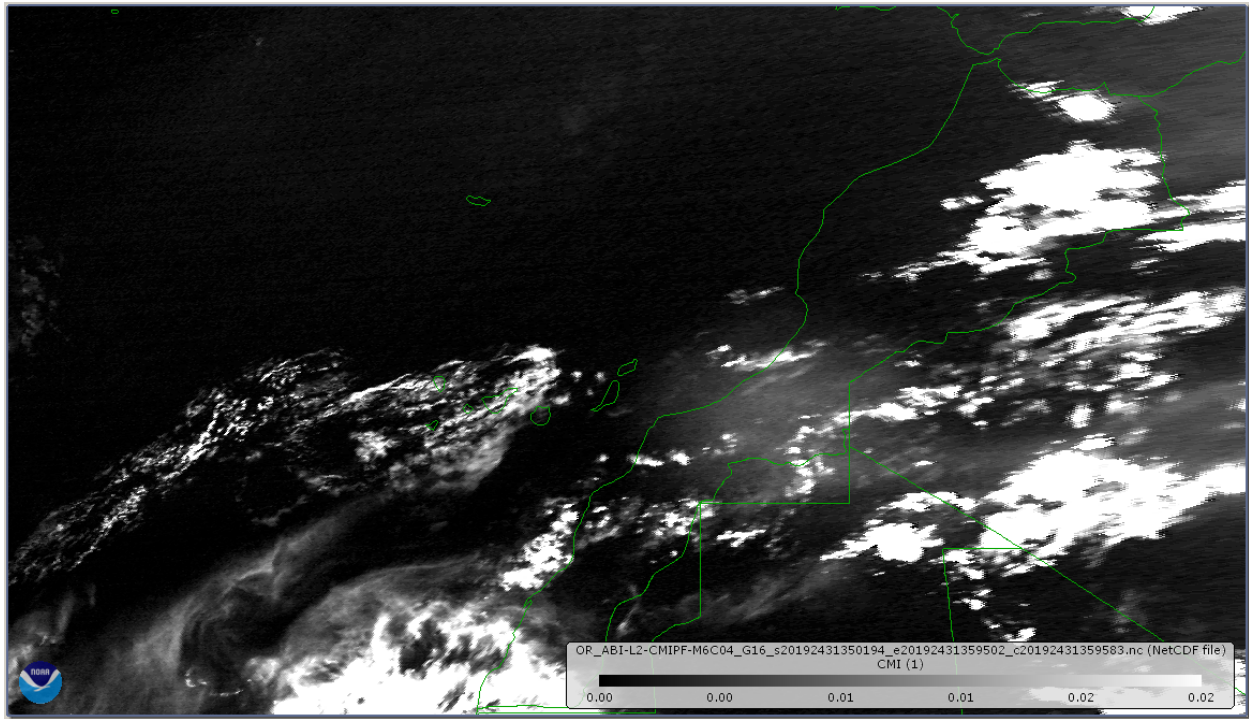
The NASA micropulse lidar (mplnet.gsfc.nasa.gov) at Santa Cruz, Tenerife, (due west of the OMPS extinction-profile curtain in Figure 4) measured the CCC as it blew west. A snapshot of the normalized backscatter ratio profile at 22UTC 31 August is below.

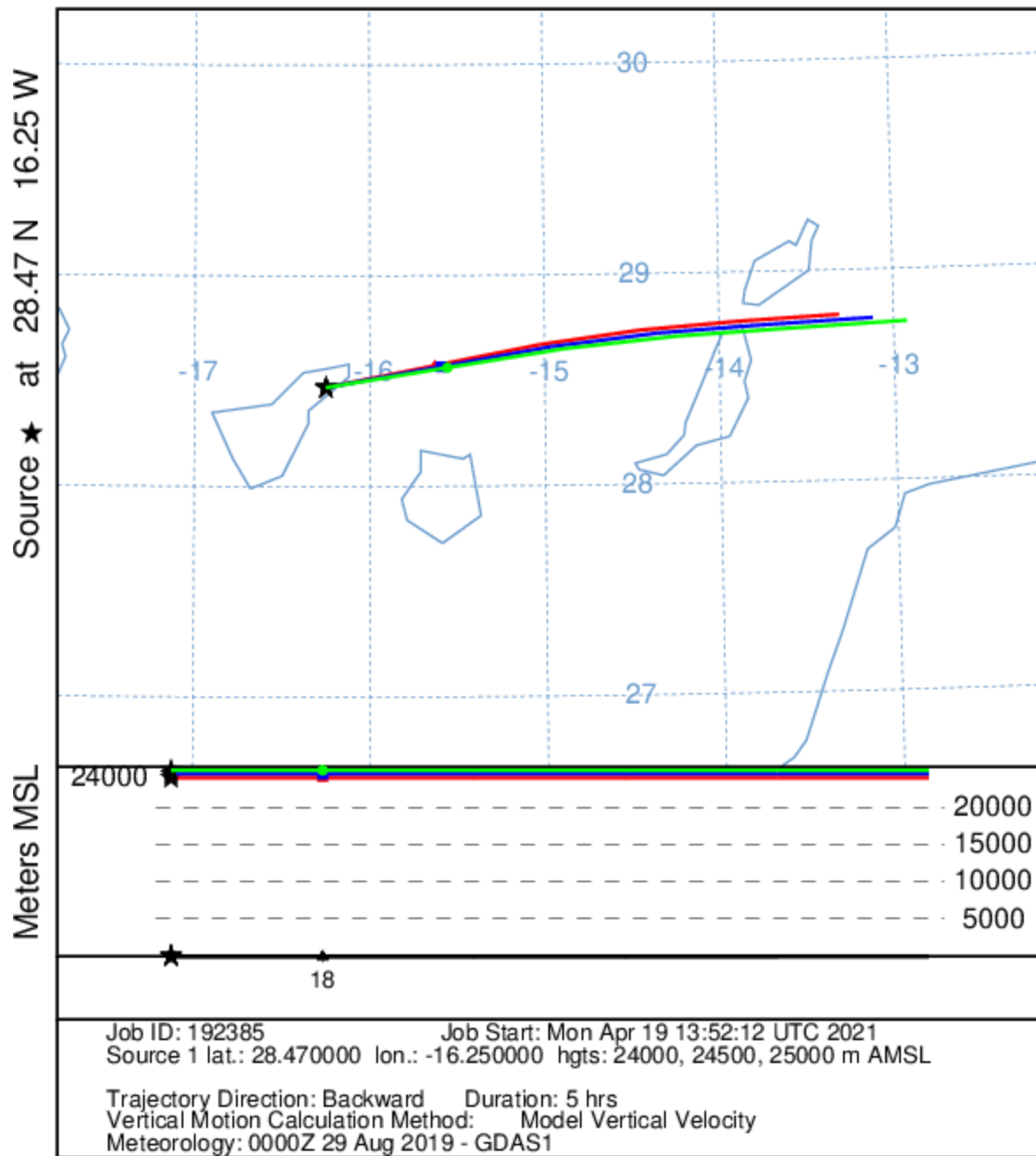MPLNET Santa_Cruz_Tenerife 2019-08-31 22:10:30 V3_L1_NRB: NRB
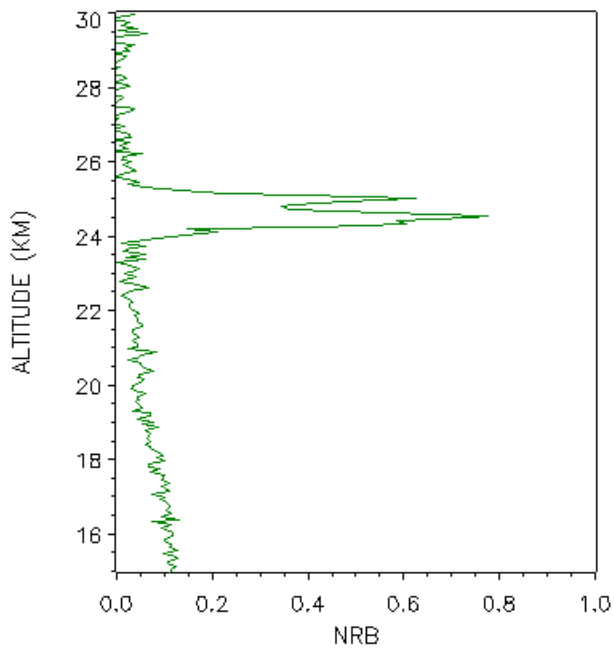
Energy: 6.518 uJ (Set Point: 6.244 uJ)
Box Temperature: 23.883 C (Set Point: 23.380 C)
Detector Temperature: 26.841 C (Set Point: NaN C)
Laser Temperature: 29.920 C (Set Point: NaN C)
Solar Background: 4.320E−05 MHz

QA Flag: Good

Back trajectories to OMPS time 31 August show that the plume over Tenerife indeed passed over the OMPS curtain at OMPS measurement time.

NOAA HYSPLIT MODEL
Backward trajectories ending at 1900 UTC 31 Aug 19
GDAS Meteorological Data

Job ID: 192385   Job Start: Mon Apr 19 13:52:12 UTC 2021
Source 1 lat.: 28.470000 lon.: -16.250000 hgts: 24000, 24500, 25000 m AMSL

Trajectory Direction: Backward   Duration: 5 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 29 Aug 2019 - GDAS1

The nadir GOES image and the precise lidar profile tightly constrain the plume geometry encountered by OMPS. Thus it appears that the CCC conditions encountered by OMPS are

vastly inconsistent with the limited horizontal cloud extent used to illustrate and motivate the arch effect principle. See the annotations to Gteam's schematic in the separate manuscript.

Given these two additional views of the CCC encountered by OMPS LP, it is largely uncertain as to how to interpret the OMPS extinction profiles in Figures 10 and 4. This does not appear to be a candidate for the arch effect, and the maximum extinction in the CCC—according to the example of it shown in the 1 September CALIOP curtain (Figure 13)—may be as much as an order of magnitude greater than retrieved by OMPS (CALIOP backscatter x lidar ratio of 50 sr). Considering all these factors, Gteam is encouraged to either clarify and bolster their arguments for this arch effect example, or find another case where it can be independently shown that a sub-FOV-filling plume element is sampled by OMPS.

If such an example can be demonstrated, it still seems unlikely that the aging/spreading Raikoke plume as sampled by OMPS LP justifies wholistic application of the arch-effect adjustment. The shear variety and complexity of plume presentations (e.g. the various CALIOP curtains illustrated by Gteam) to OMPS during the analysis time frame indicate that compact-to-the-point-of-sub-FOV plume elements are rare. This includes the CCC and the meridionally broader Raikoke layers at lower altitudes do not meet the size-limited view as depicted in Figure 3.

It is essential to characterize the schematic angles and "object" (also referred to as a "cloud") in terms of their geophysical horizontal and vertical dimensions. It is all important to know what size range of cloud in the along-track FOV direction creates the arch effect. It is difficult to know from the schematic how realistic the cloud object is, but it appears to be tiny in relation to the 180 km OMPS LP FOV.

To make a compelling argument for the arch effect, three things would have to be presented that are lacking. 1. A case study involving an independent plume-object physically characterized, 2. Such an element being located within the OMPS LP FOV (i.e. a space-time match), and 3. The plume element being demonstrably smaller than the FOV (as drawn schematically in Figure 3). An example of such a case study is provided in Penning de Vries et al. (2014) (cited by Gteam), who attempted to reconcile SCIAMACHY limb-scatter profiles of the Nabro volcano stratospheric plume with simultaneous nadir SO2 imagery. It is notable that the Nabro example was when the plume was less than two days old; demonstrably compact.

How is an "arch" identified in the OMPS data? How do we know when an arch shape might be geophysically accurate vs. one that is an artifact of a tiny cloud? Gteam describe a wholesale processing of the OMPS LP extinction profiles, applying the arch effect correction to the whole set. At least that is how I understood the method description. If this is the case, does that suggest that Gteam considers the arch effect a global

vulnerability? Much more clarification of the correction application is needed.

I could find no discussion of the vulnerability of OMPS-LP extinction profiles to saturation in the presence of optically thick aerosol plumes. Given the likelihood that the Raikoke sulfates presented widespread scenes of such optically dense conditions, as did previous eruption plumes like Nabro, Sarychev Peak, and Kasatochi (Fomm et al., JGR, 2014; Lurton et al., https://doi.org/10.5194/acp-18-3223-2018) it is important for Gteam to directly address if/how this issue was dealt with in their various visualizations of OMPS LP extinction and SAOD.

Figure 14 and related discussion of CCC: Chouza et al (2020) https://doi.org/10.5194/acp-20-6821-2020 do a very similar tracking of the CCC (although they do not describe in CCC terms). It is not cited by Gteam. There appears to be a similarity in how the CCC is tracked but then a divergence occurs in late August. Because of the similarity and relevance to this paper, Gteam are encouraged to read Chouza et al. and evaluate the similarities and discrepancies between the two treatments of the CCC.

Kloss et al. (2021) present a SAOD analysis very similar to Figure 7. In fact, the same version of OMPS LP retrieval is used for both. I did not see in GTeam what I expected, an acknowledgement of this previously published analysis and a motivation for presenting the analysis anew. The only

substantial differences in the Gteam plot are an addition of two markers for pyroCb sources and a slightly later endpoint. Neither of these visual differences are taken advantage of, from my reading. If I missed it, please advise. Regardless, it would be important for Gteam to assess whether the figure should stay and also cite Kloss et al. for the earlier rendition.

Minor Concerns

Note: Text below in quotation marks is directly from Gteam.

P4, L118: The schematic (Figure 3) shows a "feature" that is limited both horizontally and vertically. Why does this statement call out only the vertically limited aspect? What's the relative importance of limitations in the vertical versus horizontal?

P4, L122: "If we believe that these lower altitude values do not represent a true aerosol signal…" How is this belief ascertained?

Figure 3 caption: The caption needs to describe the meaning of the horizontal solid and dash-dot line. Presumably these show the OMPS instrument vertical FOV. But if so, why are the line styles different?

Figure 4. Please add an x-axis marking/labeling in deg. latitude, to give the reader a useful geographic refence frame.

P6, L153: The Junge layer peaks much higher according to Kremser https://doi.org/10.1002/2015RG000511
citing Junge. An examination of CALIOP data shows lots of Raikoke aerosols widespread, lower down, on both sides of the CCC.

P6, L158: Please define/describe "artifact density." Why is the term "density" used? Why not "extinction?"

P6, L157-161: This paragraph has several vague or unintuitive terms, such as "real," "artifact density," "cleaned." Please replace these terms or define them.

P6, L158: How is an "isolated" feature determined from OMPS data alone? There is no discussion of complementary data (e.g. OMPS nadir) informing the limb data.

P7, L185: "The sensitivity of the satellite data we examined…" What satellite data? OMPS NM? According to Figures 5 and 7, the aerosols are detected much longer.

Figure 5 caption: "5: The daily zonal mean (45-85N) SO2 mass (assuming a cloud height of 13 km) and the aerosol extinction

coefficient at 675 nm (13-18 km.)." This is 6 OMPS LP altitude bins. Which extinction is it? Max, min, average?

Figure 5 caption: "ash fallout" How does the reader know how much ash fallout is observed? What does ash fallout have to do with SO2? Please explain.

Figure 5 caption: "…extinction starts to decrease due to gravitational sedimentation, but very slowly (from OMPS LP and NM data)" What does this mean? How do these inform about sedimentation? Sedimentation is one of several processes that an diminish extinction. How is sedimentation demonstrated herein?

Figure 6 caption: "main plume" How is the "main plume" defined?" This is the first invocation of "main plume."

Figure 6 caption: "south branch" Is "south branch" synonymous with CCC, or is it more general? Perhaps just stick with "CCC" if they are one and the same.

P9, L227: "After that, the detection limit of the instrument…" What id the detection limit? Please state the value.

P9, L239: No pyroCbs are mentioned by Kloss et al. as far as I can tell. If they are, please provide some more detail. If not,

what is the justification for adding these symbols to the figure? There is no evident increase of SAOD clearly attributable to these symbols in the plot.

P10, L241: "The top panel in Fig. 8 shows that the maximum altitudes of the plume are around 25 km, when the plume penetrates the tropics." How is "when" shown? Figure 7 seems to show that Raikoke material only got to the subtropics.

Figure 8(a): Please explain the two apparent outliers of ~25 km layers over Northern Europe. They seem to be all by themselves; no indication of a ramp up to those altitudes from nearby layer observations.

Figure 8(a) caption: What is the number of days shown? Span of time? Are all the reported dates consecutive, or are there gaps? If there are significant gaps, these dates should be mentioned.

Figure 8 caption: The sAOD panel is not described.

Figure 8(b) caption: How are the data comprising the white line determined? The white line doesn't conform closely to the color shading. I.e. it crosses extinction contours in time. Please explain fully.

Figure 9 caption: The Junge layer is higher than the range indicated here.

P16, L309: Specifically, how were the CCC detections done out to 22 September? I could not find a definitive explanation.

Figure 14 caption: "OMI" is mentioned here for the first time as part of this analysis, but it is not mentioned in the Data and Methods section. This should be done, unless OMI is not part of the present analysis.

P18, L349: "A carbonaceous aerosol plume associated with wild fires in British Columbia in August 2017 reached the stratosphere a few days following the initial injection above the tropopause…" Confusing. It is stated that the plume was injected above the tropopause, but only reached the stratosphere a few days later. Both clauses are attributed to Torres et al. Please clarify.

P18, L352: "…19 to 26 kilometers is observed for a cloud consisting of sulfate aerosol." From the material presented to this point, how does the reader know it is sulfate? If compositional information has not been provided, a citation is

needed. Alternatively, the paper could just state that sulfate is assumed.

P18, L376: What are "large-scale mixing events?" Please explain using a meteorological perspective.

P18, L279: "SO2 reduction" is vague and general. Provide more detail to this description, such as "e-folding time"

P18, L383: "…the CCC circled the globe almost three times at the latitude of 30oN…" The CCC ended up almost 10 deg. south of this according to figures herein. Perhaps elaborate a little more on the CCC's slow movement even south of 30.

P18, L383: "…increased its height from 19 to 28 km." "28" is higher than shown if Fig. 14. The text in the body of the paper only gives pressure and theta. Suggestion: enhance the description of the endpoint altitude in the main text and state "26 km" or whatever actual number the authors think is represented by the figure.

P18, L387-389: This sentence is confusing, combining 3 ideas: arch effect, 45 km background, and aerosol that can pollute the background. Please flesh out these points more exhaustively.

More generally, the Conclusion section is too thin, and doesn't flow logically from points made in the body. A thorough rework of this section is called for.

Tracking aerosols and SO_2 clouds from the Raikoke eruption: 3D view from satellite observations

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Abstract. The June 21, 2019 eruption of the Raikoke volcano (Kuril Islands, Russia, 48°N, 153°E) produced significant amounts of volcanic aerosols (sulfate and ash) and sulfur dioxide (SO_2) gas that penetrated into the lower stratosphere. The dispersed SO_2 and sulfate aerosols in the stratosphere were still detectable by multiple satellite sensors for three months after the eruption. For this study of SO_2 and aerosol clouds we use data obtained from two of the Ozone Mapping Profiler Suite (OMPS) sensors on the Suomi National Polar-orbiting Partnership (SNPP) satellite: total column SO_2 from the Nadir Mapper (NM) and aerosol extinction profiles from the Limb Profiler (LP) as well as other satellite data sets. The LP standard aerosol extinction product at 674 nm has been re-processed with an adjustment correcting for limb viewing geometry effects. It was shown that the amount of SO_2 decreases with a characteristic period of 8-18 days and the peak of sulfate aerosol recorded at a wavelength of 674 nm lags the initial peak of SO_2 by 1.5 months. Using satellite observations and a trajectory model, we examined the dynamics of an unusual atmospheric feature that was observed, a stratospheric coherent circular cloud (CCC) of SO_2 and aerosol from July 18 to September 22, 2019.

1 Introduction

An eruption of the Raikoke volcano (See Fig. 1, 0.55 km altitude, uninhabited island at 48.29°N, 153.4°E) occurred on June 21, 2019 at 6 p.m. UT. The eruption was so strong that a cloud of ash and volcanic gases was ejected to a height of 17-19 km, putting a significant part of the Raikoke volcanic plume into the stratosphere and above the heights of clouds and the tropopause, which is ~ 10-12 km for such northern latitudes (see Fig. 1). The Microwave Limb Sounder (MLS) data for June 23-27, 2019 show that the observed parts of the SO_2 cloud had a height of 11 to 18 km, with a peak concentration at 14 km. The maximum volcanic cloud top height rises more than 6 km within 4 days after the eruption due to aerosol–radiation interaction (Muser et al., 2020).

The study of volcanic plumes and the transformation of their SO_2 into aerosols is interesting for several reasons. Firstly, clouds of ash and sulfate aerosol are aviation hazards. Secondly, the lifetime of SO_2 depends on its injection height, total mass, dispersion pattern and the concentrations of oxidants that vary with season and location (Krotkov et al., 2010). SO_2 from volcanic eruptions has a longer lifetime if it is injected into the stratosphere. SO_2 in the stratosphere is converted to sulfate aerosol, which interacts with the long-lived Junge layer—the naturally occurring background stratospheric aerosol layer. Further, the dynamics of the Junge layer itself is not well understood. In addition, stratospheric aerosols are an important factor for climate forcing because aerosols modify both the shortwave and longwave radiation in the atmosphere and reaching the Earth's surface (Toohey et al., 2019; von Savigny et al., 2020). Thus, each case of volcanic injection of

large amounts of SO$_2$ into the stratosphere is of great interest from the point of view of Earth sciences. Our ability to study volcanic clouds and their impacts has evolved in recent decades along with satellite technology (Bovensmann et al., 1999; Carn et al., 2003, 2007, 2008, 2009, 2017; Penning de Vries et al., 2014; Sandvik et al., 2019; Hedelt et al., 2019; Theys et al., 2019; Fisher et al., 2019). The spectral data obtained from satellites has made it possible to analyze in detail volcanic eruptions, such as of the Pinatubo (Bluth et al., 1992) and Kasatochi (Bourassa et al., 2010; Krotkov et al., 2010).
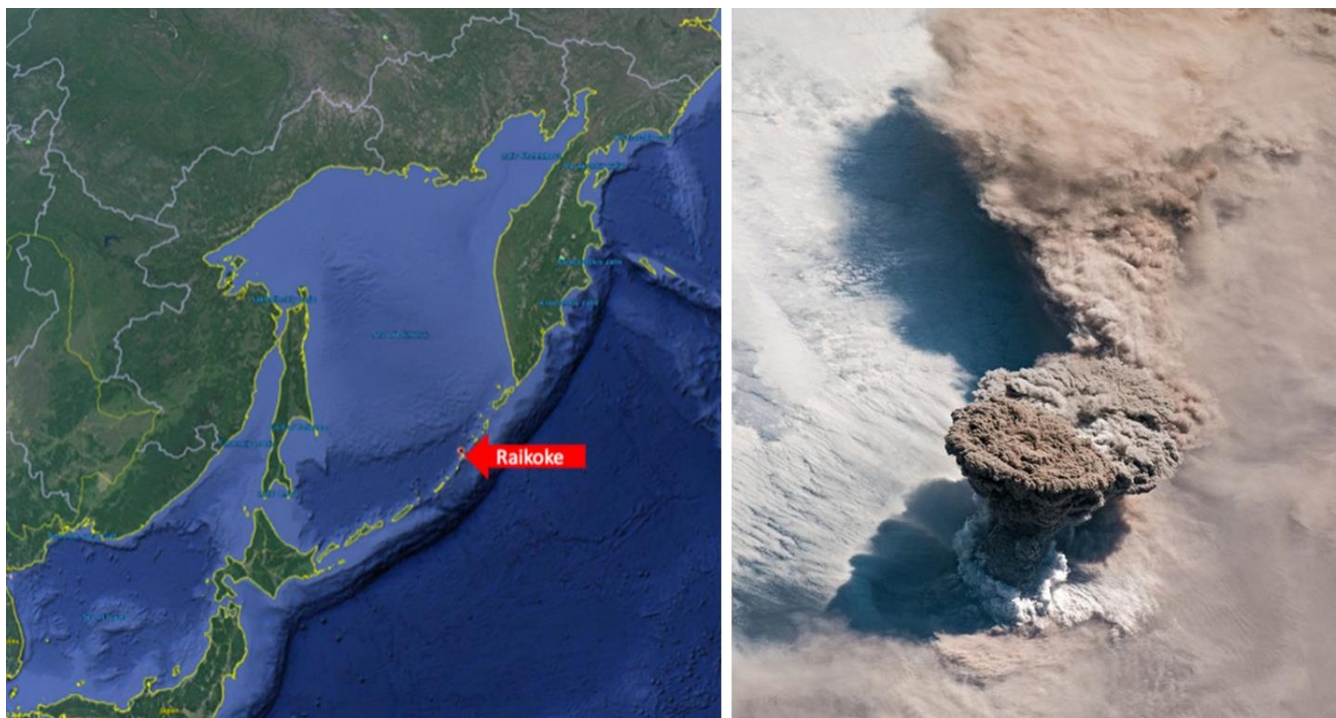
Figure 1: Left: Raikoke volcano (Kuril Islands, Russia, 48°N, 153°E) (Google Maps, Imagery: NASA; Map data: SK telecom)). Right: photo of the Raikoke eruption (06 2019), which shows that the volcanic plume rose much higher than the layer of tropospheric clouds (Photo: iss059e119250 June 22, 2019, ISS/NASA, https://visibleearth.nasa.gov/images/145226/raikoke-erupts/145230w)

The 2019 eruption of Raikoke was well observed by a number of satellite sensors, for example, the Ozone Monitoring Instrument (OMI), the Tropospheric Monitoring Instrument (TROPOMI), the Ozone Mapping Profiler Suite (OMPS) Nadir Mapper (NM) and Limb Profiler (LP) and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). Studying the nadir data of OMPS NM and TROPOMI, our attention was drawn to an interesting phenomenon in the presence of compact stratospheric SO$_2$ clouds, hereafter referred to as coherent circular clouds or CCC, each about 300 km in diameter: the first noted on June 29, 2019 centered at 52°N, 157°W (near Alaska) and the second observed starting on July 21, 2019 centered at 46.5°N, 167°E (near Kamchatka). The first of these moved towards the North Pole while the second moved to ~ 30°N, where it remained for more than two months, having made almost three complete revolutions around the Earth.

A dense cloud of SO$_2$ and aerosol that formed after the eruption of the Raikoke volcano spread over a week at latitudes above 40°N (see TROPOMI SO$_2$ map on June 29, 2019 in Fig. 2). Atmospheric currents stretched the cloud into long jets, twisted them with spirals and even formed CCCs, as shown in Fig. 2 in the region near the coordinates 52°N, 156°W. To analyze in detail the evolution and dynamics of the Raikoke volcanic cloud, we examine 1) the amount of SO$_2$ and aerosol emitted during the eruption using nadir and limb data; 2) the mutual evolution of SO$_2$ and aerosol for 100 days, starting from

the moment of the eruption with an adjustment correcting for limb viewing geometry effects; 3) the propagation of the CCCs with a case study near the end of July 2019 using a trajectory model.
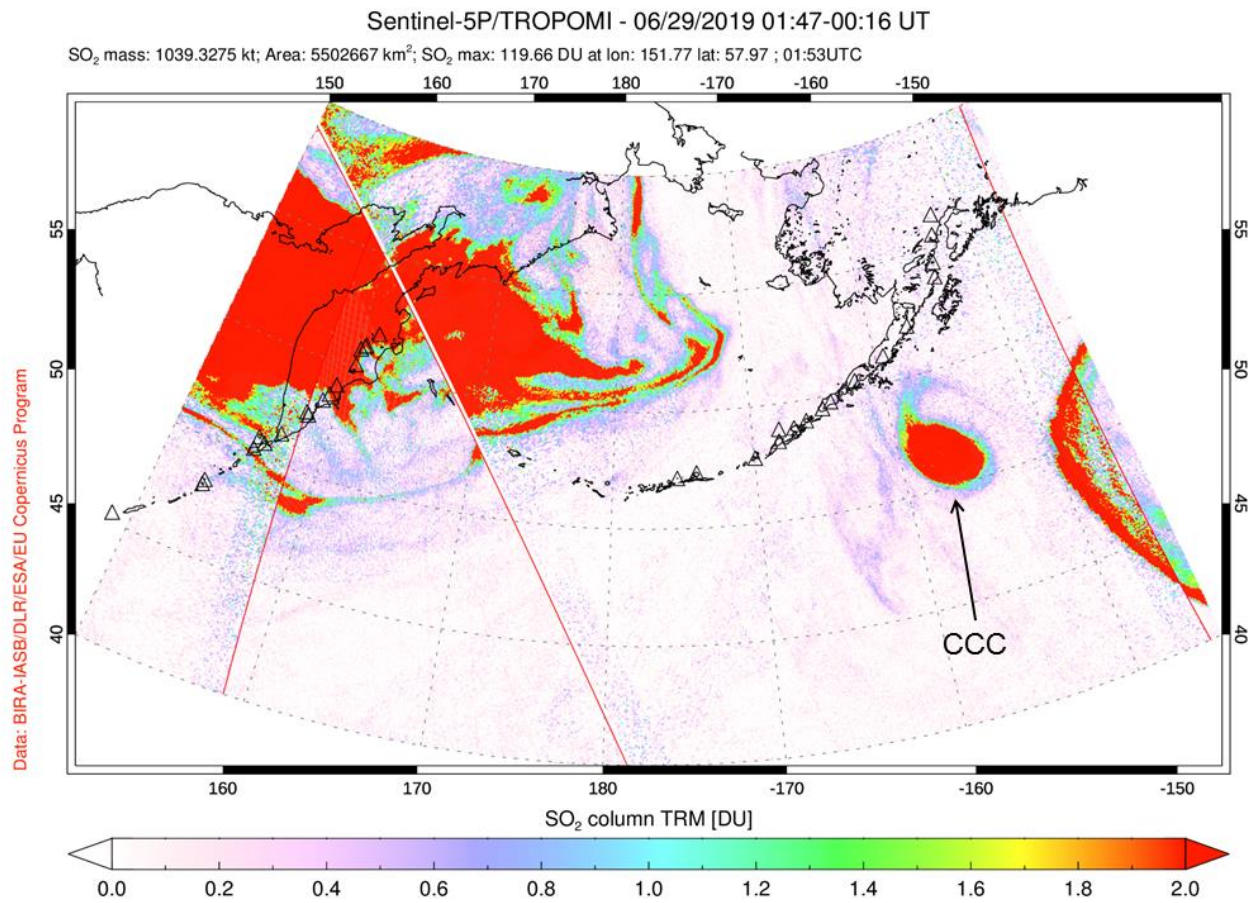
70

Figure 2: TROPOMI image of SO$_2$ clouds, a week after the Raikoke eruption (Image: courtesy Copernicus
75 **TROPOMI/Sentinel-5Precursor SO$_2$ data, https://so2.gsfc.nasa.gov/). The triangles represent the location of the**
volcanoes.

2 Data and Methods

In this section, we detail the instruments and methods used to 1) examine SO$_2$ and aerosol distributions obtained from nadir
80 for SO$_2$ and limb and lidar for aerosol; 2) calculate zonal mean values of SO$_2$ and aerosol as a function of a height using
nadir data for SO$_2$ and limb data for aerosol; 3) employ trajectory modeling for analysis of individual clouds.

3

2.1 Satellite Mapping of SO_2

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We use SO_2 total column data obtained by two satellite spectrometers: 1) TROPOMI (see Fig. 2, Theys et al., 2017, 2019) on the Sentinel 5 precursor satellite and OMPS Nadir Mapper (NM) on the Suomi National Polar-orbiting Partnership, SNPP, satellite (Li et al., 2013, 2015; Zhang et al., 2017). Both instruments observe the Earth's backscatter radiance and solar irradiance from visible through ultraviolet wavelengths with two-dimensional charge-coupled devices (CCDs), measuring in

90 the spectral domain in one dimension and in the spatial domain (across a satellite track) in the other dimension. The satellite motion provides measurements along the satellite track. The spatial resolution of TROPOMI (3.5 km by 7 km) is much finer than that from OMPS measurements made in the nominal mode (50 km by 50 km) and the spectral resolution is higher for TROPOMI as well (~ 0.5 nm as compared with ~ 1 nm for OMPS). The satellite swaths are wide (2700 km for TROPOMI and 2800 km for OMPS), providing nearly daily global coverage. SO_2 is retrieved with backscatter solar spectral

95 measurements at ultraviolet wavelengths (312-390 nm). TROPOMI uses a spectral fitting algorithm based on differential optical absorption spectroscopy (DOAS) (Theys et al., 2017, 2019). OMPS uses spectral fitting approach with a principal component analysis (PCA) scheme (Li et al., 2013).

2.2 Aerosol profile data

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We use aerosol data from CALIOP (Fairlie et al., 2014) and OMPS LP (Loughman et al., 2018; Chen et al., 2018). The height of an aerosol cloud can be estimated with both OMPS LP and CALIOP. We use the CALIOP lidar images for aerosol, specifically 532 nm total attenuated backscatter signal with a spatial resolution of 40 km along track and a vertical resolution of 120 m (altitude < 20 km) and 360 m (altitude > 20 km).

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OMPS LP hyperspectral measurements capture aerosol data with a sampling of ~1 km in altitude. OMPS LP views the atmosphere in a backward direction along the orbit track with three vertical slits, one (central) aligned with the orbit track and the other two (left and right) separated by ±4.25° horizontally. Here, we use data from version 1.5 (Chen et al., 2018).

110 Limb measurements of the atmosphere that view an altitude H at the tangent point also view altitudes above H in the foreground and background of the line of sight. However, the converse situation is also true: an object at a fixed altitude H will appear to be at a lower altitude h if it is located closer or farther than the tangent point. This is shown schematically in Fig. 3, where a single cloud is located at true altitude H and 5 successive LP measurement events (each event generates a new profile) are illustrated. For event C, the cloud location is at the tangent point, and the apparent altitude h equals the true

115 altitude H. For events A and B, the cloud position is closer to the LP instrument than the tangent point, and h is less than H. For events D and E, the cloud position is farther from the LP instrument than the tangent point, and again $h < H$.

Plotting successive altitude profiles of LP signal strength (e.g., extinction coefficient) when a vertically limited feature (such as a cloud) is present gives the impression of an "arch" in the data. Figure 4, where extinction values are color-coded for

120 intensity, and a lower threshold of 1×10^{-3} km^{-1} is applied, illustrates this effect. The maximum values (yellow) appear to vary in altitude by 2-3 km due to the projection effect shown in Fig. 3. Note that this effect will be present for high, thin clouds such as cirrus clouds as well as for aerosol plumes. If we believe that these lower altitude values do not represent a true aerosol signal, we need to apply a correction in order to accurately determine overall aerosol loading.
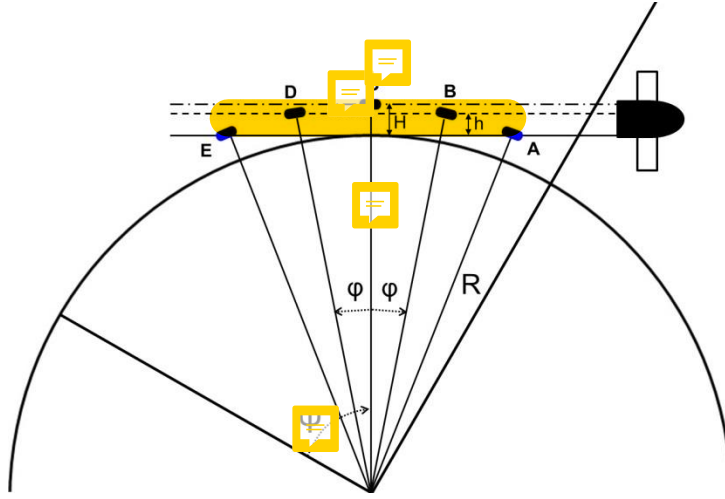
Figure 3: The diagram for observations of the limb sensor, showing the relationship between the observed (h) and real (H) heights of five clouds, as well as φ - angular displacement of the position of the clouds.

The relationship between true cloud height H and apparent cloud height h in Figure 3 is given by a simple function (DeLand and Gorkavyi, 2021):

$$\cos \varphi = \frac{R+h}{R+H} \qquad (1)$$

where R is the radius of the Earth; φ is the angular displacement. Table 1 gives some specific examples of the angular displacement predicted for specific combinations of H and h. This displacement is approximately equal to a shift in latitude for a sun-synchronous satellite such as S-NPP (inclination = 98.9°) outside the polar regions. From this table, we see that for a true cloud at $H = 25$ km, the apparent cloud signal at h = 15 km would be displaced by ~3.2° both earlier and later along the orbit. This total separation of ~6.4° is consistent with the "arch width" at 15 km shown in Fig. 4. Note also that because of differences in overall path length, we expect events A and B in Fig. 3 (left branch of "arch" in Fig. 4) to have a stronger signal than the corresponding events D and E at the same apparent altitude. We can therefore use Eq. (1) to calculate and apply a correction for determining the magnitude and position of an aerosol cloud.

Table 1. The visible height h and angular displacement φ for the cloud with real height H.

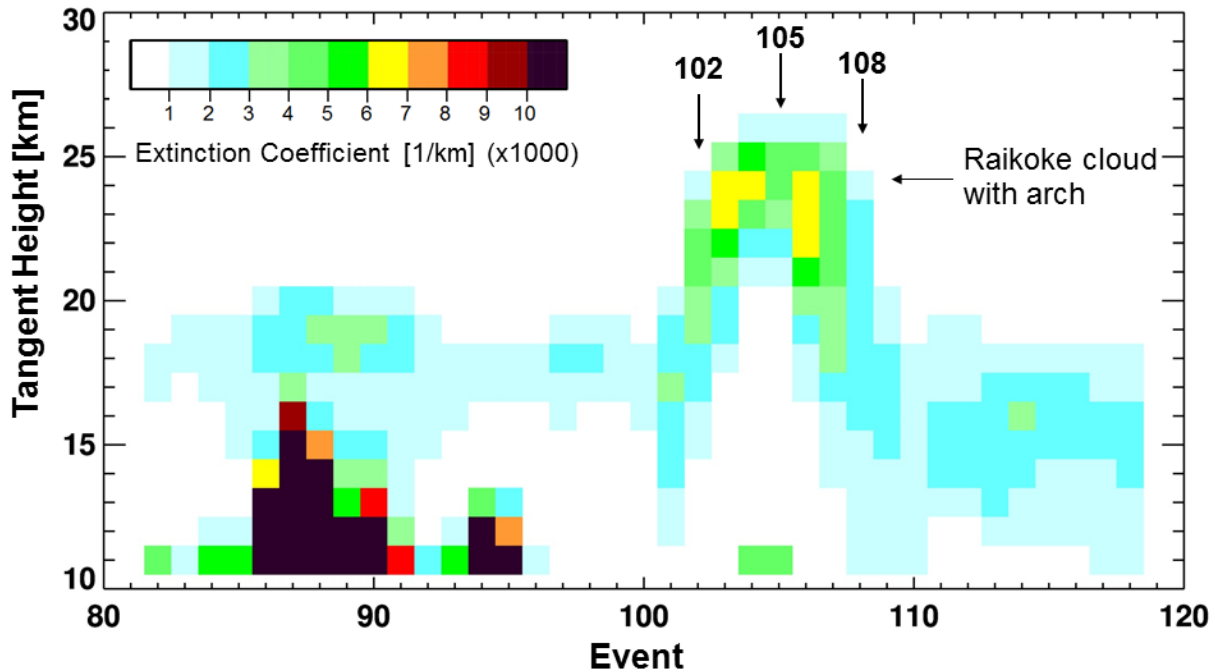| $h\backslash H$	$H =$10 km	15 km	20 km	25 km	30 km
$h = 0$ km	$\varphi = 3.21°$	3.93°	4.53°	5.07°	5.55°
5 km	2.27°	3.21°	3.93°	4.53°	5.07°
10 km	-	2.27°	3.21°	3.92°	4.53°
15 km	-	-	2.27°	3.20°	3.92°
20 km	-	-	-	2.27°	3.20°
25 km	-	-	-	-	2.26°

5

Figure 4: The extinction coefficient profiles (674 nm) from OMPS LP for the orbit 40636 (central slit, same orbit as in Fig. 6, which shows the profiles for events 102, 105, 108) for August 31, 2019. For accurately calculating the extinction coefficient it is necessary to take into account the arch effect, which is clearly visible (OMPS LP data).

150 The arch in Fig. 4 is formed by several measurements of one CCC, which has a height of 23-25 km (see the analysis of this CCC in Sect. 3.2). Parts of the arch (between events 101 and 109) below 23 km are artifacts of multiple registrations of the same object and should be removed when calculating the total amount of aerosol at given latitudes or heights. The aerosol layer at an altitude of 18-20 km with a decrease to the north to 15-16 km is the Junge layer. It merges in frames 101-109 with a volcanic cloud arch that partially shields the Junge layer in frames 103-106. The tropical cloud in frame 88 is up to 16 km

155 altitude.

The principle of our posterior algorithm for neutralizing the "arch effect" is as follows: the data in the uppermost pixel of the arch are considered real. For a given pixel, artifact densities are calculated in pixels in the side branches of the arch. These artifact densities are subtracted from the initial arch density. The procedure is repeated for the second highest pixel in the

160 already modified arch image. This continues until the side branches of the arch completely disappear. The top pixels with the corrected density are summed into the cloud with the cleaned data.

The arch effect is a specific example of the effects of inhomogeneity along the line of sight that is an issue for all types of limb sounders. One way to account for such effects is to use a radiative transfer scheme that is able to model such effects and

165 then to use multiple observations in a tomographic retrieval (e.g., Livesay et al., 2006; Zawanda et al., 2018). Such an approach with OMPS LP aerosol profiles was discussed by Loughman et al. (2018) but has not yet been implemented. Instead, we have developed a posterior adjustment method that is effective in correcting for the arch-type effects that appear with isolated features within the lines of sight for a series of observations.

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2.3 The NASA ftraj trajectory model

175 The "ftraj" trajectory model from NASA's Goddard Space Flight Center Atmospheric Chemistry and Dynamics Laboratory uses a fourth-order Runge-Kutta integration scheme to track parcels isentropically, with optional diabatic adjustments (see Schoeberl and Sparling, 1995). The model is driven with winds at 1/4° horizontal resolution and spaced every six hours apart from the Goddard Earth Observing System (GEOS) forward processing system produced by the NASA Global Modeling and Assimilation Office (GMAO). Each model run was initialized with 3000 parcels distributed randomly in a cylinder of radius 150 km centered about the location of the observed cloud and stretching between the lowest and highest potential

180 temperature (θ) value in the observed cloud. Those θ values were derived by interpolating the GEOS fields to the parcels' initial longitude, latitude, altitude, and time. Both the starting and ending cloud observations extended through a range of altitudes and hence θ values.

3 Results

185 The sensitivity of the satellite data we examined is such that the aerosol cloud from the Raikoke volcano was observable for 3 months following the eruption. Thus, a synergistic study of Raikoke volcanic emissions using various satellite-based instruments provides a good opportunity to study the dynamics of volcanic cloud dispersion. For quantitative analysis, we will use 2D SO_2 vertical column density (VCD) maps obtained by OMPS NM, as well as vertical profiles of aerosol clouds that are obtained from the OMPS LP.

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3.1 SO_2 and aerosol evolution

The retrieved Raikoke SO_2 mass increased rapidly following the dispersion of the volcanic cloud after the eruption and then exponentially decayed with an average e-folding time scale of 8-10 days. The amount of stratospheric SO_2 decreases due to

195 photochemical conversion to sulfuric acid though gas - phase reaction with the hydroxyl radical, OH. Sulfuric acid nucleates new particles and condenses onto pre-existing particles to form long-lived stratospheric sulfate aerosol. Figure 5 shows the change in the total SO_2 mass (kt) retrieved by the OMPS NM and aerosol extinction coefficient retrieved with the OMPS LP in the latitude range 45°-65°N after the Raikoke eruption. The aerosol extinction data in Fig. 5 are shown before and after removing the "arch effect". As can be seen from Fig. 5, the aerosol extinction coefficient after removing the arch effect is
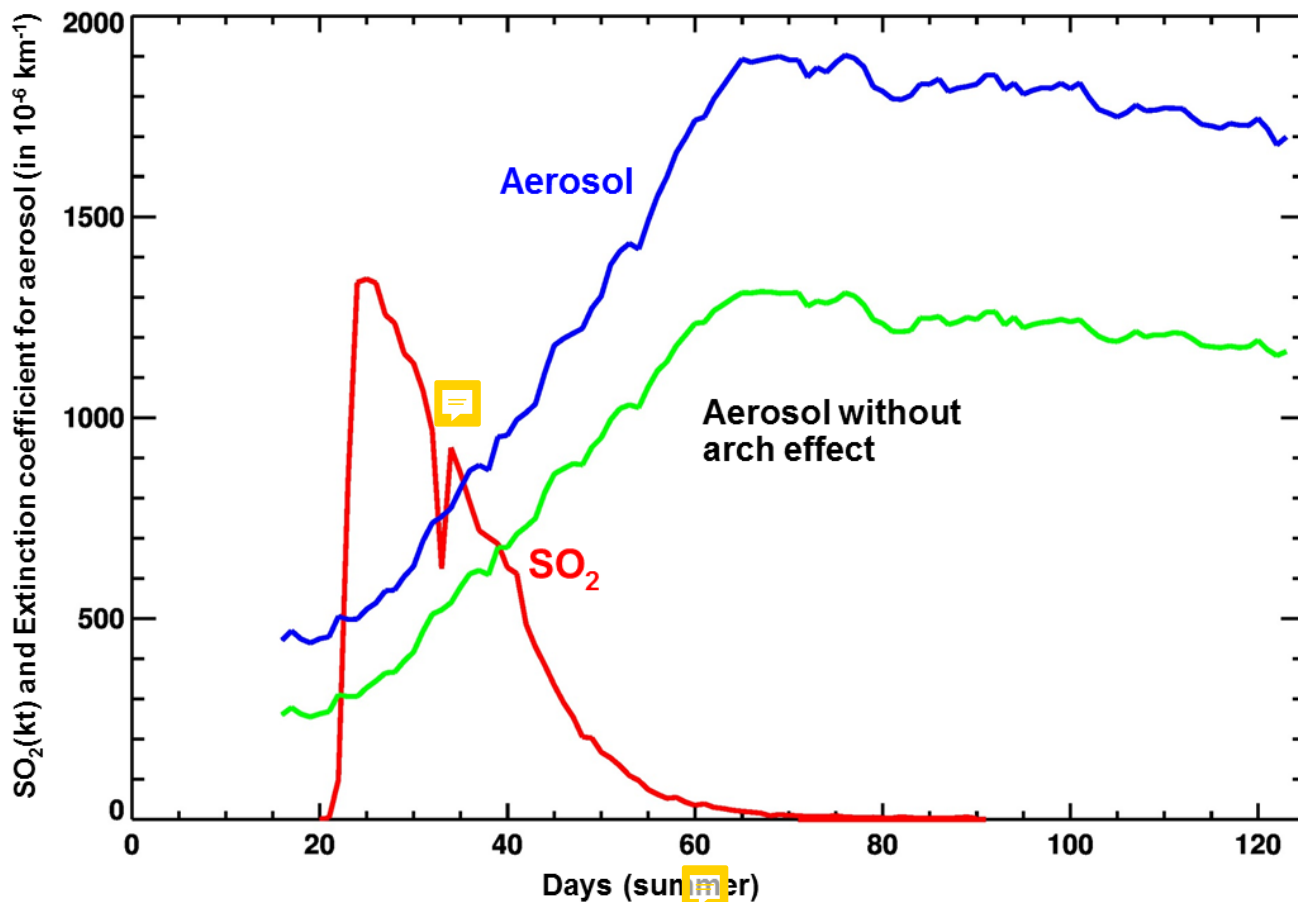
200 reduced by 30-50%.

Figure 5: The daily zonal mean (45-85N) SO$_2$ mass (assuming a cloud height of 13 km) and the aerosol Extinction coefficient at 675 nm (13-18 km.). The retrieved SO$_2$ mass increases for 2 days after the eruption during rapid dispersion of the fresh opaque volcanic cloud. Following initial dispersion and fallout of ash the SO$_2$ mass decreases exponentially due to chemical conversion to sulfate. Aerosol extinction has opposite behavior: it increases by a factor of 4 as the concentration of SO$_2$ decreases, reaching a maximum ~50 days after the eruption. After that, the aerosol extinction starts to decrease due to gravitational sedimentation, but very slowly (from OMPS LP and NM data).

210   As can be seen from Fig. 6, the cloud SO$_2$ mass decreases exponentially with variable (8-18) day timescale. Aura/OMI previously measured volcanic SO$_2$ mass decay following the August 8, 2008 Kasatochi eruption (30°N–90°N). The overall apparent e-folding time for Raikoke is ~ 8-10 days (Fig. 6, solid black lines), in good agreement with Kasatochi e-folding time ~9 days (Krotkov et al., 2010). But there are uncertainties associated with the estimated e-folding time, as discussed below.
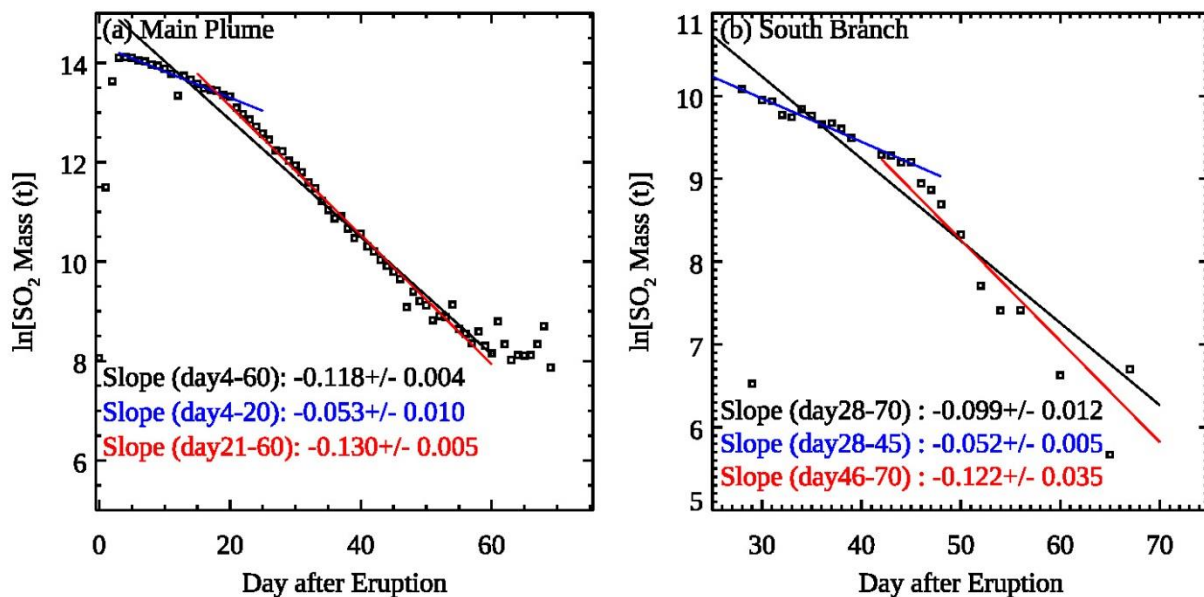
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Figure 6: Time series of the logarithm of the estimated total SO$_2$ mass (in ton) within (a) the main plume and (b) the CCC to the south after the eruption. Assuming a first order loss rate for SO$_2$, the slope of the linear fit (straight line) for (a) indicates a decay rate of 0.118 day^{-1}, or an e-folding lifetime of ~8.5 days. Similarly, for (b), the decay rate is 0.099 day^{-1} and the e-folding lifetime is ~10 days. There are uncertainties in the estimated lifetime. For (a) the main plume, the estimated lifetime is ~18.9 days between 4 and 20 days after eruption, and ~7.7 days between days 21 and 60 after the eruption. For the south branch (b), the estimated lifetime is ~19.2 days between 28 and 45 days after eruption, and ~8.2 days between days 46 and 70 after the eruption.

Note that the left panel in Fig. 6 shows the following deviations from the average of the e-folding lifetime of 8.5 days: within the first 20 days after the eruption, the SO$_2$ mass falls with an apparent e-folding lifetime of 18.9 days, and between days 21 and 60, the decrease in SO$_2$ mass accelerates and the apparent e-folding lifetime is only ~8 days. The e-folding lifetime of 18.9 days, derived from the earlier stage after the eruption, probably reflects the conversion of SO$_2$ into sulfate aerosol. After that, the detection limit of the instrument becomes a more important factor, leading to faster apparent decay of the total observed SO$_2$. As the plume spreads out to larger areas, and more and more pixels with SO$_2$ fall below the detection limit of the OMPS NM sensor, the apparent decay rate becomes larger, likely reflecting the effects of both actual SO$_2$ loss and the diminished OMPS sensitivity to weaker SO$_2$ signals. Figure 6b shows a similar pattern for change in SO$_2$ in the CCC. For the period between days 28 and 45 after eruption, the e-folding lifetime is ~19 days. This suggests that the time after the eruption does not play a big role here, only the compactness of the CCC with a high level of SO$_2$ is essential. As in case Fig. 6a, the second phase of the evolution of the CCC is characterized by a faster apparent (the e-folding lifetime ~ 8 days) rate of decrease in the recorded SO$_2$.

3.2 Stratospheric aerosol from limb observations

The stratospheric aerosol optical depth shows the plume latitudinal distribution, with most of the aerosol being transported poleward (Fig. 7). Figure 7 also shows that the effects of the eruption lasted for almost a year. Increased aerosol loading in the lower stratosphere can also be attributed to two pyroCumulonimbus (pyroCb) events that took place before and after the eruption, Alberta fires (June 18) and Siberian fires (July 2) (Kloss et al., 2021). The zonal mean aerosol extinction profile

240 between 20˚N and 90˚N shows the vertical transport of the Raikoke plume to higher altitudes and its persistence in the lower stratosphere (Fig. 8). The top panel in Fig. 8 shows that the maximum altitudes of the plume are around 25 km, when the plume penetrates the tropics.
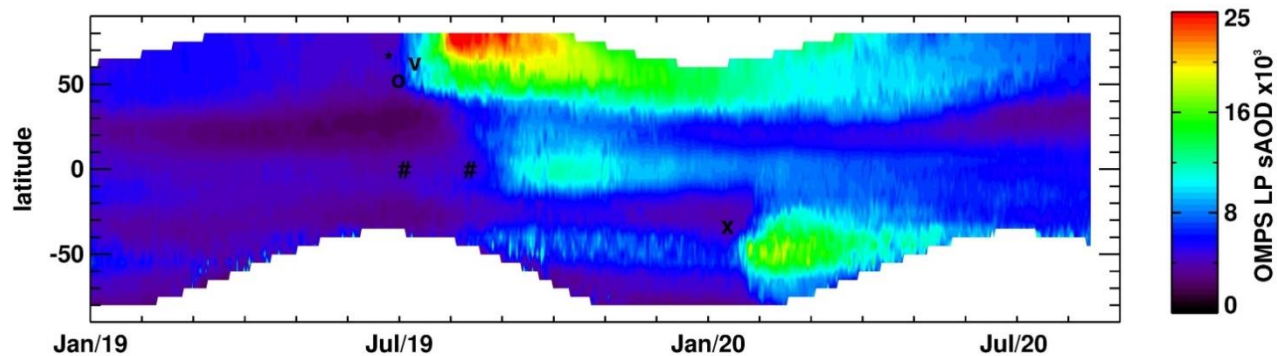
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Figure 7: Latitudinal distribution of the stratospheric aerosol optical depth (x 10^3, sAOD) at 675 nm. "o" – Raikoke eruption, "*" – Alberta fires, "v" – Siberian fires, "#" – Ulawun eruptions, "x" – Australian fires. The sAOD daily time series were derived by integrating aerosol extinction profiles derived from OMPS LP above the tropopause to 35 km and averaged every five degrees of latitude

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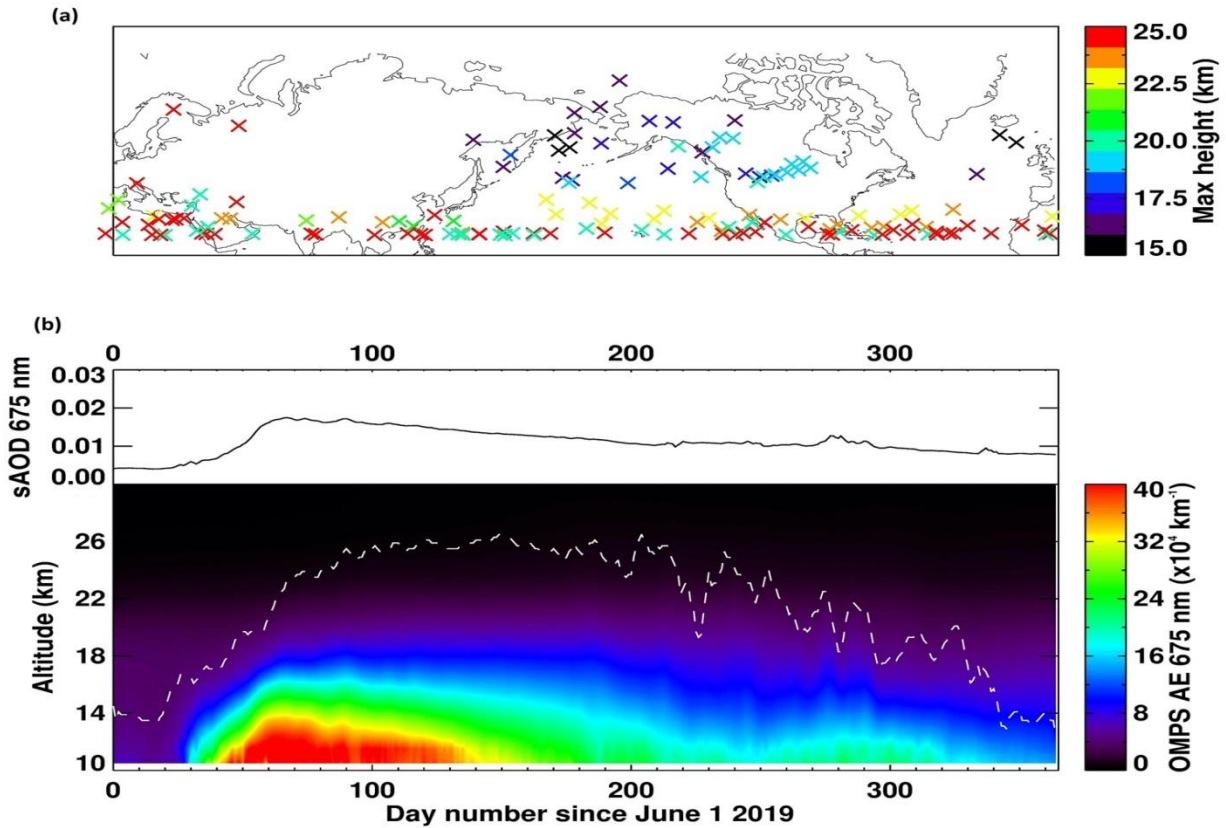
Figure 8: (a) Daily location of the maximum altitude reached by the Raikoke aerosol plume as detected by OMPS LP. The color scale shows altitude values in km. Figure 8(b): (bottom) Daily zonal mean aerosol extinction profiles at 675 nm (km^{-1}) between 20°S-90° measured by OMPS LP from June 2019 to June 2020 and smoothed spatially using a 5 point boxcar averaging. Only profiles measured above tropopause+1 km are used. The white line is the aerosol plume maximum altitude in km.

Figure 9 shows the ratio of a daily zonal mean of the aerosol extinction coefficient for July 1 and 21 and September 1 and 30, 2019 to a similar daily zonal mean for a quiet period (June 16-20, 2019) before the eruption. The means were computed without taking into account the "arch effect" (see DeLand and Gorkavyi (2021) and Sect. 2.2). Therefore, the lower parts of the layers shown may have an overestimated extinction coefficient. In addition to increasing the density of the Junge layer (the contribution of seasonal changes is possible here), an interesting CCC appears at an altitude of about 25 km that will be discussed in Sect. 3.3.
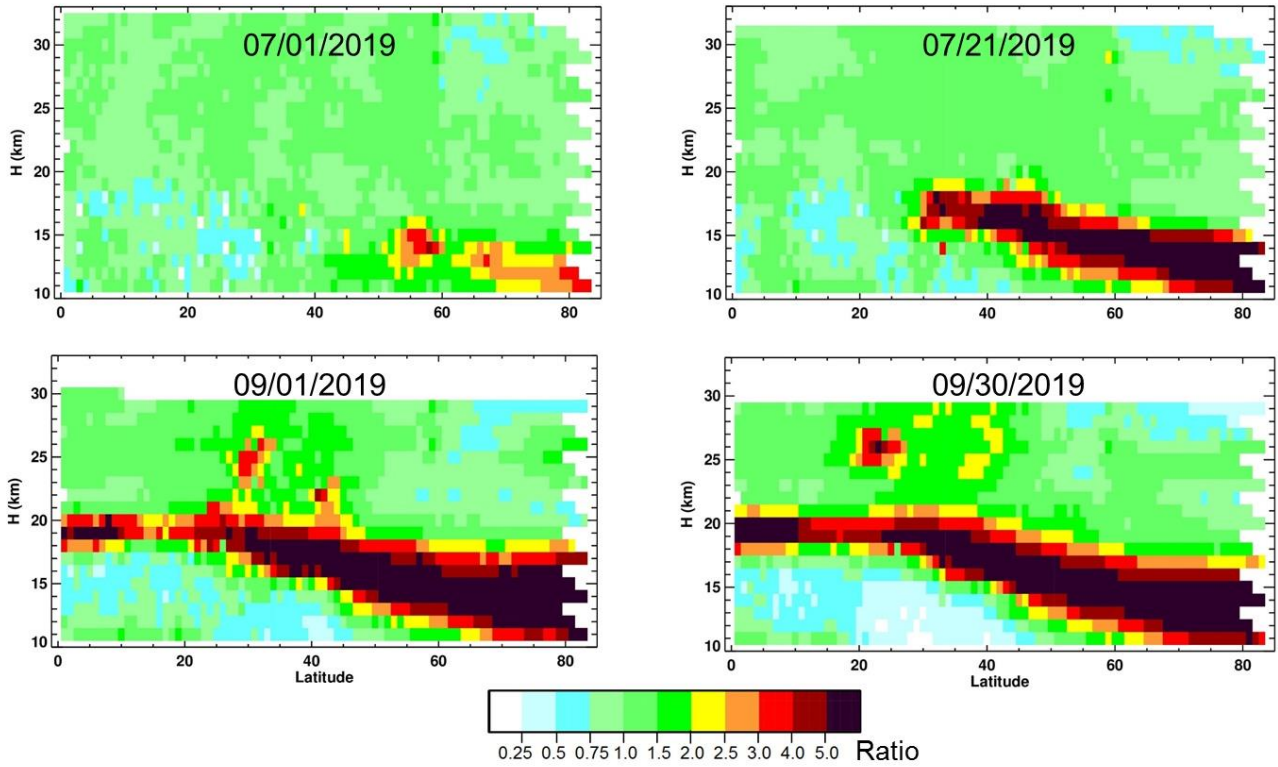
Figure 9: The ratio of the zonal mean, daily average aerosol extinction for the indicated dates after the Raikoke eruption to the averaged aerosol extinction immediately prior to the eruption shows the evolution of the volcanic aerosol in the Junge layer (13-20 km) and the occurrence of the CCC near 25 km from OMPS LP data.

Three LP aerosol extinction profiles were measured by OMPS LP on 31 August 2019, more than two months after the Raikoke eruption (Fig. 10). The extinction peak at 22-25 km represents the continued signal of the stratospheric plume from the eruption. The apparent altitude variation between these peaks also shows the impact of the LP viewing geometry on the retrieved extinction profile. Each measured profile (also termed "event") gives an along-track separation of ~125 km. The along-track field of view integrates over a distance of ~180 km for each 1 km vertical sample (DeLand and Gorkavyi, 2021).
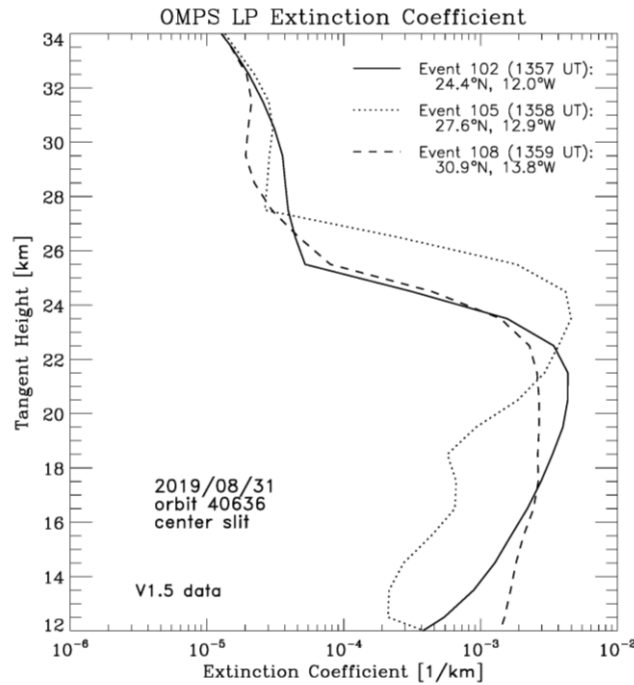
Figure 10: The aerosol formed as a result of the Raikoke eruption is clearly visible in the extinction coefficient profiles obtained by the OMPS LP aerosol extinction vertical profile months after the eruption.

3.3 Coherent circular cloud (CCC)

The CCC is part of the original aerosol/SO_2 cloud resulting from the eruption of the Raikoke volcano. It formed one month after the eruption in the same area where the volcano is located (Fig. 11). On July 24, 2019, as the CALIOP aerosol data show, the CCC had a height of 19-20 km and quickly shifted to the south (Fig. 11). On July 30, 2019, it reached a latitude of 30˚N and was extended across China, fell into easterly winds and moved west at about 17 m/s (Fig. 12). On August 4, it was recorded over the Persian Gulf region, on August 5 - over the Egyptian region, and on August 8 - near the Azores.
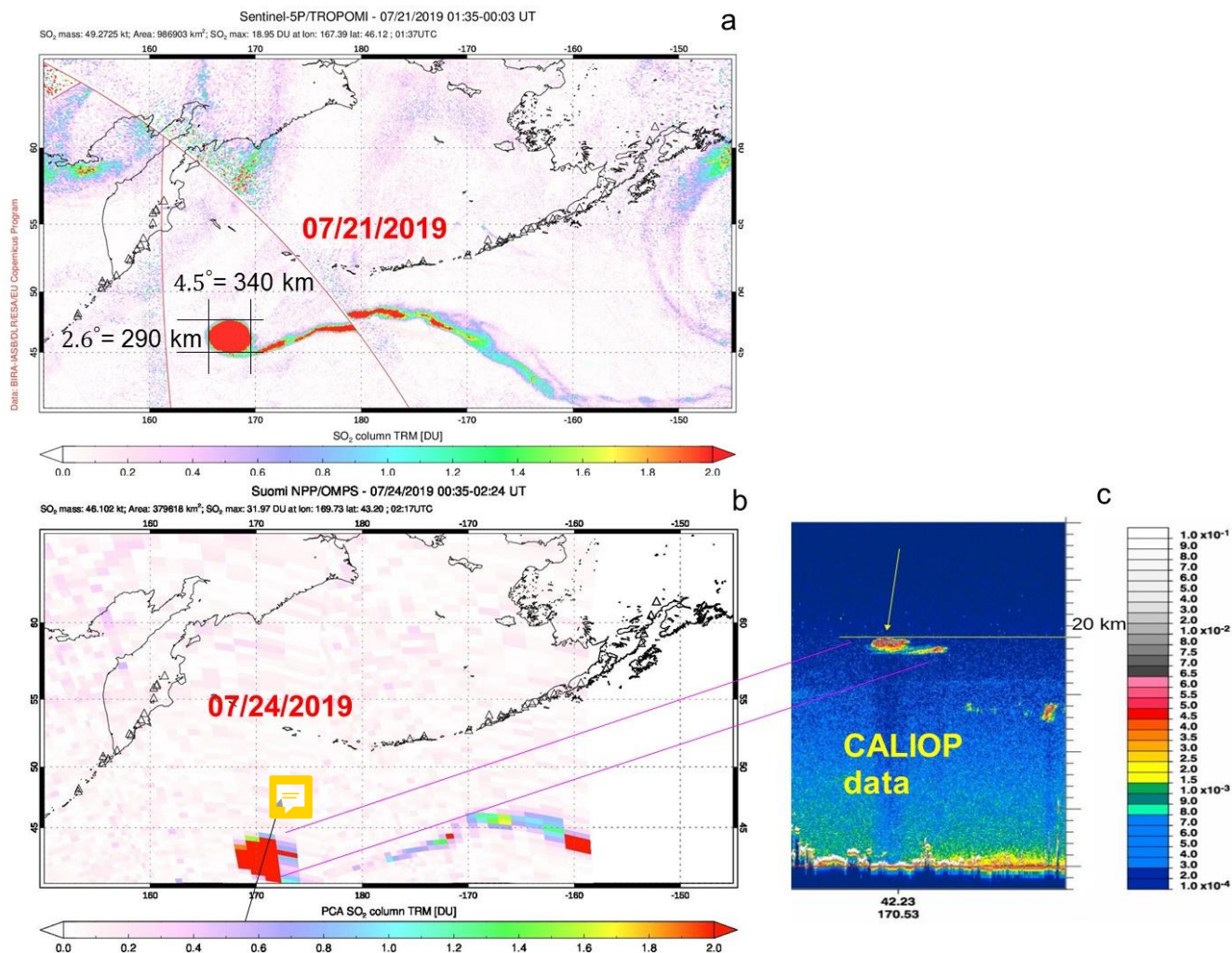
13

Figure 11: a-b) TROPOMI and OMPS NM SO$_2$ data for 07/21/2019 and 07/24/2019 show the formation of CCC, which appeared near Kamchatka and shifted to latitude ~ 30˚N and moved to west at a speed of 1400 km per day. From OMPS NM data we estimated the initial mass of SO$_2$ in this cloud to be ~20 kt; **c)** CALIOP total attenuated backscatter data (km^{-1} sr^{-1}) show the height of the CCC to be 18.5-20 km.
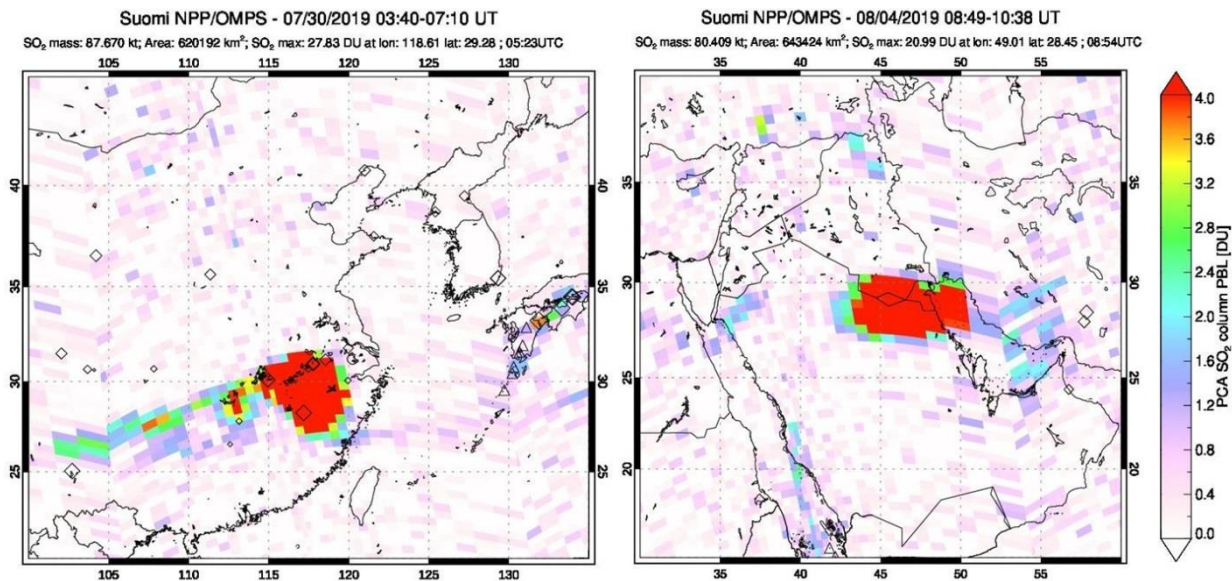
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Figure 12: SO₂ column data shows the westward drift of the CCC with speed ~ 14-15°/day, ~ 1400 km/day, ~ 60 km/h.

300   The CCC was tracked until the end of August, while making 1.5 orbits around the Earth. CALIOP and OMPS LP (see Fig. 13 with CALIOP data from August 18 and September 1, 2019 and Fig. 4, 10 from August 31, 2019 – OMPS LP data) were able to track the movement of the accompanying aerosol part of this CCC up to September 22 as it made ~ 3 orbits around the Earth.
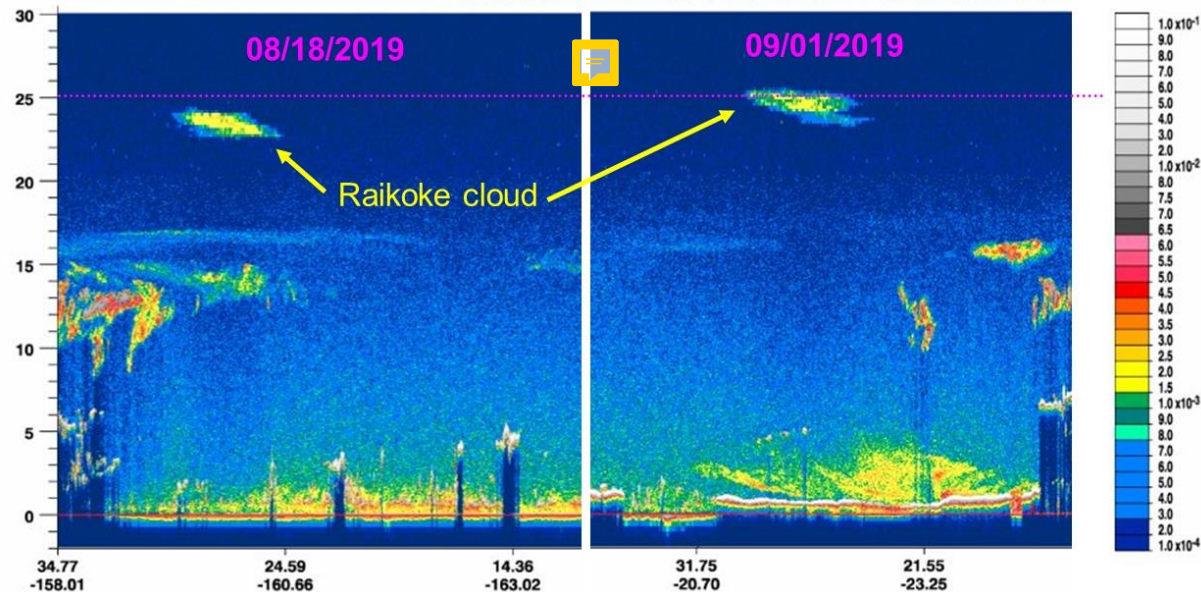

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Figure 13: The CALIOP total attenuated backscatter data (km⁻¹ sr⁻¹) for 08/18 and 09/01/19, where bright aerosol layers are visible at the indicated latitudes and heights show an increase in the height of the layer over time.

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Figure 14 shows satellite observation of the CCC from July 18 o September 22, 2019. Figure 15 shows the results of
310 backward trajectory modeling of the CCC for one month period with initial days August 18 and September 19, 2019. The basic idea is to use the trajectories to see if this long-lived CCC can be correctly transported using assimilated meteorological observations.
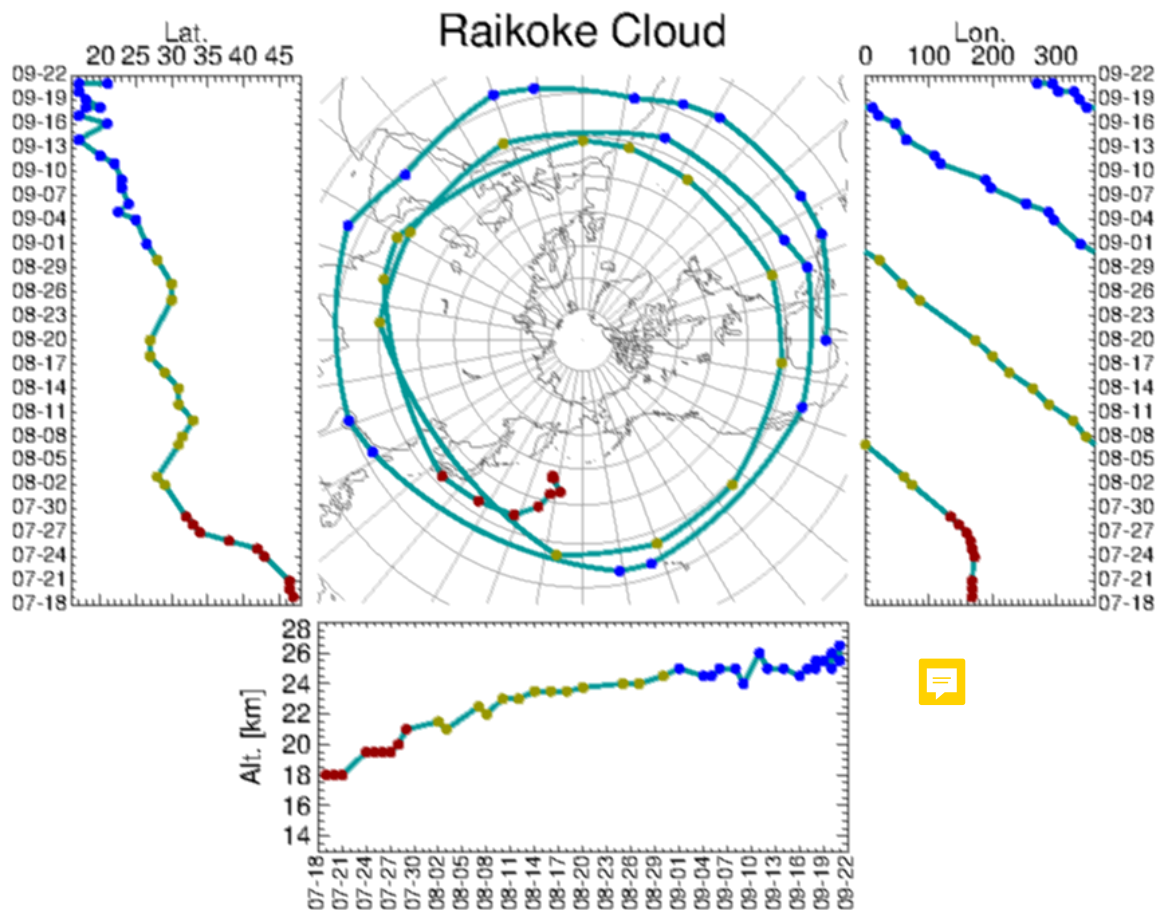
315 **Figure 14: The observed path of the CCC as inferred from OMI/OMPS/TROPOMI and CALIOP observations. The individual observations are shown by dots color-coded by month.**

We have two hypotheses to evaluate. The first hypothesis ("the great red spot" hypothesis) is that the observed CCC is contained within a gyre (whirlpool) that existed from around the time of the eruption on June 21 to September 1 or even later, advected around the globe by the large-scale circulation. The second hypothesis (the "dead fish" hypothesis) is that the
320 CCC was simply advected around by the large-scale winds without being sheared apart because the environment of the summer stratosphere is fairly quiescent.

We inspected the meteorological fields from the GMAO GEOS (Rienecker et al., 2008) and from the Global Data Assimilation System (GDAS) by the National Centers for Environmental Prediction (NCEP) at the location and altitude of
325 the observed volcanic CCC on July 21 and 24; the winds show no discernible gyre. This is to be expected, as the satellite data that go into these assimilation products are unlikely to pick up features on sufficiently small scales that they would show up clearly and prominently in the winds. Therefore, it is not possible to test the "great red spot" hypothesis using a trajectory model with the wind products available to us.
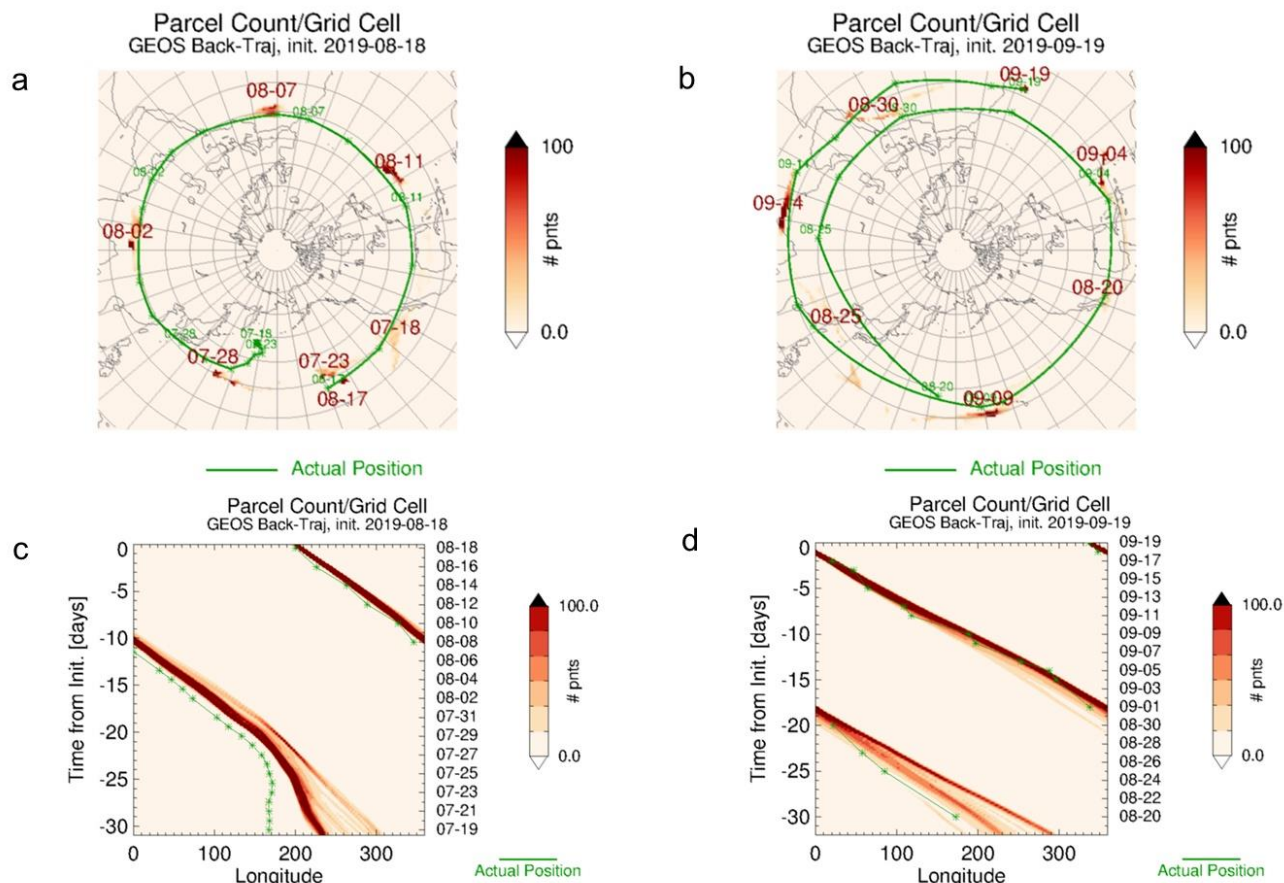
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Figure 15: Backward trajectory modeling: a) This panel shows several discrete time snapshots from the Goddard "ftraj" back-trajectory modeling for 3000 parcels within a 150 km radius of the observed CCC position on initialized on 2019-08-18; **b)** The trajectory model run was initialized with the observed cloud position on 2019-09-19 and then run backwards to 2019-08-18; **c)** This shows the same model results as for a) but using a different visualization. Here the parcels are binned and counted in longitude and time, regardless of altitude or latitude; **d)** Similar to c), except that the model run is initialized at 2019-09-19 and ran backwards to 2019-08-18.

We can examine the "dead fish" hypothesis; if a trajectory model run shows a cloud of parcels maintaining its integrity over a long period of time, then this hypothesis becomes plausible. There are difficulties, however. First, the winds used to drive the trajectory model have spatial and temporal resolutions that are coarse enough that trajectories cannot be modelled with perfect accuracy. One can expect that a cloud of parcels will lose many of its parcels along the way. It would be unrealistic to expect to duplicate the volcanic cloud's integrity and position as a function of time. Instead, the test is whether the simulated cloud of parcels can maintain its existence over the course of a month or more during this time period. Second, a more serious problem is that of altitude. We can expect a volcanic CCC to experience self-lofting, and the CCC indeed is observed to increase in altitude with time. However, the winds vary with altitude and thus the parcel trajectories will depend critically on getting their altitude changes right.

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A carbonaceous aerosol plume associated with wild fires in British Columbia in August 2017 reached the stratosphere a few
350 days following the initial injection above the tropopause and resulting from self-lofting triggered by solar heating (Torres et
al., 2020). The case of a volcanic CCC, which we are considering, is interesting in that the effect of an increase in height
from 19 to 26 kilometers is observed for a cloud consisting of sulfate aerosol. In this case, the main increase in the height of
the CCC occurred during its movement at approximately the same latitude. Early on (July 19) it is at 80 hPa (450 K), but by
September 1 it had risen to 30 hPa (590 K). This is a diabatic self-lofting heating rate of about 3 K per day. Note that this
355 CCC, increasing its height, simultaneously contained a detectable amount of SO_2, which is also confirmed by independent
satellite data from ACE (Cameron et al., 2021).

The thermodynamic processes within the cloud are not well-known, so rather than attempting to model the cloud's self-
lofting and the overall vertical advection, we impose an approximation to the cloud's vertical motion. We determined an
360 average daily θ adjustment that would map the column of parcels from its initial θ range to its final range. Back-trajectories
were then computed isentropically for one day at a time. After each day, the parcels' potential temperature values were
changed by their daily adjustment. In this way, the vertical motion of the CCC is guided by observations, while the
horizontal motions are unconstrained. Note that this adjustment procedure implies two assumptions: that the self-lofting is
linear in time, and that it is linear in θ as well. The map (see Fig.15) displays the parcel population count as a "heat map" or
365 density display: the more intense the color at a given point, the more parcels are near that point (the resolution of the map is
1° x1^o). The plot below the map shows a similar "heat map" of parcel population count as a function of longitude (x-axis)
and altitude (y-axis); the resolution here is 1° longitude by 0.5 km altitude.

Figure15a superimposes several discrete time snapshots from the trajectory model output, starting with the observed cloud
370 position on 2019-08-18. The modelled positions are generally close to the observed, except that the model fails to pick up the
effects of the synoptic weather feature beginning around July 23. Figure15b shows the output from another model run,
initialized with the observed CCC position on 2019-09-19 and then run backwards to 2019-08-18. Figure15c,d shows the
same model results but using a Hovmueller-type plot.

375 The CCC appears not to be a whirlpool or roll-up, but rather a CCC with easterly flow and drifting ("dead fish" model).
Neither the large-scale mixing events nor gravity waves penetrate well into the summer stratosphere to mix features, so they
tend to move intact if injected at the right time.

6 Conclusion

We studied the process of converting SO_2 to aerosol. The typical time for SO_2 reduction is 8-10 days, which coincides with
380 the results for Kasatochi (Krotkov et al., 2010). The peak sulfate aerosol is 1.5 months behind the time of maximum release
of SO_2. We examined the cloud of SO_2 and aerosol, which was observed for 2.5 months. The CCC is embedded in the
summer easterly flow in the stratosphere and demonstrated a diabatic "self-lofting" heating rate of about 3 K per day. For 2.5
months, the CCC circled the globe almost three times at the latitude of 30^oN and increased its height from 19 to 28 km. The
latitudinal distribution of stratospheric optical depth shows that most of the aerosol from Raikoke plume is transported
385 poleward.

The arch effect characteristic of the limb observations should be taken into account when calculating the optical thickness of
aerosol clouds, determining a characteristic cloud height or using calibration heights of ~ 45 km, which should be free of
aerosol but can be contaminated with polar mesospheric clouds (PMCs) and other high aerosol clouds.

Data availability

The SO_2 data for OMPS, OMI and TROPOMI data are available at https://so2.gsfc.nasa.gov/ and Goddard Earth Sciences
Data   and   Information   Services   Center   (GES   DISC)   at

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Competing interests

The authors declare that they have no conflicts of interest.

Authors's contributions

400 NG, CL, LL, SC, MD, MS, GT developed computer codes and algorithms, analyzed the results and wrote the manuscript. NK, JJ, AV set the task of developing, supported the development of the algorithm, analyzed the results and wrote the manuscript. PN, OT, PC analyzed the data and results and wrote the manuscript.

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