S5P+I SO2 LAYER HEIGHT

VALIDATION REPORT

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1 Purpose and objective

The purpose of this document is to describe the validation operations of the S5P+I SO_2 Layer Height Level-2 products. This is the final version of the Validation Report.

2 Document overview

In Section 1, 2 and 3 the purpose of this document, applicable documents and acronyms, are given. In Section 4 a short overview about the retrieval algorithm is given, along with an overview about the product requirements. In Section 5 the validation/verification datasets are presented and the main algorithm parameters are given. In Section 6 the actual validation efforts are presented, whereas in Section 7 the main findings are summarized.

3 References, terms and acronyms

3.1 Applicable Documents

The following project documents contain provisions which, through reference in this text, become applicable to the extent specified in this document.

Table 1 Applicable Documents

Document Title		Document ID	Issue
[AD01]	Statement of Work, ESA Express Procurement Plus - [EXPRO+] Sentinel-5p Innovation (S5p+I)	EOP-SD-SOW-2018-049	2.0 (20.08.2018)

3.2 Reference Documents

The following standards or documents are considered reference for this document.

Table 2 Reference Documents

Title		Document ID	Issue
[RD01]	Sentinel-5P TROPOMI SO2 ATBD	S5P-BIRA-L2-400E-ATBD	2.2.0
[RD02]	Sentinel-5 Precursor/TROPOMI Level 2 Product User Manual Sulphur Dioxide SO_2	S5P-L2-DLR-PUM-400E	2.1.0
[RD03]	Sentinel-5P TROPOMI SO_2 LH ATBD	S5P+I-SO2LH-D4-ATBD-v4	4.0
[RD04]	Sentinel-5P TROPOMI SO_2 LH RB	S5P+I_SO2LH_RB_D1	1.0
[RD05]	Sentinel-5P TROPOMI ATBD of the Aerosol Layer Height	S5P-KNMI-L2-0006-RP	1.1.0
[RD06]	Sentinel-5P TROPOMI ATBD of the UV aerosol index	S5P-KNMI-L2-0008-RP	1.1

3.3 Electronic references

Title		Source	Issue
[ER01]	S5P SO2 LH Data Pool (DP)	https://atmos.eoc.dlr.de/so2-lh/datapool/	1.0
[ER02]	IASI/MetOp SO_2 ACSAF column data	https://iasi.aeris-data.fr/so2_iasi_a_arch/	
[ER03]	NASA CALIOP CALIPSO data	https://www-calipso.larc.nasa.gov/	

3.4 Terms and Abbreviations

Abbreviations specific to the report are found in the following table:

Abbreviation	Meaning
ACSAF	Atmospheric Composition Monitoring Satellite Application Facility
AOPP	Atmospheric, Oceanic and Planetary Physics sub-department of the University of Oxford
ATBD	Algorithm Theoretic Baseline Document
BIRA-IASB	Royal Belgian Institute for Space Aeronomy
CALIOP	Cloud-Aerosol Lidar with Orthogonal Polarization
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
CNES	Centre National d' Études Spatiales
D.U.	Dobson Units
DOAS	Differential Optical Absorption Spectroscopy
ECMWF	European Centre for Medium-Range Weather Forecasts
EO	Earth Observation
FM	Forward model
FP_ILM	Full-Physics Inverse Learning Machine
FRESCO	Fast Retrieval Scheme for Clouds from the Oxygen A Band
HARP	Software component of the Atmospheric Toolbox
HYSPLIT	Hybrid Single Particle Lagrangian Integrated Trajectory Model
IASI	Infrared Atmospheric Sounding Interferometer
LH	Layer height
LIDORT	Linearized Discrete Ordinate Radiative Transfer
MAX-DOAS	Multi Axis Differential Optical Absorption Spectroscopy
MLS	Microwave Limb Sounder
NN	Neural Network
NOVAC	Network for Observation of Volcanic and Atmospheric Change
OE	Optimal Estimation
OMPS	Ozone Mapping & Profiler Suite
RTM	Radiative Transfer Model
RTTOV	Radiative Transfer for TOVS

Abbreviation	Meaning
S4	Sentinel-4
S5 L2PP	Sentinel-5 Level 2 Prototype Processors
S5P	Sentinel-5 precursor
SACS	Support to Aviation Control Service
SO_2	Sulfur dioxide
TAB	Total Attenuated Backscatter
TROPOMI	Tropospheric Ozone Measurement Instrument
UVAI	UV Aerosol Index
VCD	Vertical column density
VEI	Volcanic Explosivity Index
VFM	Vertical Feature Mask
VIIRS	Visible Infrared Imaging Radiometer Suite

4 SO_2 layer height retrieval algorithm

4.1 Algorithm description

The 'Full-Physics Inverse Learning Machine' (hereafter referred to as FP_ILM) for the retrieval of the SO_2 layer height is based on Hedelt *et al.* (2019) which was developed for the retrieval of the SO_2 LH based on TROPOMI data. It was further optimized in the framework of this ESA Sentinel-5P Innovations project.

Conceptually, the FP_ILM consists of a training phase, in which the inversion operator is trained using synthetic data generated with an appropriate radiative transfer model (RTM), and an operational phase, in which the inversion operator is applied to real satellite measurements. The main advantage of the FP_ILM over classical direct fitting approaches is that the time-consuming training phase involving complex RT modeling is performed offline; the inverse operator itself is robust and computationally simple and therefore extremely fast.

During the training phase, the Linearized Discrete Ordinate Radiative Transfer model (LIDORT) with inelastic rotational Raman scattering (RRS) implementation (Spurr *et al.*, 2008) is deployed to compute simulated reflectance spectra in the wavelength range 311 - 335 nm, which are then convolved with the TROPOMI Instrument Spectral Response Function (ISRF). To extract the information about the layer height and to reduce the dimensionality of the spectral dataset, a principal component analysis (PCA) is applied to the simulated spectra, which also include noise. By thus characterizing the set of simulated measurements with fewer parameters, a simpler, more stable and computationally efficient inversion scheme can be realized. The dimensionality-reduced spectra, together with information about the O_3 VCD, the viewing angles, the surface pressure and albedo of each training data-point, are then used as input to train a feedforward artificial neural network including regression, with the corresponding SO_2 LH as the output layer.

In the operational phase, the first step is to use the principal component scores acquired during the training phase to transform a given TROPOMI spectral measurement data set to one with a lower dimension. Once this is done, the neural network inverse function is then applied to retrieve the SO_2 LH.

A detailed algorithm description can be found in the SO_2 LH Algorithm Theoretical Baseline Document (ATBD) [RD03].

4.2 Product requirements

In the framework of the S5P+I SO_2 LH product, the Sentinel-5 Precursor Innovation SO_2 LH project Requirements Baseline Document [RD04] clearly states that the uncertainty of **the SO_2 LH has to be smaller than 1 km (breakthrough) to 2 km (threshold) for SO_2 VCD > 25 D.U.**

5 Reference Measurements

This section provides information on the independent satellite observations to be used for the product validation.

5.1 Satellite measurements

For the aims of the detailed cross-comparison of the TROPOMI Sentinel-5p SO_2 Layer Height products with existing EO-based equivalent/alternative datasets, which shall be performed in order to gain a thorough understanding of the range of validity, limits and benefits of the different existing products, we perform the following verification and validation study using IASI/MetOp-A, /MetOp-B and /MetopC SO_2 Layer Height, as well as CALIOP/CALIPSO ash Layer Height observations.

5.1.1 IASI/MetOp SO_2 layer height verification.

Two different IASI/Metop SO_2 column and SO_2 LHs will be used for the validation of the S5P SO_2 LHs: the EUMETSAT ACSAF Brescia v201510 product (Clarisse et al., 2012; 2014; Astoreca et al., 2018) as well as the University of Oxford product (Carboni et al., 2016.)

i. The IASI/MetOp SO_2 ACSAF column data are fully described in Clarisse et al., 2012, where a novel algorithm for the sounding of volcanic SO_2 plume above ~5 km altitude was presented and applied to IASI. The algorithm is able to view a wide variety of total column ranges (from 0.5 to 5000 D.U.), exhibits a low theoretical uncertainty (3–5 %) and near real time applicability and was thence demonstrated on the recent eruptions of Sarychev in Russia, Kasatochi in Alaska, Grimsvötn in Iceland, Puyehue-Cordon Caulle in Chile and Nabro in Eritrea. A validation of this algorithm on the Nabro eruption observations using forward trajectories and CALIOP coincident measurements is presented in Clarisse et al., 2014, where the expansion of the algorithm to also provide SO_2 LHs is also described. This dataset includes five SO_2 column data at assumed layer heights of 7, 10, 13, 16 and 25 km, as well as a retrieved best estimate for the SO_2 LH. It is important to note that the SO_2 LHs provided by this algorithm are quantized every 0.5km, which renders simple scatter-type comparisons not as straightforward. This dataset is publicly available from [ER02]

ii. The University of Oxford employs an optimal estimation scheme (Carboni et al. 2012, 2016) to estimate SO_2 column amount, the height of the SO_2 profile, and the surface skin temperature to IASI/MetOp-A and /MetOp-B measurements. The IASI spectral range encompasses the three SO_2 absorption features two of which are used in the SO_2 retrievals. These are the ν1 (1000-1200cm^{-1}) and ν3 (1300-1410 cm^{-1}) bands. The strongest SO_2 band is the ν3, around 7.3μm, which is contained within a strong water vapour (H_2O) absorption band and subsequently, it is not very sensitive to emissions from the surface and lower troposphere. However, above the lower troposphere, this band contains valuable information on the vertical profile of SO_2. Fortunately, differences between the H_2O and SO_2 absorption spectra allow the signals from the two gases to be decoupled in high resolution measurements. The SO_2 ν1 band, at around 8.7 μm (1000 to 1200 cm^{-1}) is within an atmospheric window. This allows the radiation from the surface to reach the satellite from deep within the atmosphere enabling the retrieval of SO_2 amount down to the surface. The detection scheme is a linear retrieval with one free parameter, the column amount of SO_2, while the vertical distribution of SO_2 and the atmospheric vertical profiles (temperature and trace gases) are assumed. In the retrieval scheme a detection is considered 'positive' if the output of the linear retrieval is greater than a defined positive threshold (0.49 effective DU, following Walker et al. 2012). The detection limits for a standard atmosphere (with no thermal contrast) are estimated to be: 17 DU for a SO_2 plume between 0-2 km, 3 DU between 2-4 km, and 1.3 DU between 4-6 km (Walker et al., 2011). The detection scheme can miss part of an SO_2 plume under certain circumstances, such as low-altitude plumes, conditions of negative thermal contrast (i.e. where the surface is colder than the atmosphere), and where clouds are present above the SO_2 plume, masking the signal from the underlying atmosphere. An iterative retrieval is performed for the pixels that give positive detection results. All the channels in the ranges 1000-1200 and 1300-1410 cm-1 (the 7.3 and 8.7μm SO_2 bands) are simultaneously used in the iterative optimal estimation retrieval scheme to obtain the SO_2 amount, the altitude of the plume and the surface temperature. The scheme iteratively fits the forward model (simulations) with the measurements, through the error covariance matrix, to seek a minimum of a cost function. The forward model is based on RTTOV (Radiative Transfer for TOVS) which is a very fast radiative transfer model for passive visible, infrared and microwave downward-viewing satellite radiometers, spectrometers and interferometers. It is a FORTRAN 90 code for simulating satellite radiances, designed to be incorporated within user applications (Saunders et al., 2018). It has been extended to include SO_2 explicitly and uses ECMWF profiles interpolated to the measurement time and location. The error covariance matrix used is the 'global error covariance matrix' in Carboni et al. (2012). It is defined to represent the effects of atmospheric variability not represented in the forward model (FM), as well as instrument noise. This includes the effects of cloud and trace gases which are not explicitly modelled. The matrix is constructed from differences between FM calculations (for clear-sky driven with ECMWF profiles) and actual IASI observations for a wide range of conditions, when we are confident that negligible amounts of SO_2 are present. Only quality controlled pixels are considered; these are values where the minimization routine converges within 10 iterations, the SO_2 amount is positive, the plume pressure is between 0 and 1100mb and the cost function is less than 10. A comprehensive error budget for every pixel is included in the retrieval. Rigorous error propagation, including the incorporation of FM parameter error, is built into the system, providing quality control and comprehensive error estimates on the retrieval results. The IASI SO_2 retrieval is not affected by underlying cloud. If the SO_2 is within or below a cloud layer its signal will be masked and the retrieval will underestimate the SO_2 amount.

5.1.2 CALIOP/CALIPSO ash layer height verification.

As an independent satellite source for the validation of the ash layer height the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument onboard the CALIPSO mission is used. CALIPSO is the first polarization lidar to provide global atmospheric measurements and is quite able to identify volcanic eruption plumes and data from its instruments have been used in numerous publications, such as Carboni et al., 2016; Carn et al., 2015; Ge et al., 2016; Hughes et al., 2012; Krotkov et al., 2010; Raffuse et al., 2012; Vira et al., 2017; Wang et al., 2012, to name by a few relevant to the SO_2 Layer Height identification and retrieval. For the validation of the TROPOMI/S5P SO_2 LH products level 2 data different CALIPSO products are used: these consist of three basic types of information: layer products, profile products and the vertical feature mask (VFM). Layer products provide layer-integrated or layer-averaged properties of detected aerosol and cloud layers. Profile products provide retrieved extinction and backscatter profiles within these layers. Because information on the spatial locations of cloud and aerosol layers is of fundamental importance, the VFM was developed to provide information on cloud and aerosol locations and types. The methodology described in Winker et al, 2012 will be applied in the comparisons in a manner similar to the one provided in Koukouli et al., 2014a; 2014b, where the aerosol optical depth, extracted from the IASI observations, was used as a proxy locator for the volcanic SO_2 plume. For this methodology, the following CALIPSO level-2 data products are analysed: the Vertical Feature Mask and the aerosol layer products. The Vertical Feature Mask describes the vertical and horizontal distribution of layers, including both cloud and aerosol.

A more detailed description of the CALIPSO level-2 data products used in this report are given in Section 6.2.1.1 where an example day is presented for clarity.

Table 3. Space-borne datasets used in the S5P SO_2 LH verification.

Instrument	Platform	Time span	Coverage	Pixel size	Geophysical Product
TROPOMI	Sentinel-5p	2017-todate	Global, once per local solar time	7 x 3.5 km^2 [5 x 3.5 km^2 after August 2020]	L2 SO_2 column L2 SO_2 LH L2 SO_2 LH error L2 ALH product
IASI	MetOp-A MetOp-B MetOp-C	2007-todate 2013-todate 2019-todate	Global, twice per day [every 12h LST]	12km in diameter	SO_2 column at retrieved plume height SO2 layer height
CALIOP	CALIPSO	2006-todate	Global, every 16-days	100-300 m	VFM & aerosol layer products

6 Validation approach

We will use the final version (v4.0) of the S5P LH results for this validation report. A detailed description of the algorithm can be found in [RD01]. To compare and validate the S5P SO_2 LH results with other datasets, we will focus on eruptions with an extended plume such that independent satellite data with a different overpass time are also available. Although this is not a major issue for IASI data (global daily coverage, low SO_2 threshold), collocated CALIOP/CALIPSO measurements have to be carefully identified. Furthermore, to assess the error of the retrieved SO_2 LH in the presence of absorbing and non-absorbing aerosols, we will also select volcanic eruptions with known ash or sulphate emissions.

For the comparisons with the IASI products, the data parameters enumerated in Table 4 were extracted, as described in Table 11 of the [RD01]:

Table 4. Parameters extracted from the S5P SO2 I+ LH product files for this validation.

Parameter	Short explanation	Unit
Time	Julian Date and Time of the measurement	n/a
Longitude [center]	Latitude at the center of the pixel	deg
Latitude [center]	Longitude at the center of the pixel	deg
SO_2 layer height	SO_2 layer (mid) height	m
SO_2 layer height error	SO_2 layer (mid) height error	m
Improved VCD	SO_2 VCD calculated using SO_2 LH	mol/cm^2
QA value	QA value indicating quality of retrieval	n/a
LH flag	Flag indicating warnings and errors during the retrieval	n/a

In the following we have selected a number of volcanic eruptions that presented in this version of the Validation Report.

- The **Sinabung** stratovolcano (2,460 m summit elevation) on the island of Sumatra has been highly active until September 2010 and was quiet until a new eruptive phase began in September 2013 which lasted until March 2018, see Venzke, 2018. On **19 February 2018**, Sinabung erupted violently at around 02:55h UTC with its largest explosion to date, emitting a volcanic ash plume that rose to at least 16.8 km altitude and an SO_2 plume of up to 50 DU of SO_2 that was observed by several satellite instruments, including TROPOMI (around 06:30h UTC), OMI, IASI (overpass time around 03:30h UTC and 15:00h UTC), MLS (overpass time around 07:10h UTC). There was also an overpass of CALIPSO/CALIOP at about 07:15h UTC, over the volcanic plume.
- On **22 June 2019** at 04:00h local time, the **Raikoke** stratovolcano located on the Kuril Islands (Russia, 551 m summit elevation) erupted explosively (VEI ≥ 4) after being dormant since 1924. There were several strong distinct explosions, producing a dense ash and SO_2 plume that rose until 13 km altitude the first days and was entrained into the stratosphere (see Sennert, 2019). This was the strongest volcanic eruption since the Merapi eruption in 2011, producing a colossal SO_2 plume with an SO_2 loading of more than 900 DU on 22 June 2019, that was dispersed by strong winds over Russia and North America, and was detectable even two weeks after the volcanic eruption. MLS was able to perform SO_2 profile measurements and also CALIPSO was able to detect the ash plume during the first days after the eruption.

- The **Taal** Volcano in Batangas, Philippines erupted on the afternoon of **January 12th, 2020**, 43 years after its previous eruption in 1977. Stronger explosions began around 3 pm that spewed an ash column exceeding a kilometre high. By 7:30 pm, volcanic activities intensified as continuous eruptions generated a tall 10 to 15 kilometres steam-laden tephra column. On January 13, the activity on Taal's main crater had transitioned into a lava fountain.
- **Nishinoshima** lies about 600 miles (1,000 km) south of Tokyo, Japan, in the Ogasawara Arc. A young volcano, it has gone through several expansions in recent history. During a major eruption in 1973-1974, several new islands coalesced and expanded the size of the small island. In 2013, another vigorous eruption, which began offshore and continued until late 2015, eventually covered the island with lava flows and again enlarged the island. The volcano has been increasingly active through most of 2020, with **July 2020** seeing extreme activity, including a series of powerful vulcanian-type eruptions from July 30-August 1st, characterized by dense, ash-rich cloud of gas exploding from the volcanic crater and typically rising high above the peak.
- On the morning of **April 9th 2021**, **La Soufrière** volcano, on the Caribbean island of Saint Vincent, began erupting, spewing ash at least 25,000 feet in the air. The volcano continued to erupt over the next several days, with multiple violent explosions. Ash blanketed Saint Vincent and winds carried ash to Barbados, about 120 miles east. A violent eruption on April 12th generated pyroclastic flows, a high-density mix of hot lava blocks, pumice, ash, and volcanic gas that moves at very high speeds down volcanic slopes. La Soufrière last erupted in 1979.

A note that applies to the validation shown hereafter is to acknowledge the use of the Atmospheric Toolbox® HARP and CODA routines for the ingestion, manipulation and collocation of the satellite observations.

6.1 Comparison between the S5P/TROPOMI and the IASI/Metop SO_2 layer heights

In the S5P SO_2 LH Data Pool [ER01], SO_2 LH v4.0 data are currently present in NRT and OFFL processing mode and are continuously updated on a daily basis in case a volcanic eruption occurred. These currently cover the entire operational mission time of S5P, which started in May 2018 in a non-continuous manner (i.e. only for dates where a volcanic eruption occurred) up to now. Note that also an additional time period in the pre-operational mission phase was investigated (February 2018), which covers the strong volcanic eruption of Mt. Sinabung. In order to compare the observations to the collocative datasets, a well-established gridding technique was applied to the S5P SO_2 LH data, bringing them onto a 0.1x0.1° grid at 6h increments for each day. The choice of the temporal field was applied since the S5P and MetOp orbits differ on average by 3-4h and this temporal range was found to be the optimal trade-off between resulting in a successful collocative dataset while also ensuring the comparisons view the same parts of the SO_2 plumes. Recall also that IASI, an infrared sounder, also performs observations 12h later, during night-time. For high enough latitudes, the time zones collapse onto another, so in the case of the Raikoke plumes a collocation closer in time can be achieved.

A selection of filters was also applied to the S5P SO_2 LH data, before gridding. After applying the required filters shown in Table 5, left column, and quality controls on the databases to exclude erroneous values [for e.g. negative, non-physical, etc.], we proceeded in collocating to the provided IASI/AOPP and IASI ULB/LATMOS datasets.

6.1.1 IASI/Metop observations by the AOPP algorithm

The SO_2 LH data provided by the IASI/AOPP algorithm were gridded onto the same 0.1x0.1° grid at 6h increments for each day, as per the S5P SO_2 LH data. Furthermore, they were filtered following recommendations given in the SO_2 LH ATBD (cf. Sections 5.3.6 and 5.3.7 in [RD03]), as shown in Table 5, right column.

Table 5. Filtering of the S5P and the IASI/AOPP SO_2 Layer Height.

S5P FILTERS	IASI/METOP AOPP FILTERS
Retrieved SO_2 load > 20 D.U. qa_value > 0.5 LH flag < 16	SO_2 layer height ≤ 25km SO_2 layer height error ≤ SO_2 layer height Retrieved altitude ≠ Apriori altitude [400 mbars]

Observations during the Raikoke, Kuril Islands eruption, between the 22nd of June and the 31 of August 2019 and during the Taal, Philippines, eruption, on the 13th of January 2020, made by the IASI/Metop instruments were provided by the University of Oxford team, based on the Atmospheric Oceanic and Planetary Physics, AOPP, algorithm. As it is well-established that the three, currently in orbit, IASI instruments provide nearly identical level-2 products, and in order to increase our available collocations, their measurements were all grouped into one verification database. Note that there is a slightly different overpass time, typically within 1h, between instruments, however since we grid the datasets every 6h – to account for the difference between the S5P and Metop orbits – this time difference does not affect the comparisons.

6.1.1.1 Raikoke 2019 eruption comparisons.

Spatial collocations for 6h intervals in the days between the 22nd of June 2019 and the 20th of July 2019 are shown in *Figure 1*, where the histogram representation of the SO_2 layer heights (LH) are shown in the left column and the scatter plots in the right column. The three rows show the effect of choosing different filtering criteria for the S5P SO_2 LHs: all cases are filtered using a qa_value > 0.5, however in the upper row, the original S5P SO_2 LH points are filtered for an associated SO_2 load > 15 D.U., in the middle row, for an associated SO_2 load > 20 D. U. while in the lower row, for an associated SO_2 load > 20 D. U. and LHflag < 16. While we note a decrease in the amount of collocated pixels, from ~24,000 to ~17,000, the mean IASI to S5P differences [show as a histogram in red] improve from 1.09±4.08km, to 0.62±3.7km to 0.61±3.72km in the stricter filtering case. Overall, the IASI AOPP SO_2 LH for Raikoke reports a mean height of ~11.5±2.5km while S5P reports ~10.5±3.5km, in excellent agreement. The correlations and linear behaviour of the comparisons is not as promising, however such representation is beneficial since one can note a large number for S5P pixels reported heights above 25km, reaching even 35km, not reproduced by IASI/AOPP which places those pixels between 10 and 15km.

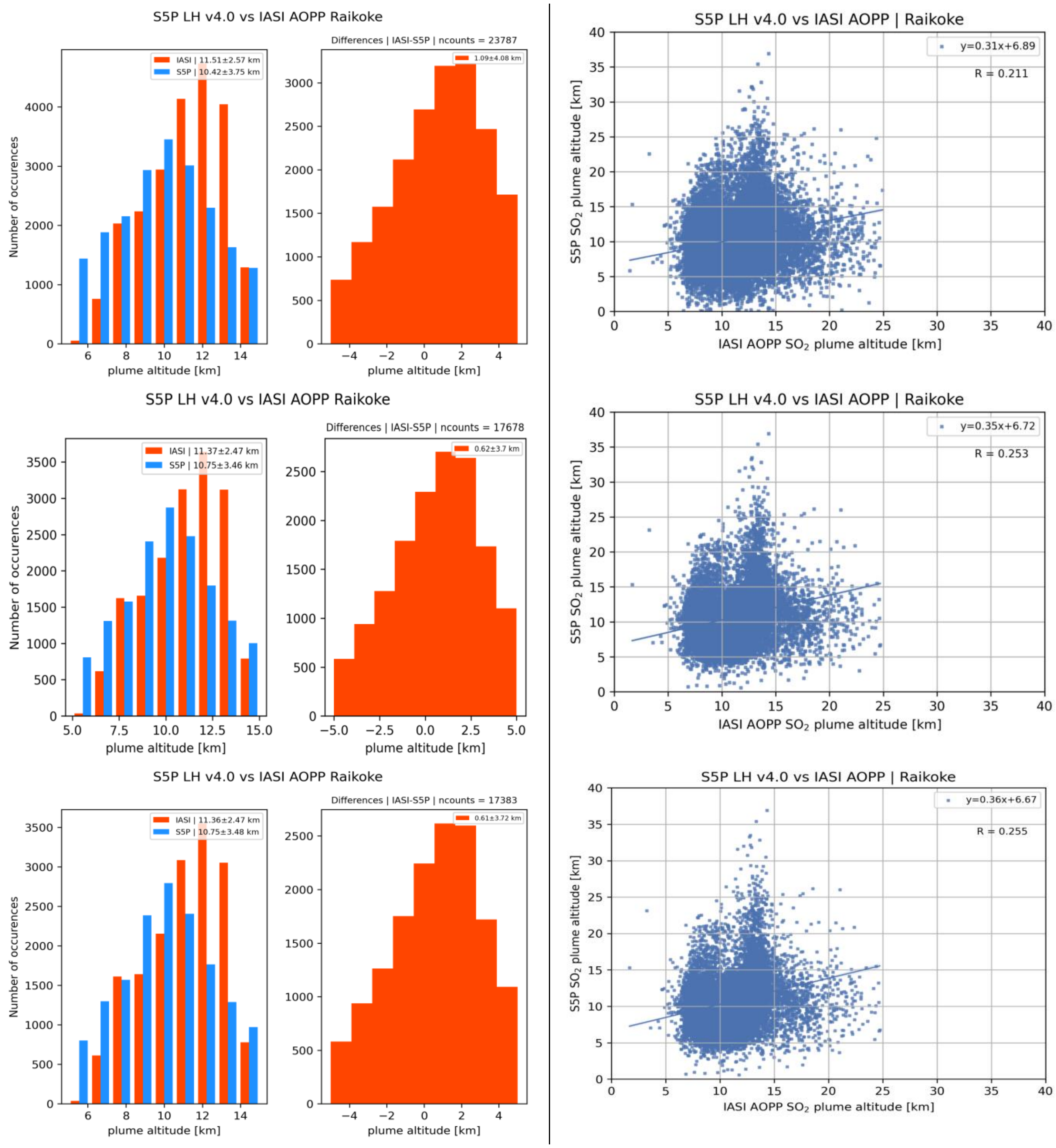
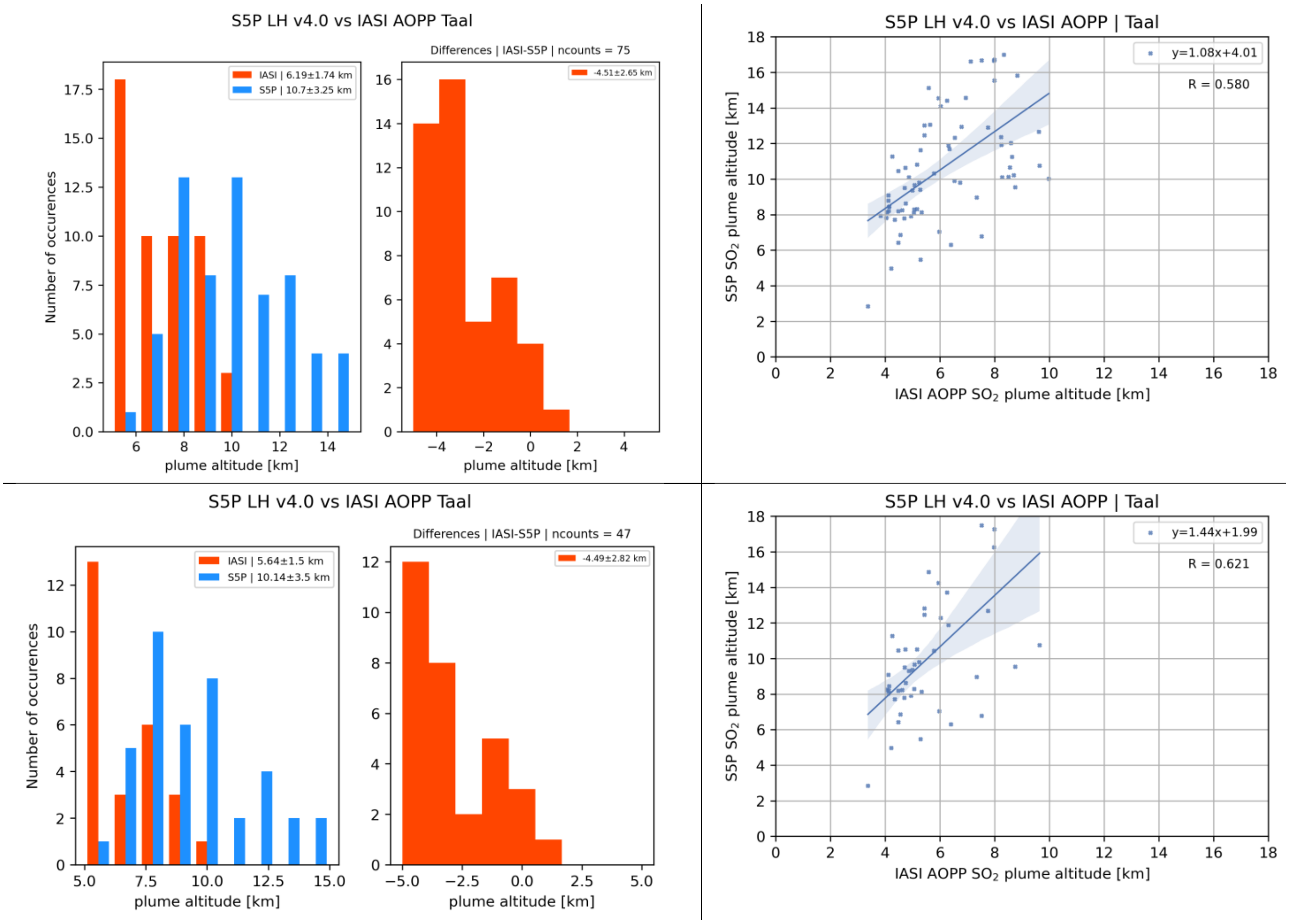


Figure 1. *Raikoke eruption. Collocated gridded IASI/AOPP [red] and S5P [blue] SO_2 plume heights in histogram [left] and scatter plot representation [right column]. Upper row: S5P SO_2 LH filtered for qa_value>0.5and SO_2 load > 15 D.U. Middle row: S5P SO_2 LH filtered for qa_value>0.5and SO_2 load > 20 D.U. Lower row: S5P SO_2 LH filtered for qa_value>0.5, SO_2 load > 20 D.U. and LHflag < 16.*

6.1.1.2 Taal 2020 eruption comparisons

Spatial collocations for 6h intervals for the 13th of January 2020 are shown in *Figure 2*, where the histogram representation of the SO_2 LHs are shown in the left column and the scatter plots in the right column, following the same format of *Figure 1*. In this case, the overall SO_2 LHs reported differ substantially with IASI reporting heights ~5.5±1.5km while S5P reports higher columns, at ~10±3.5km. However, the scatter plot comparison [right column] reveals common SO_2 LHs patterns for the two sensors, with substantial correlations ~0.6, and a constant bias in the SO_2 LHs reported of ~4km, with S5P overestimating the infrared sensor.



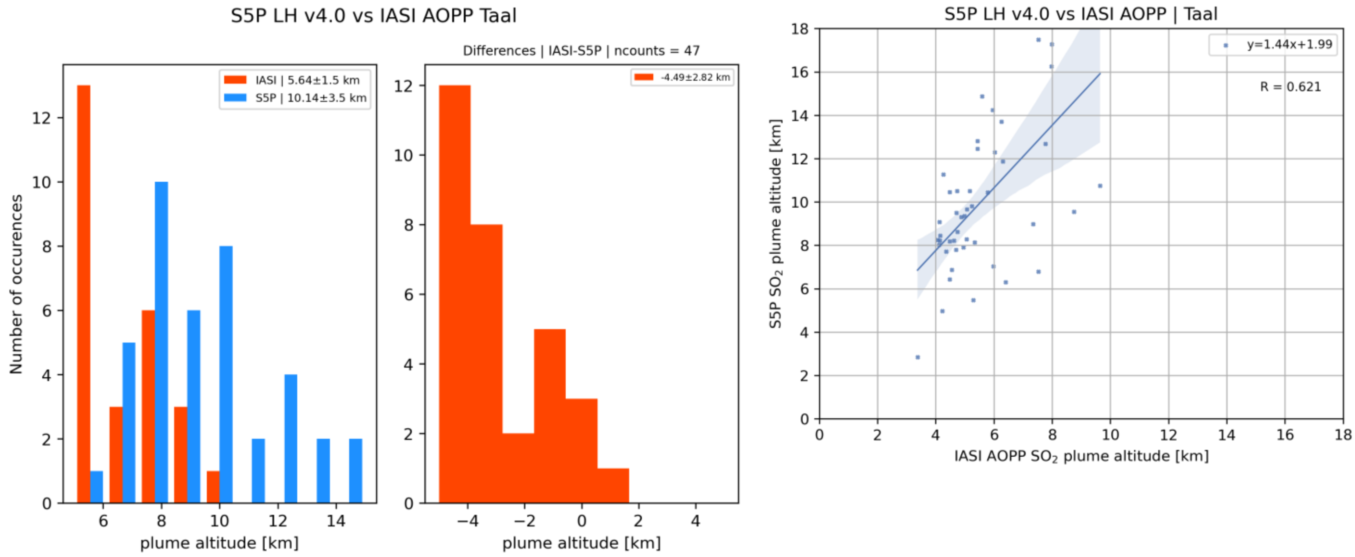


Figure 2. *Taal eruption. Collocated gridded IASI/AOPP [red] and S5P [blue] SO_2 plume heights in histogram [left] and scatter plot representation [right column]. Upper row: S5P $SO_{2\ LH}$ filtered for qa_value>0.5 and SO_2 load > 15 D.U. Middle row: S5P SO_2 LH filtered for qa_value>0.5 and SO_2 load > 20 D.U. Lower row: S5P $SO_{2\ LH}$ filtered for qa_value>0.5, SO_2 load > 20 D.U. and LHflag < 16.*

6.1.1.3 La Soufriere, 2021, eruption

Spatial collocations for 6h intervals for the 10th and the 11th of April 2021 are shown in *Figure 3*, where the histogram representation of the SO_2 LHs are shown in the left column and the scatter plots in the right column, following the same format of *Figure 1*. In this case, as for Raikoke, both sensors report quite high SO_2 LHs, at ~13.5km, with a mean difference of -0.35±3.5km for the case of the strict filtering [bottom row of *Figure 3*]. The scatter plot comparison [right column] reveals a number of low SO_2 LHs reported by IASI/AOPP which are not observed by S5P and which result in the low correlations found.

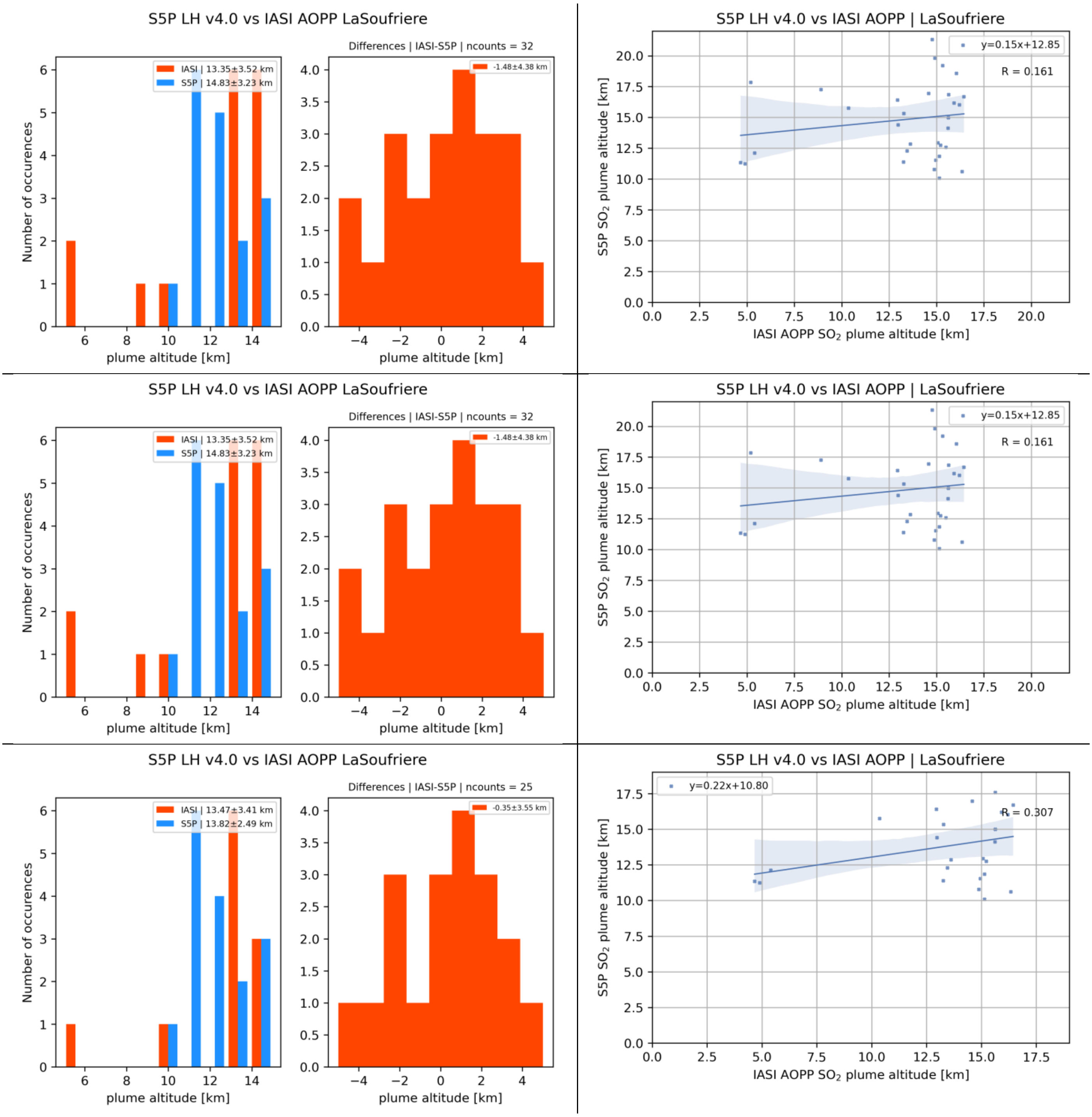


Figure 3. *La Soufriere eruption. Collocated gridded IASI/AOPP [red] and S5P [blue] SO_2 plume heights in histogram [left] and scatter plot representation [right column]. Upper row: S5P SO_2 LH filtered for qa_value>0.5 and SO_2 load > 15 D.U. Middle row: S5P SO_2 LH filtered for qa_value>0.5 and SO_2 load > 20 D.U. Lower row: S5P SO_2 LH filtered for qa_value>0.5, SO_2 load > 20 D.U. and LHflag < 16.*

6.1.2 IASI/Metop observations by the ULB-LATMOS/ACSAF algorithm

A similar gridding and collocation sequence, as for the IASI AOPP data, was followed for the IASI ULB/LATMOS dataset. Due to the fact that the IASI ULB/LATMOS SO_2 layer heights are quantized every 0.5km, we compare against the S5P/TROPOMI LHs using histogram representations only. In the following, we present comparisons against four eruptions within the S5P timeline, namely Raikoke 2019, Taal 2020, Nishinoshima 2020 and La Soufriere 2021, for the stricter filtering case only, i.e that the original S5P SO_2 pixels are filtered for qa_value>0.5, SO_2 load > 20 D.U. and LHflag < 16. In *Figure 4*, the Raikoke collocations are shown between the 22nd of June 2019 and the 20th of July, 2019 in the upper left, the Taal collocations for the 13th of January 2020 in the upper right, the Nishinoshima collocations for the 10th, 11th and 16th of July 2020 in the lower left and the La Soufriere collocations for the 10th and 11th of April 2021 in the lower right.

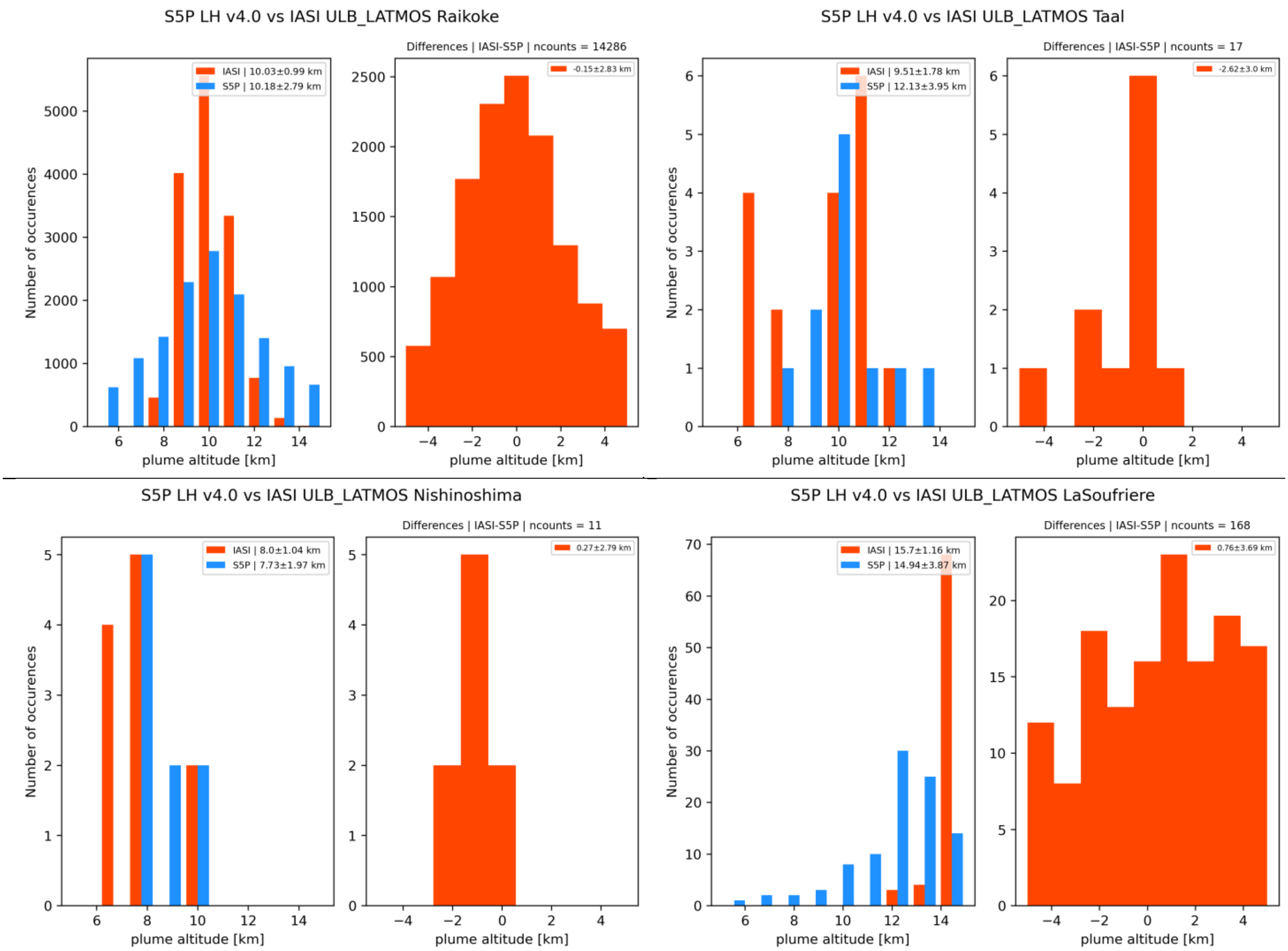


Figure 4. *IASI ULB/LATMOS and S5P SO_2 LHs collocation comparisons for S5P SO_2 pixels filtered for qa_value>0.5, SO_2 load > 20 D.U. and LHflag < 16 are shown for Raikoke [upper left], Taal [upper right], Nishinoshima [lower left] and La Soufriere [lower right.]*

An excellent agreement, well within the S5P product requirements, is found in the comparisons between IASI ULB/LATMOS and S5P SO_2 LHs. For Raikoke, both sensors report on average LHs at ~10km, with the IASI std at 1km and S5P std at ~3km and a mean difference of ~0±3km. For Taal, IASI reports lower heights, at 9.5±2km while and S5P places the plume at ~12±4km with a mean difference of~2.5±3km. For Nishinoshima, both sensors place the plume at the same altitude, with IASI at ~8±1km and S5P ~8±2km and mean difference of~0±3km. Finally, for the La Soufriere eruptions, again, both sensors report high plume levels, at ~15km, with the IASI std at 1km and the S5P std at ~4km, and overall mean difference of ~1±3.5km.

6.1.3 Summary of the comparisons with the IASI/Metop observations

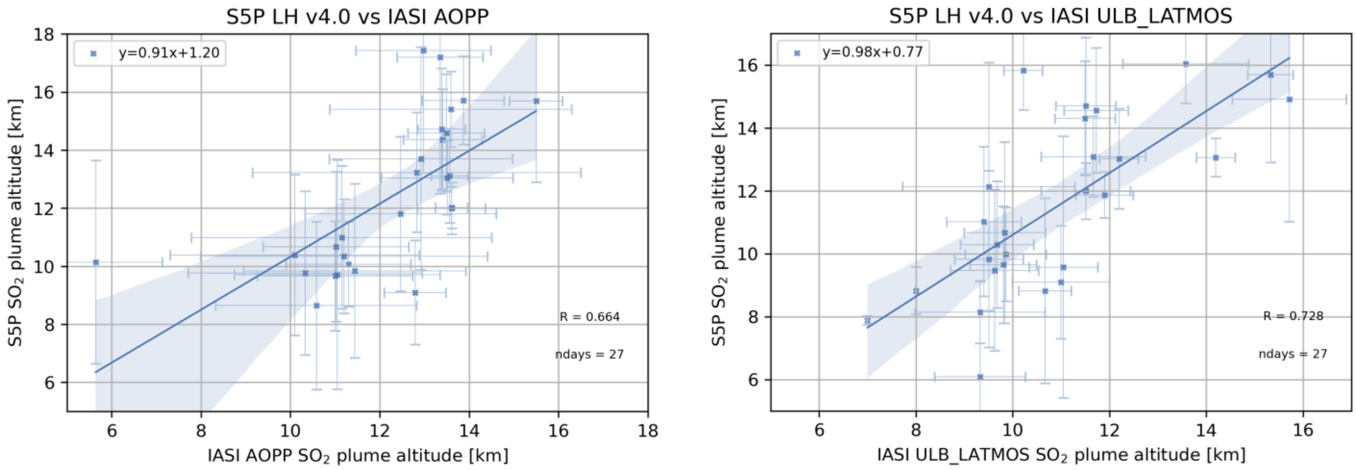


Figure 5. *Scatter plot of the mean daily average reported SO_2 LHs by S5P and IASI/AOPP [left] and IASI ULB/LATMOS [right] for all available collocated eruptive days.*

- The S5P/TROPOMI SO_2 layer height product has been compared against IASI/Metop SO_2 layer height for the eruptive periods of the Raikoke volcano, 22 June to 30 July 2019, the Taal volcano, 13 January 2020, the Nishinoshima eruptive period, July 2020, and the La Soufriere eruptive days, April 10^{th} to 11^{th}, 2021. Two different algorithms were examined, the official EUMETSAT ACSAF algorithm, labeled ULB/LATMOS, and the University of Oxford, AOPP, algorithm. All three IASI instruments, flying on board MetopA, MetopB and MetopC, where considered simultaneously in the correlative databases.
- For the comparisons between S5P and IASI/AOPP SO_2 LHs, for the strict filtering of the S5P dataset, for the Raikoke eruptive days the difference between dataset is 0.61±3.72km, while the IASI/AOPP SO_2 LH reports a mean height of ~11.5±2.5km and S5P reports ~10.5±3.5km, in excellent agreement. For the Taal eruptive day, the SO_2 LHs reported differ substantially with IASI/AOPP reporting heights ~5.5±1.5km while S5P reports higher columns, at ~10±3.5km. The scatter plot comparison reveals common SO_2 LHs patterns for the two sensors, with substantial correlations ~0.6, and a constant bias in the SO_2 LHs reported of ~4km, with S5P overestimating the infrared sensor. We present a summary plot of the comparisons between S5p

and IASI/AOPP SO_2 LHs, in Figure 5, left, where the mean plume height reported for each of the 27 days of collocations is shown as a scatter plot. The mean SO_2 LHs, as expected, follow quite closely a straight line, with slope of 0.9 and y-intercept of ~1.2km, with the correlation coefficient is also very promising, at 0.66. 24 days belong to the Raikoke eruptive period, 2 days to La Soufriere, while the Taal comparison for the 13th of January 2020 appears as the outlier point, with low IASI/AOPP SO_2 LH (~6km) while S5P reports a high LH at ~10km.

- For the comparisons between S5P and IASI ULB/LATMOS SO_2 LHs, again for the strict filtering of the S5P dataset, for the Raikoke eruptive days a mean difference of ~0±3km is found with both sensors reporting on average LHs at ~10km. For Taal, as for the case of the IASI/AOPP dataset, IASI ULB/LATMOS reports lower heights, at 9.5±2km while and S5P places the plume at ~12±4km with a mean difference of~2.5±3km. For Nishinoshima, both sensors place the plume at the same altitude, with IASI at ~8±1km and S5P ~8±2km and mean difference of~0±3km. Finally, for the La Soufriere eruptive days, both sensors report high plume levels, at ~15km, with the IASI std at 1km and the S5P std at ~4km, and overall mean difference of ~1±3.5km. We present a summary plot of the comparisons between S5p and IASI ULB/LATMOS SO_2 LHs, in Figure 5, right, where the mean plume height reported for each of the 27 days of collocations is shown as a scatter plot. The mean SO_2 LHs, as expected, follow quite closely a straight line, with slope of ~1 and y-intercept of ~0.8km, with the correlation coefficient is also very promising, at 0.73. Approximately 20 days belong to the Raikoke eruptive period, and the rest to Taal, Nishinoshima and La Soufriere.

6.2 Comparison between the S5P/TROPOMI SO_2 layer height and the CALIOP/CALIPSO ash layer height

The objective of this section is the validation of the S5P/TROPOMI SO_2 layer height against the ash height sensed by the CALIOP/CALIPSO lidar instrument. CALIOP is highly valuable in validating aerosol layer height estimates, but spatial collocations with its narrow swath are relatively rare. To obtain any collocations with the CALIOP narrow swatch, usually large temporal differences between the observations need to be allowed, which then lead to large spatial collocation uncertainties. To compare and validate the S5P SO_2 layer height, we focus on eruptions with an extended plume such that independent satellite data with a different overpass time are also available. In this report we have selected the case of Mt Sinabung, Sumarta, eruption on the 19th of February 2018,the Raikoke, Kuril Islands, eruption on the 22nd of June 2019, the Nishinoshima eruptive period of July 2020 and the La Soufriere eruption in April 2021, to be compared against the space-born lidar observations.

6.2.1 CALIOP on board the CALIPSO space-born platform

CALIPSO, *Cloud-Aerosol and Lidar Infrared Pathfinder Observations*, is a joint NASA/CNES (Centre National d' Études Spatiales) satellite and part of the A-Train constellation of satellites. It is designed to study aerosols and clouds and aims to provide profiling information at a global scale for improving our knowledge and understanding the role of the aerosol in the atmospheric processes. The main instrument, CALIOP, *Cloud-Aerosol Lidar with Orthogonal Polarization*, is a dual-wavelength (532 and 1064 nm) elastic backscatter lidar with the capability of polarization-sensitive observations at 532 nm (Winker et al., 2006; 2007). The high-resolution profiling ability coupled with accurate depolarization measurements make CALIPSO an indispensable tool to monitor specific aerosol species and clouds (Liu et al., 2008). The optical properties retrieval is based on the successful cooperation of three modules whose main mission objective is to produce the CALIPSO Level 2 data. As an independent satellite source for the validation of the ash layer height, the CALIOP instrument can be used as it measures both ash and aerosol absorption profiles. CALIPSO is the first polarization lidar to provide global atmospheric measurements and is quite able to identify volcanic eruption plumes related to the SO_2 Layer Height identification and retrieval. We can use CALIPSO data close to the volcanic source since ash (and/or aerosols) are initially collocated with the SO_2 cloud, before the gas and ash plumes separate. We note that the footprint of CALIOP measurements is only 100m, hence the global coverage is very low and detection of a volcanic ash plume is rare.

6.2.1.1 CALIPSO data products

CALIPSO data consist of three basic types of information: (a) layer products, (b) profile products and (c) the vertical feature mask (VFM). Layer products provide layer-integrated or layer-averaged properties of detected aerosol and cloud layers. Profile products provide retrieved extinction and backscatter profiles within these layers. Because information on the spatial locations of cloud and aerosol layers is of fundamental importance, the VFM was developed to provide information on cloud and aerosol locations and types. Layer properties include layer top and base altitude, and physical properties of the feature such as the Integrated Attenuated Backscatter or the Integrated Volume Depolarization Ratio, some of which are described below. Layer top and base altitudes are reported in units of kilometres above mean sea level. Between -0.5 km and ~8.2 km, the vertical resolution of the lidar is 30-meters. From ~8.2 km to ~20.2 km, the vertical resolution of the lidar is 60-meters. Above ~20.2 km, the vertical resolution is 180-meters. The on-board averaging scheme provides the highest resolution in the lower troposphere where the spatial variability of clouds and aerosols is the greatest and coarser resolutions higher in the atmosphere (*Figure 6*). The CALIPSO data products used in this validation study are summarized in Table 6.

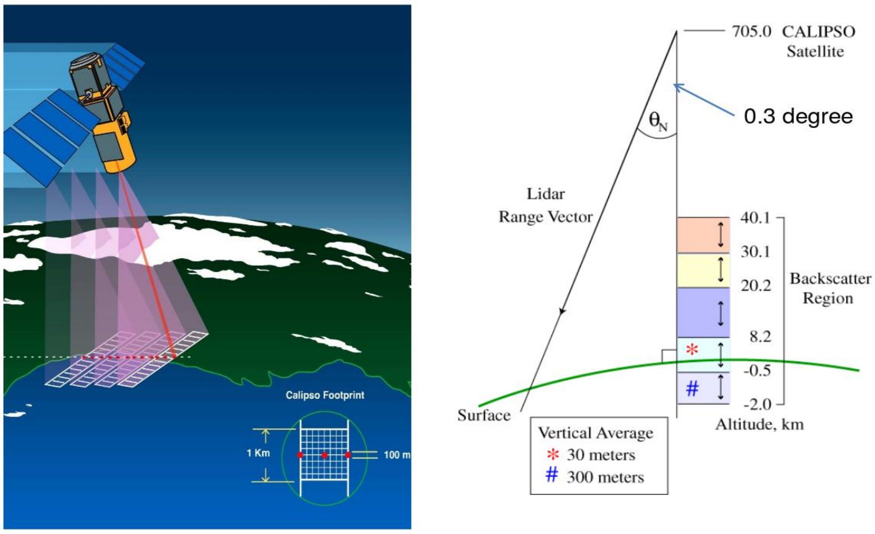


Figure 6. *Spatial resolutions for the CALIPSO on-board averaging scheme (Winker et al., 2006)*

Table 6. CALIOP/CALIPSO parameters used in this study.

Parameter	**Version**	**Level**	**Resolution Due to Averaging**		**Product filename**
			Horizontal	**Vertical (<8km)**	
Total_Attenuated_Backscatter_532	v.4.10	1	1/3 Km	30 m	**CAL_LID_L1-Standard** **CAL_LID_L1-ValStage1**
Extinction_Coefficient_532	v.3.41, v.4.20	2	5 km	60 m	**CAL_LID_L2_05kmAPro-Standard** **CAL_LID_L2_05kmAPro-Prov**
Aerosol Layer_Top/Base_Altitude	v.341, v.4.20	2	5 km	30 m	**CAL_LID_L2_05kmALay-Standard** **CAL_LID_L2_05kmALay-Prov**
Feature_Clarification_Flags	v.3.41, v.4.20	2	5 km	60 m	**CAL_LID_L2_VFM-Standard** **CAL_LID_L2_VFM-ValStage1**

The CALIPSO version v4 product determines the locations of layers within the atmosphere, discriminates aerosols from clouds and categorizes aerosol layers as one of eleven subtypes, seven in the troposphere and four in the stratosphere (Omar et al., 2009; Kim et al., 2018) providing also the optical depth of each detected aerosol layer. The tropospheric aerosol types include the following sub-types: clean marine, dust, polluted, continental/smoke, clean continental, polluted dust, elevated smoke and dusty marine. The stratospheric aerosols include the sub-types polar stratospheric, volcanic ash, sulphate/other and smoke.

NASA CALIPSO/CALIOP data are open-source and can be downloaded from [ER03].

6.2.2 Comparison of TROPOMI SO_2 LH and CALIOP ALHext

For the validation of the TROPOMI SO_2 LH, we used CALIOP level 2 version 4.10 aerosol extinction profiles at 5 km spatial resolution, retrieved from CALIOP observations of attenuated backscatter at 532 nm (Winker et al., 2010). To facilitate quantitative comparison of aerosol altitude, we used a mean extinction height calculated from the CALIOP extinction profile, following Koffi et al. (2012):

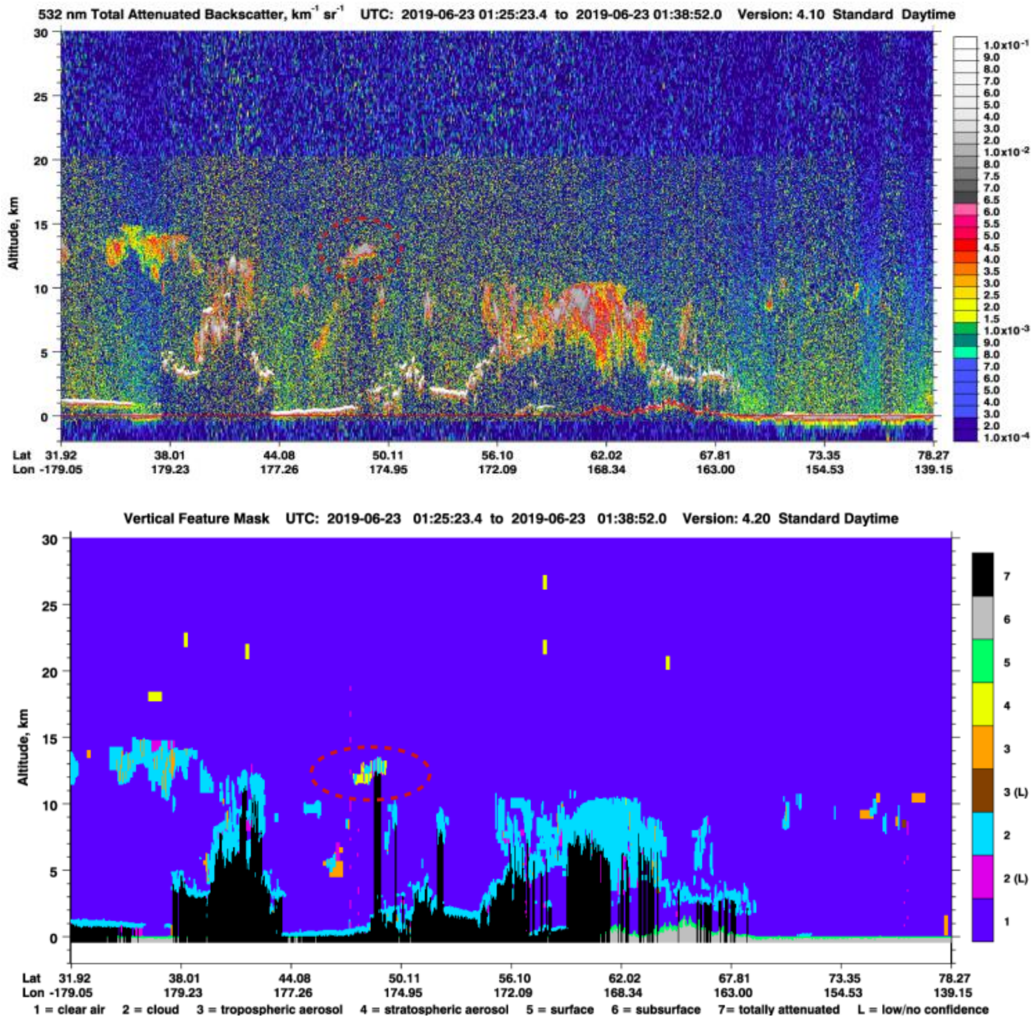
$$ALH_{ext} = \frac{\sum_{i=1}^{n} \beta_{ext,i} \cdot Z_i}{\sum_{i=1}^{n} \beta_{ext,i}}$$

where Zi is the height from sea level in the i^{th} lidar vertical level i (km), and βext,i is the aerosol extinction coefficient (km^{-1}) at the same level. In the CALIOP level 2 products, aerosol extinction is only retrieved for the layers in which aerosols are detected, depending on the instrument's signal-to-noise ratio (SNR). In the case when aerosols are present over clouds, ALHext will be situated in the centre of the aerosol layer, with any undetected aerosol layers below the cloud layer not included in the calculations due to attenuation of the signal beyond the cloud layer. In this validation report, the CALIOP 532nm channel observations are chosen for analysis as the conclusions from the analysis of the results do not change when the 1064 nm channel observations are used instead.

6.2.2.1 Comparisons for the Raikoke June 2019 eruption

In the framework of this study we checked on the availability of overpasses of CALIPSO/CALIOP after the eruption of Raikoke volcano on the 22^{nd} of June. The number of available cases for the intercomparison is subject to a certain number of constraints. The collocated data set includes observations of calculated weighted extinction (532nm) altitude and SO_2 Layer Height (in km). The closest distances between the CALIOP footprint of the CALIPSO overpass and the locations of the TROPOMI centre pixels are selected respectively, to create collocated datasets. To illustrate the reliability of TROPOMI SO_2 LH product, we discuss in detail a selected case of collocated and concurrent TROPOMI–CALIPSO observations close to the detected SO_2 plume from Raikoke eruption, on the 23^{rd} of June 2019.

We use the 532 nm Total Attenuated Backscatter (TAB) data version 4.10 from one CALIPSO orbit in a qualitative approach, i.e., detection of cloud and aerosol layers and their heights. The Total Attenuated backscatter (TAB) signal strength [*Figure 7*, top] is color-coded such that blues correspond to molecular scattering and weak aerosol scattering and aerosols generally show up as yellow–red–orange. Stronger cloud signals are plotted in grey scales, while weaker cloud returns are similar in strength to strong aerosol returns and coded in yellows and reds. The TAB is sensitive to atmospheric particles: both water and ice droplets as well as various types of aerosols. This VFM image [*Figure 7*, middle] shows the aerosol type (i.e. the output from the aerosol classification algorithm) for all aerosol layers. The VFM describes the vertical and horizontal distribution of layers, including both cloud and aerosol. A 16-bit integer value describes the data bin of each level-0 data, with every bit range describing a characteristic.



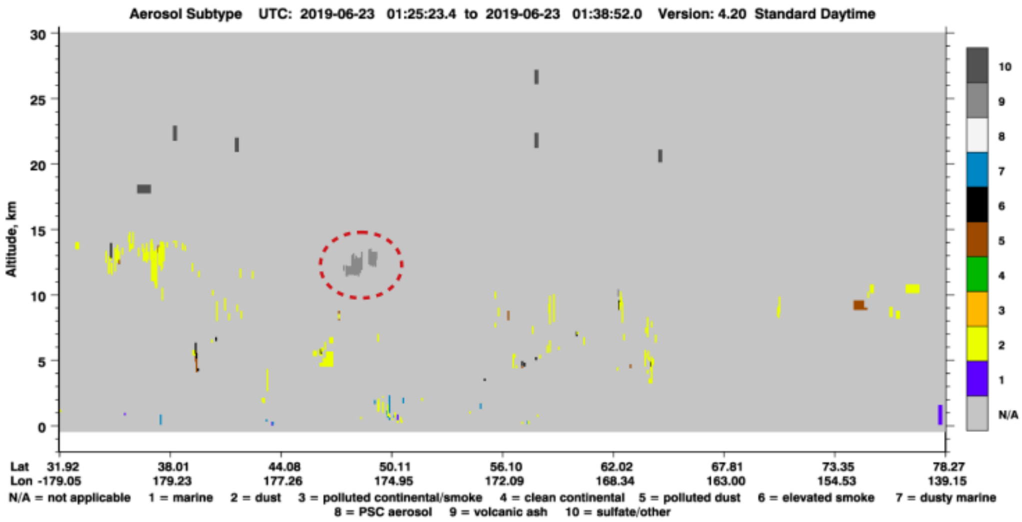


Figure 7. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on the 23rd of June. (Middle) Vertical feature mask image showing the location of all layers detected and (bottom) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (From* https://www-calipso.larc.nasa.gov/products/)

After detection of the aerosol features, they are then classified by the Scene Classification Algorithm (SCA), a suite of algorithms, into types and subtypes. As shown in *Figure 7* [bottom], the aerosol layer subtypes in the Version 4 CALIPSO product (Kim et al., 2018) include dust, marine, smoke, polluted dust, polluted continental, and clean continental, continental smoke, elevated smoke, dusty marine, PSC aerosol, volcanic ash, sulphate/other. The volcanic plume is marked with a dashed red circle. Several improvements to aerosol subtyping have been implemented in V4. The most fundamental change is that aerosol layers are now classified as either tropospheric aerosol or of certain stratospheric aerosol feature types. In previous versions, aerosol was only identified below the tropopause. Stratospheric aerosol subtypes have been introduced in V4 for ash, sulphate/other, smoke and polar stratospheric aerosol. The stratospheric aerosol subtyping algorithm performs well at identifying volcanic ash and sulphate above the tropopause based on manual verification. Note that below the tropopause, ash and sulphate plumes are given by the tropospheric aerosol subtypes: volcanic ash is often classified as dust or polluted dust and volcanic sulphate is often classified as elevated smoke. As a result, contiguous aerosol features crossing the tropopause will have aerosol subtypes which switch from tropospheric to stratospheric subtypes, depending on the relationship between the attenuated backscatter centroid altitude of the layer identified by the feature finder and the tropopause altitude. Further details can be found in the Data Quality Summary Document: https://www-calipso.larc.nasa.gov/resources/calipso_users_guide/qs/cal_lid_l2_all_v4-20.php.

The overpass considered here shows a representative case study of a CALIPSO orbit on the 23rd of June 2019 (black line), between 01:25:23 - 01:38:17 UTC. The CALIPSO attenuated backscatter coefficient cross section at 532 nm [*Figure 7*, top] shows the presence of aerosols between 10-13 km a.s.l. The CALIPSO feature-mask algorithm classifies all of the detected layers as ash aerosols [*Figure 7*, bottom].

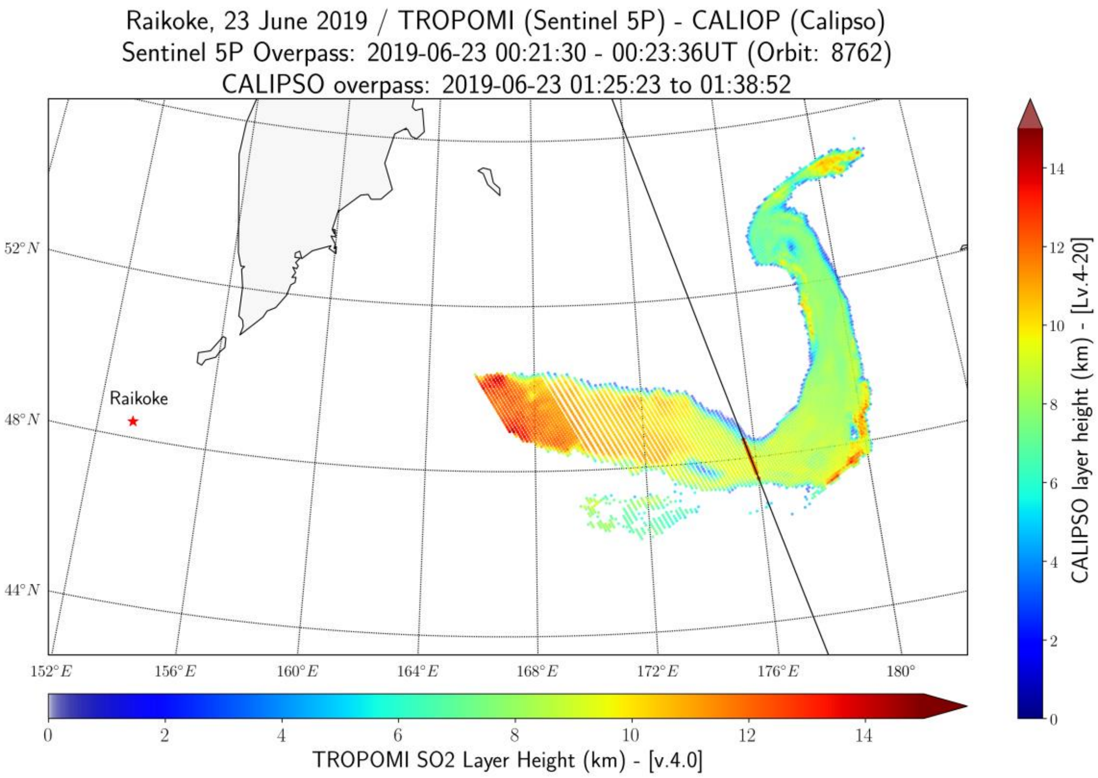


Figure 8. *SO_2 layer height retrieved or the TROPOMI measurements of the Raikoke volcanic eruption, measured on the 23rd of June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicate the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height values (in km).*

Figure 8 shows the TROPOMI SO_2 layer height pixels retrieved by the FP_ILM algorithm for SO_2 VCDs greater than or equal to 20 DU, qa_value > 0.5 and LHflag < 16 (cf. *Table 5),* overlaid with the calculated CALIPSO weighted extinction ALH pixel values (coloured circles) which are color-coded according to the range of height values (in km). The CALIOP overpass time of this area is between 01:25:23 and 01:38:26 UTC, and the TROPOMI overpass time is between 00:21:37 - 00:23:36 UTC, a time difference of approximately 60 min. Clearly, the plume shows several layers, with SO_2 layer heights ranging from 5-6 km up to 14 km on 23rd of June. In the area of the plume observed by both TROPOMI and CALIOP (47 – 50°N & 175 – 176°E), the CALIOP vertical feature mask and aerosol subtype mask identify some volcanic ash at approximately 13 km altitude, and meteorological clouds mixed with tropospheric aerosols (dust, polluted dust and elevated smoke) at lower altitudes. The clouds below the ash plume are visible in *Figure 7*, middle.

Although the collocation TROPOMI-CALIOP is not perfect [see *Figure 9*, left], the agreement between SO_2 LH and CALIOP weighted extinction altitude is satisfactory, and tends to confirm the presence of volcanic plumes. Both instruments yield high altitude values, however TROPOMI retrieves higher altitudes especially for the western part of the plume. A comparison scatterplot of collocated ash-flagged pixels is shown in *Figure 9*, right. The pixel-by-pixel comparison shows low correlation. On the following days, a large ash plume was observed which was more spatially extended. Seven TROPOMI (at 22/6 02:20; 23/6 00:20; 24/6 00:00; 25/6 01:30; 28/6 02:00; 29/6

02:00 and 30/6 01:30) and CALIPSO collocated overpasses (at 22/6 02:30; 23/6 01:30;24/6 00:30; 25/6 01:00; 28/6 03:00; 29/6 03:35 and 30/6 02:40) were identified. These volcanic cases for TROPOMI and CALIPSO overpasses are described on a per day basis in the Appendix.

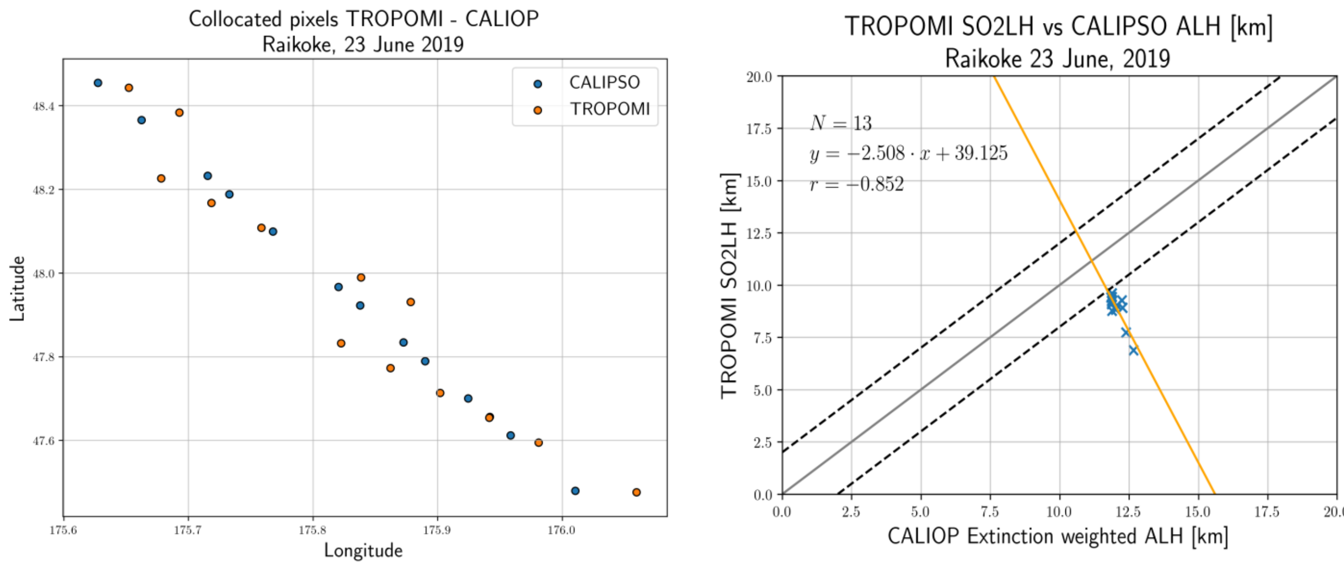


Figure 9. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI S0₂ LH and CALIPSO weighted extinction height product for the 23rd of June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations; the grey line is the 1:1 line.*

A statistical analysis has been performed using all 241 collocated pixels for the 22nd, 23rd, 24th, 25th, 28th, 29th and 30th of June 2019, for the Raikoke eruption. The number of available TROPOMI correlative observations and CALIPSO orbits used to produce the compared datasets are summarized in Table 7.

Table 7. Collocation points, criteria and statistics for the TROPOMI/S5P-CALIOP/CALIPSO comparisons.

Date	# counts	S5P range	CALIPSO range	y-int	R	slope
22 June 2019	8	7.5-10km	10-11.5km	-3.644	0.664	1.204
23 June 2019	13	6.5-10km	11.5-12.5km	39.125	-0.852	2.508
24 June 2019	22	10-15km	11.5-12.5m	73.04	-0.804	-5.025
25 June 2019	57	7.5-11km	11.5-13km	-0.642	0.732	0.806

28 June 2019	87	7.5-15km	11.5-15km	20.488	-0.393	-0.68
29 June 2019	46	10-13km	12.5-15km	19.51	-0.412	-0.616
30 June 2019	8	6.5-9.5km	11.5-14km	-2.36	0.844	8.52
All	**241**	**6-15km**	**10-15km**	**7.564**	**0.154**	**0.231**

Figure 10 shows the distribution of TROPOMI SO_2 LH and CALIOP calculated weighted heights differences for all days of collocation, as a scatter plot on the left and on a histogram representation on the right. The agreement is quite satisfactory with mean and median residual values around ~-2.4km and ~-3km respectively, and standard deviation of ~1.7km, with summary statistics given in Table 8.

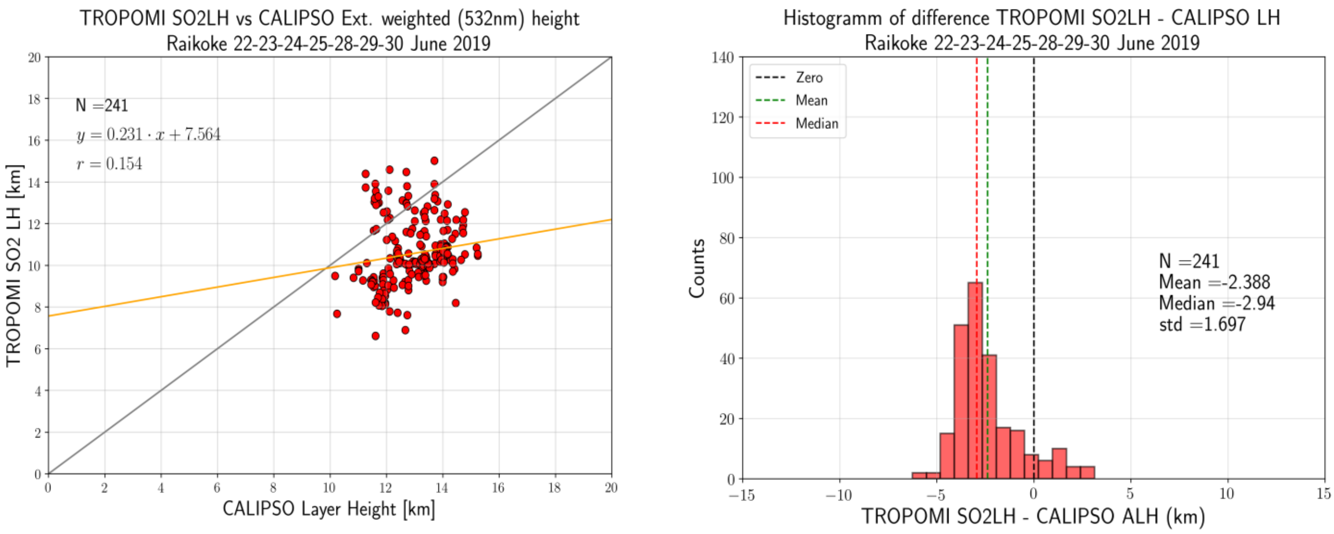


Figure 10. *(Left) Scatter plot of the TROPOMI SO_2 LH and CALIPSO weighted height for all days. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line. (Right) Histogram distribution of the absolute differences between TROPOMI SO_2 LH and the corresponding CALIPSO weighted extinction height measurements, calculated for the 241 collocated points.*

Table 8. TROPOMI/S5P–CALIOP/CALIPSO comparison statistics for the Raikoke correlative cases.

All Raikoke collocations [22,23,24,25,28,29 and 30 June 2019]	
Mean difference [km]	-2.388
Median difference [km]	-2.94

Standard deviation [km]	1.697
Min. absolute difference [km]	-6.31
Max absolute difference [km]	0.15

6.2.2.2 Comparisons for the Nishinoshima August 2020 eruption

The Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Aqua satellite acquired a true-color image of the volcano on August 1st as Nishinoshima poured huge amounts of volcanic ash into the sky (*Figure 11*, left). The overpass considered here shows a representative case study of a CALIPSO orbit on August 1st 2020 (black line), between 04:19:30 - 04:32:59 UTC [*Figure 11*, right]

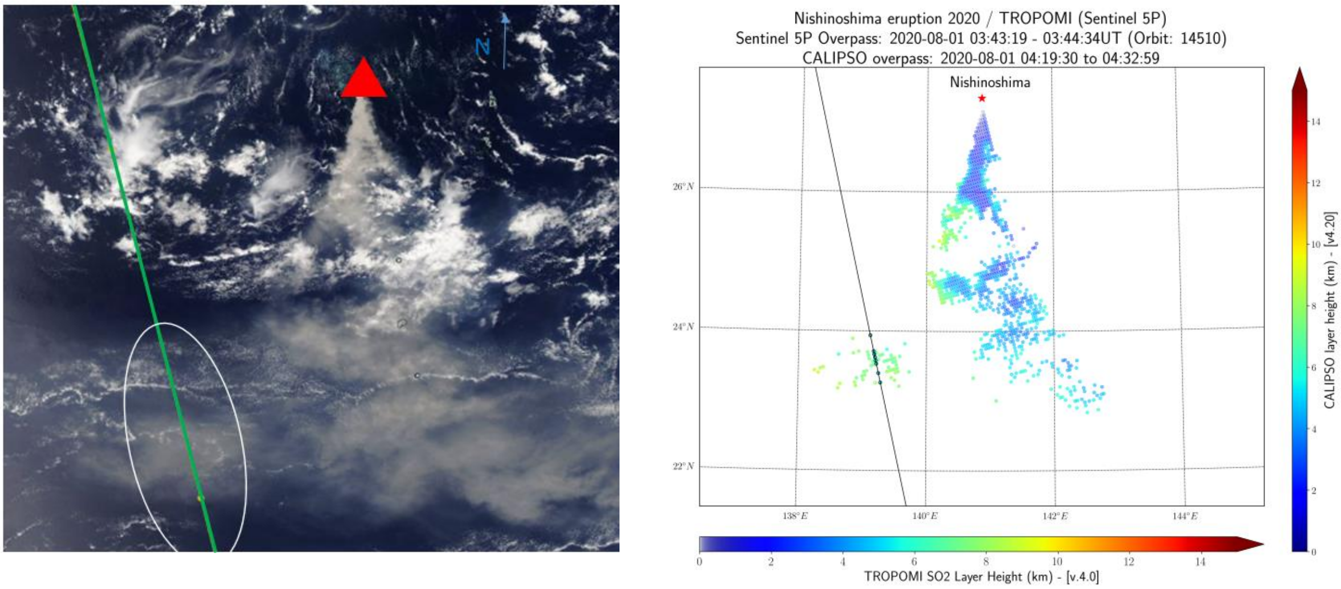


Figure 11. *[Left] Image of detect ash plume as captured by the MODIS/Terra satellite, on 1 August, (*https://worldview.earthdata.nasa.gov/*, last access: 01 June 2021). [Right] SO_2 layer height retrieved or the TROPOMI measurements of the Nishinoshima volcanic eruption, measured on the 1st of August 2020. The black line indicate the CALIPSO ground track and the coloured circles indicate weighted extinction height values (km)*

Spatial collocations for the 1st of August 2020 are shown in *Figure 12* [left], where the scatterplot of height values is shown in the right. The geographical collocations between TROPOMI and CALOP are not optimal, however the agreement between SO_2 LH and CALIOP weighted extinction altitude is satisfactory, and tends to confirm the presence of volcanic plumes. The CALIPSO observations (see *Figure 13*) confirm the presence of volcanic cloud up to 6-7 km, while S5P reports a slightly higher loads, at ~7.5km.

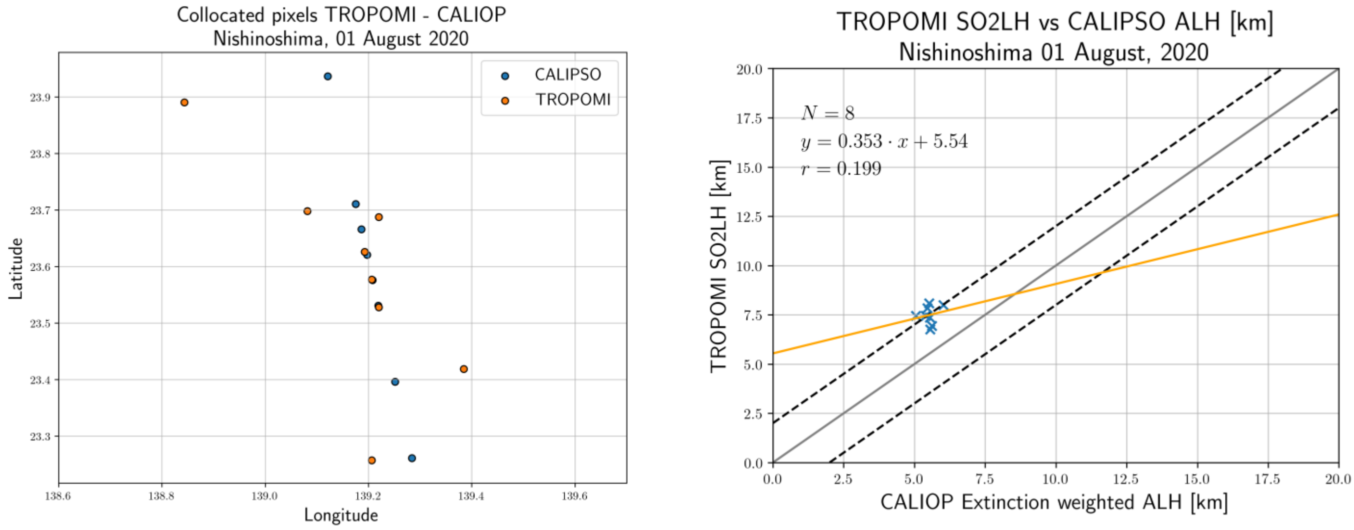
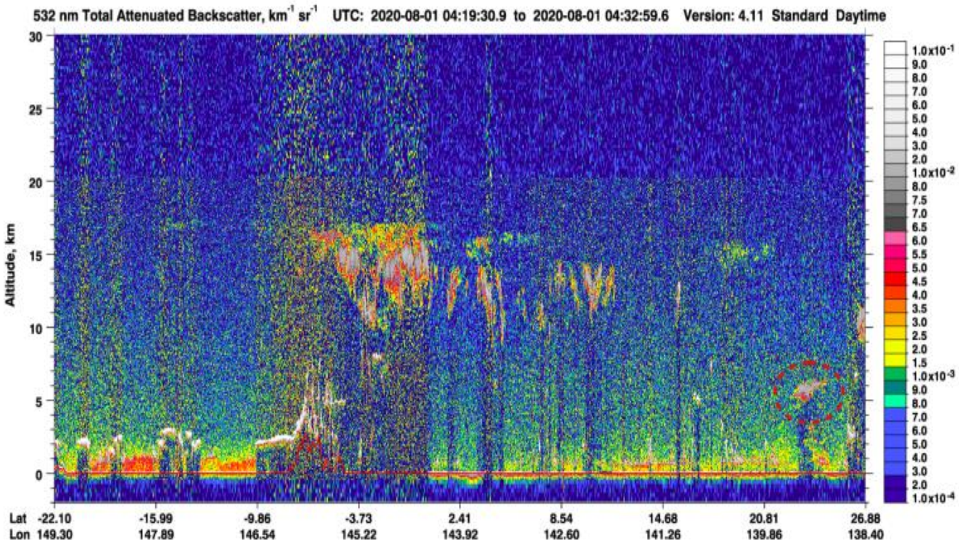
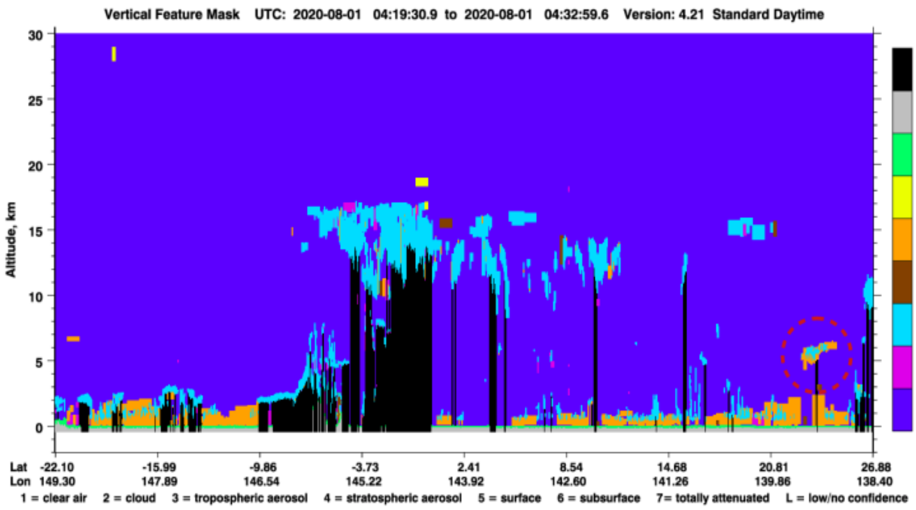


Figure 12. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 1st of August 2020. The orange line is the regression line of the TROPOMI-CALIPSO observations; the grey line is the 1:1 line.*


532 nm Total Attenuated Backscatter, km⁻¹ sr⁻¹ UTC: 2020-08-01 04:19:30.9 to 2020-08-01 04:32:59.6 Version: 4.11 Standard Daytime
Altitude, km
Lat -22.10 -15.99 -9.86 -3.73 2.41 8.54 14.68 20.81 26.88
Lon 149.30 147.89 146.54 145.22 143.92 142.60 141.26 139.86 138.40


Vertical Feature Mask UTC: 2020-08-01 04:19:30.9 to 2020-08-01 04:32:59.6 Version: 4.21 Standard Daytime
Altitude, km
Lat -22.10 -15.99 -9.86 -3.73 2.41 8.54 14.68 20.81 26.88
Lon 149.30 147.89 146.54 145.22 143.92 142.60 141.26 139.86 138.40
1 = clear air 2 = cloud 3 = tropospheric aerosol 4 = stratospheric aerosol 5 = surface 6 = subsurface 7= totally attenuated L = low/no confidence

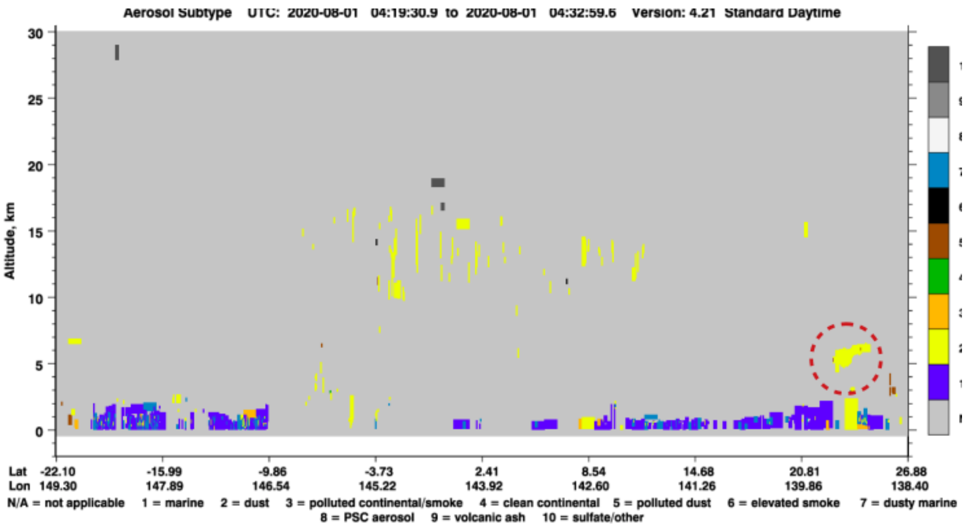


Figure 13. *(Top) CALIOP total attenuated backscatter profile for the Nishinoshima eruption on 01 August 2020 (middle) Vertical feature mask image showing the location of all layers detected and (bottom panel) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (*https://www-calipso.larc.nasa.gov/products/ *)*

6.2.2.3 Comparisons for the La Soufriere April 2021 eruption

On the afternoon of April 11, 2021, the VIIRS satellite instrument on board Suomi NPP acquired the image shown in *Figure 14*, left, showing that winds carried much of the ash and gas east from Saint Vincent reaching the Barbados. The scene was quite complex as there were many low, mid- and high level clouds. Despite this complication, the massive triangular volcanic plume from La Soufrière is well visible in the imagery, while ash is visible as brown-yellow layers. *Figure 14*, right, illustrates the locations and heights of detected ash layers observed by CALIOP and TROPOMI, for April 1st. The black line shows the CALIPSO orbit track.

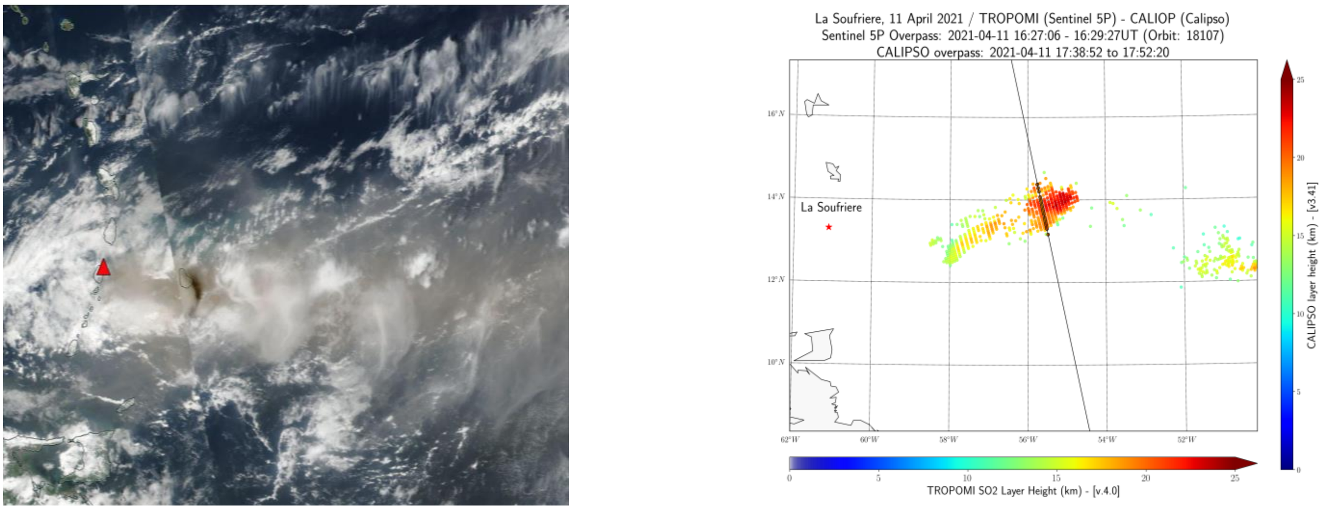


Figure 14. *[Left] Image of detect ash plume as captured by the VIIRS/Suomi-NPP satellite, on the 11st of April (*https://worldview.earthdata.nasa.gov/*, last access: 11 April 2021). [Right] TROPOMI SO_2 layer height retrieved for*

the La Soufriere volcanic eruption. The black line indicates the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height values (in km).

Spatial collocations for the 11st of April 2021 are shown in *Figure 15* (left), where the scatterplot of collocations is shown in the right column and the scatter plots in the right column. In this case, both CALIPSO and TROPOMI collocated pixels confirms the presence of volcanic cloud up to and around ~20km. For this case, v3 data products are available only which do not provide subtyping (*Figure 16*).

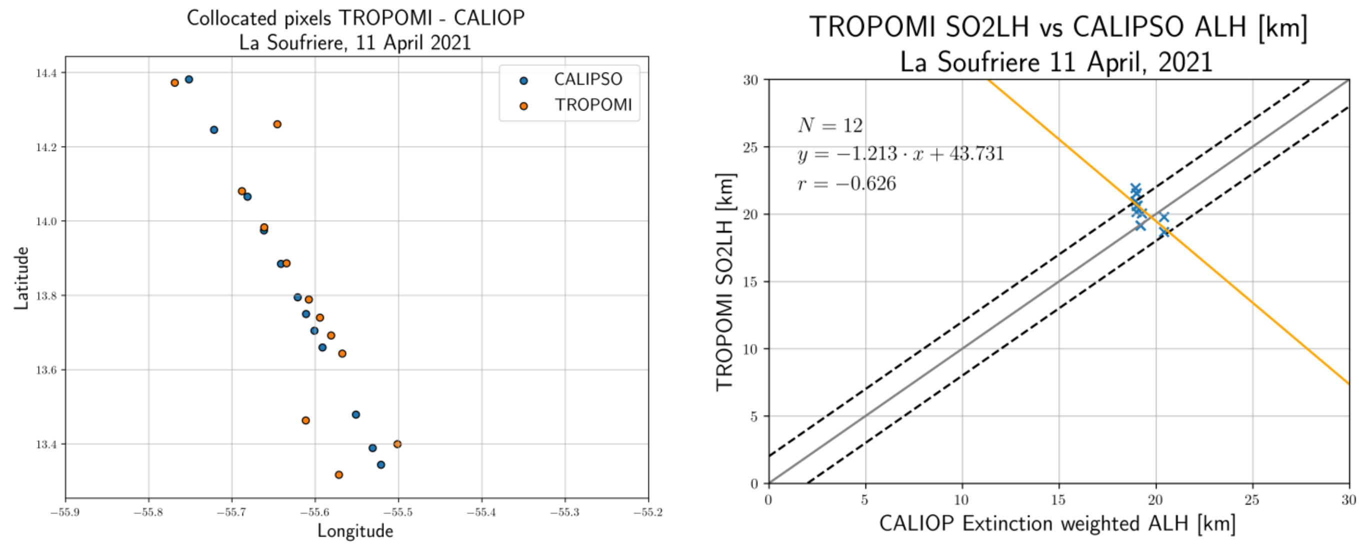


Figure 15. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 11 April 2021. The orange line is the regression line of the TROPOMI-CALIPSO observations; the grey line is the 1:1 line.*

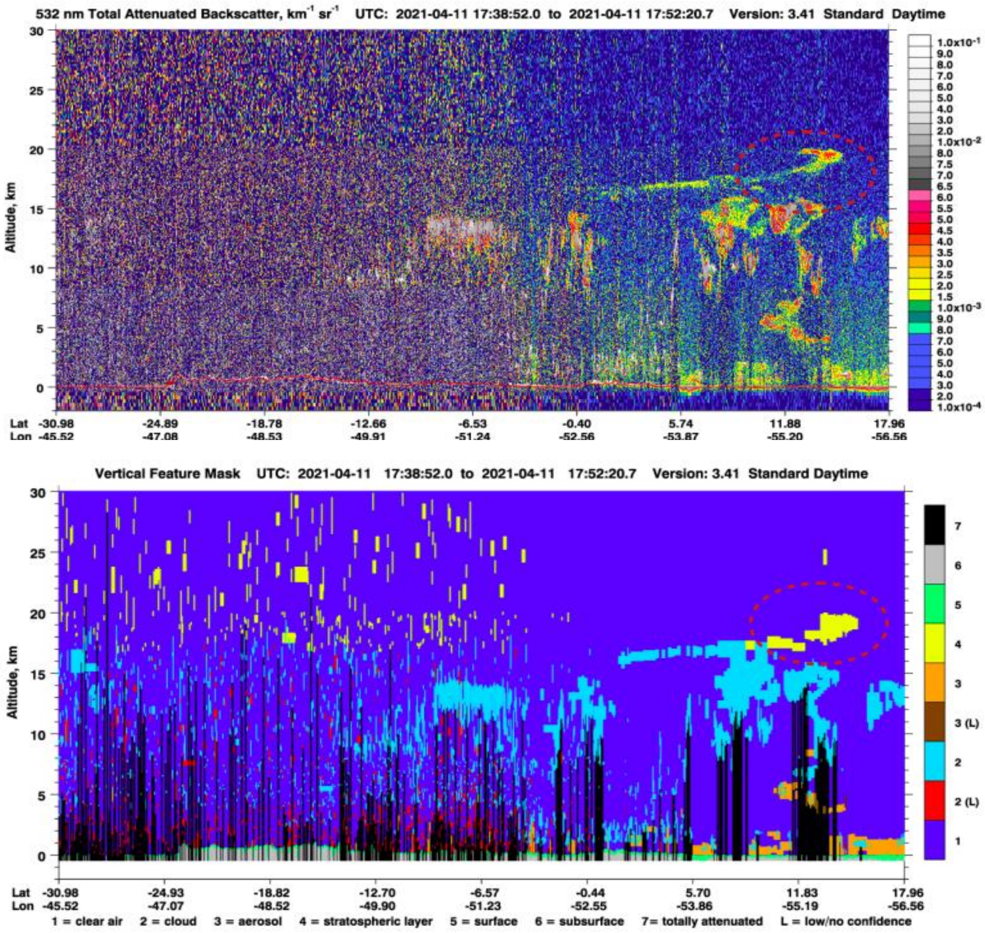


Figure 16. *(Top) CALIOP total attenuated backscatter profile for the La Soufriere eruption on 11 April 2021 (middle) Vertical feature mask image showing the location of all layers detected and (bottom panel) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (*https://www-calipso.larc.nasa.gov/products/*)*

6.2.2.4 Comparisons for the Sinabung February 2018 eruption

On 19 February 2018, at 08:53 local time, the Indonesian stratovolcano Mount Sinabung on Sumatra (2460 m summit elevation) generated a dark grey plume with a high volume of ash that quickly rose to an estimated 15–17 km a.s.l. Although the eruption was small in spatial extent and rather short-lived, by mere accident there was a perfect overpass with the CALIOP instrument (*Figure 17*, right). The CALIOP track goes straight through the core of the volcanic ash cloud and across the north–south gradient in cloud tops. At 11:10 a.m. local time (04:10 Universal Time) on February 19, the Moderate Resolution Imaging Spectroradiometer (MODIS) on the NASA Terra satellite captured a natural-color image of the eruption, just a few hours after it began (*Figure 17*, left). A gray ash plume can be seen to rise nearly directly upward over the volcano and far over the island.

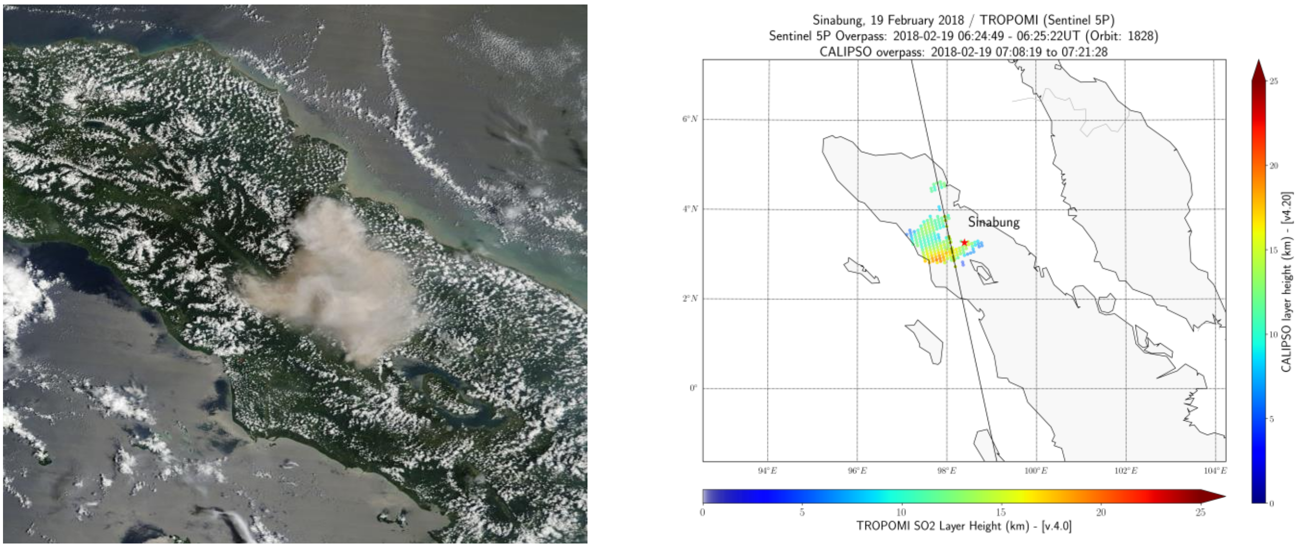


Figure 17. *Sinabung eruption on the 19th of February 2018. (Left) MODIS/Terra satellite captured image around 04:10 UTC. (Right) SO_2 LH for the TROPOMI measurements around 06:30 UTC and the CALIPSO ground track, with measurements as colored circles at an overpass time around 07:10 UTC.*

The CALIOP overpass time of this area is between 07:09:56 and 07:11:26 UTC, and the TROPOMI overpass time is between 06:24:23 and 06:26:00 UTC, a time difference of approximately 45 min. The CALIOP data clearly show not only a cloud–ash layer around 15 km altitude but also two cloud–ash structures extending from the ground up to approximately 10 km altitude, *Figure 18*. As shown in *Figure 19* (top) the presence of clouds (with light blue in VFM plot) appear along the CALIPSO path indicated by the stronger attenuated backscatter than the aerosol layer.

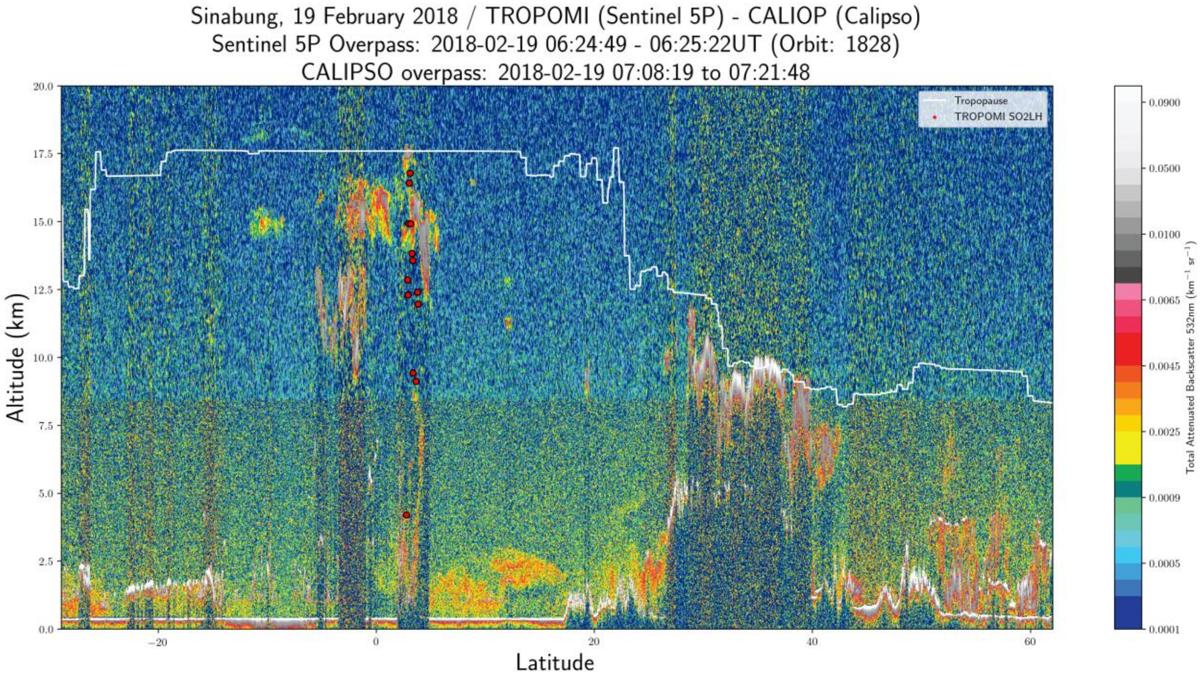


Figure 18. *Sinabung, 19th of Februray 2018, 07:15UTC. The colours show the CALIOP/CALIPSO total attenuated backscatter at 532nm, the red dots show the TROPOMI SO2 LH and the white line the tropopause level.*

This case of mixing between ash and clouds over a volcanic eruption renders the retrieval of the ash plume altitude very difficult, since the CALIPSO algorithm cannot separate clouds from aerosols, especially when the aerosol amount is low. As shown in *Figure 19*, bottom panel, the CALIPSO feature mask does not identify hardly any of these backscatter signals as aerosol. The high-altitude structures are flagged as regular clouds, and the below-cloud structure as "totally attenuated", even though clearly the attenuation is not complete. The lack of aerosol masking in the feature mask most likely is related to liquid water or ice contaminating the volcanic ash (Hedelt et al., 2019). The maximum TROPOMI SO_2 LH value agrees with the maximum backscatter height in CALIOP between 2-3° latitude. The CALIOP data also suggest that backscatter signals going northern of 3° latitude, are weaker, which might indicate less dense ash or clouds. In case of a semi-transparent cloud or ash plume, it could be expected that TROPOMI SO_2 LH heights are lower than the actual height of the cloud or ash plume due the presence of bright clouds.

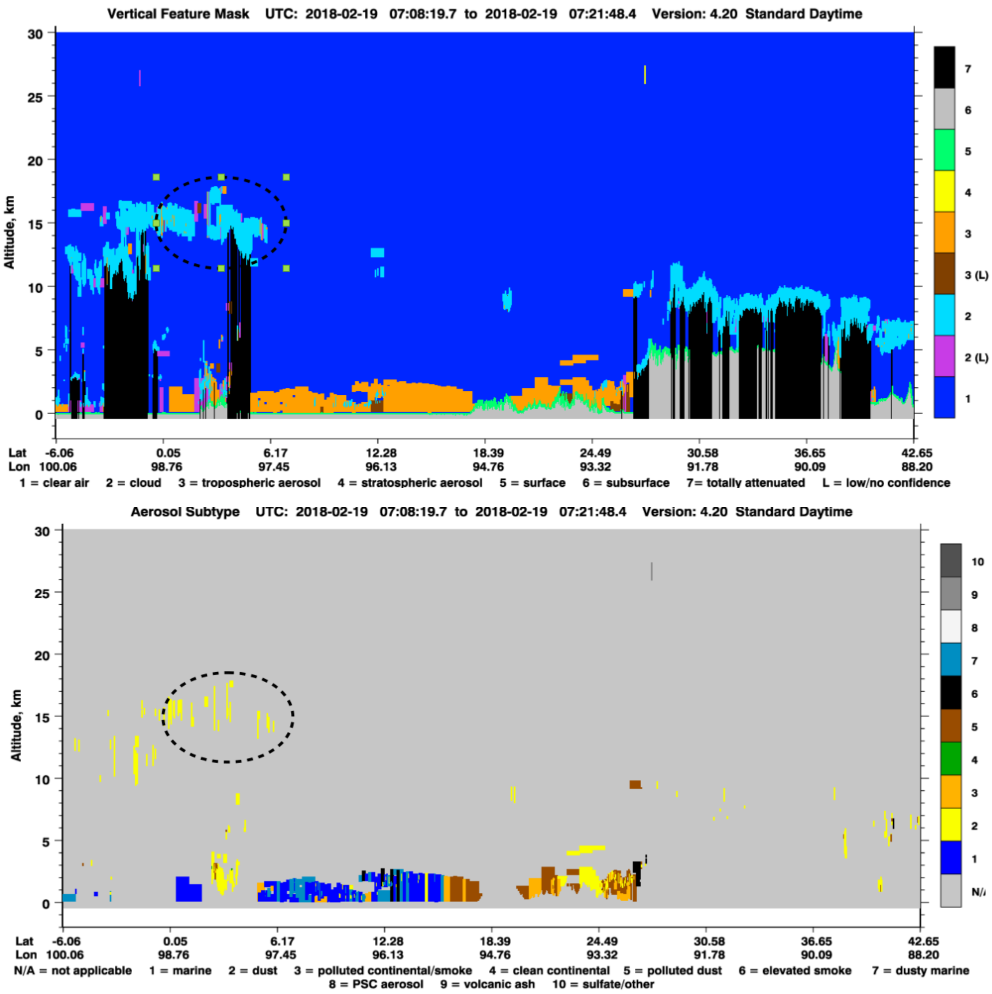


Figure 19. *Sinabung eruption on 19 February 2018 (top) Vertical feature mask image showing the location of all layers detected and (bottom panel) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP (https://www-calipso.larc.nasa.gov/products/).*

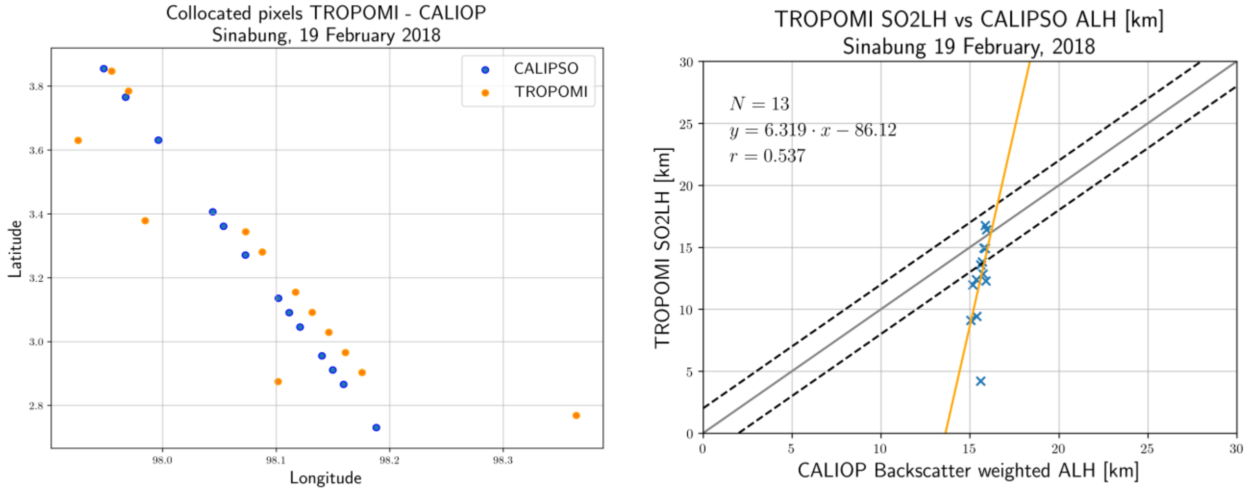


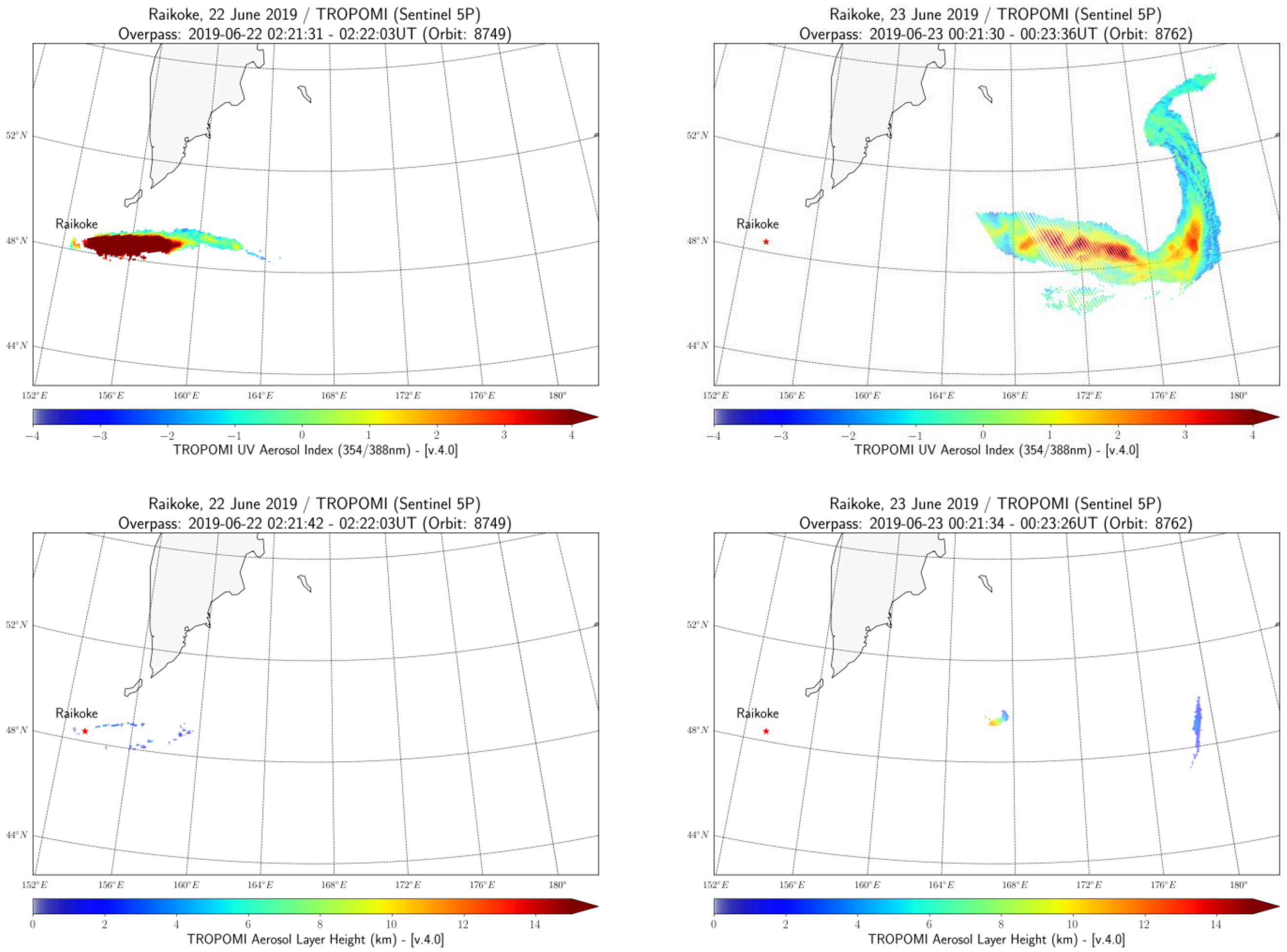
Figure 20. *(Left) The latitude/longitudes of the collocated pixels. (Right) Comparison between TROPOMI SO_2 LH and CALIPSO weighted backscatter height product for the 19 February 2018. The orange line is the regression line of the TROPOMI-CALIPSO observations; the grey line is the 1:1 line.*

As far as the actual comparison in concerned, the locations of the collocated points are shown in *Figure 20*, left, while the statistics are given in the right panel. As already discussed above, the CALIPSO algorithm considers that the ash plume is strictly located at 15km, unable to view the underlying plumes due to cloud contamination. A similar issue is present in the TROPOMI algorithms, which render this case study as quite challenging; the clear separation of cloud, ash and SO_2 layers is impossible to perform in this case.

6.2.2.5 Investigating the S5P ALH observations as possible verification dataset

The TROPOMI Aerosol Layer Height (ALH) product focuses on the retrieval of vertically localized aerosol layers in the free troposphere, such as desert dust, biomass burning aerosol, or volcanic ash plumes in layers for cloud-free scenes. The retrieval of the aerosol height is based on absorption in the O_2 A band in the near-infrared wavelength range (759 and 770 nm) and the operational TROPOMI ALH processor is based on neural network methodology. The data contain the aerosol_mid_pressure and aerosol_mid_height, which provide the air pressure at the centre of the aerosol layer and the height at the centre of the aerosol layer relative to the geoid, respectively. The aerosol layer mid pressure is computed for pixels that are relatively cloud-free and have an UVAI, UV Aerosol Index, greater than 0. As a user guideline, it is recommended to use only those pixels that have an UVAI larger than 1, and contain no sun glint as the ALH is very sensitive to cloud contamination. However, aerosols and clouds can be difficult to distinguish, and ALH is computed for all FRESCO effective cloud fractions smaller than 0.05. The *Fast Retrieval Scheme for Clouds from the Oxygen A Band*, Fresco, algorithm is described by Koelemeijer et al., 2011 and Wang et al., 2012. Cloud masks are available from FRESCO and VIIRS, and are strongly recommended to filter for residual clouds. A sun glint mask is also available to screen sun glint regions, which are not filtered beforehand. These and other sources of uncertainties are filtered out using qa_value values > 0.5. For further details on the product and the algorithm refer to [RD05] and [RD06].

Both the S5P/TROPOMI Aerosol Layer Height and Aerosol Index OFFL observations for the week 22nd to 29th of June 2019 where downloaded, filtered and examined for the possibility of collocations to the SO_2 LH observations. In *Figure 21*, the S5P/TROPOMI OFFL Aerosol Index [upper row], Aerosol Layer Height [middle row] and v4.0 SO_2 Layer Height [bottom row] are shown for the 22nd [left] and the 23rd [right] of June 2019 over the Raikoke eruption. While there appear to be a number of observations, in the thick parts of the plumes from the 22nd and the 23rd with associated AI values > 1 [upper row], there exist no ALH retrievals for these parts of the plumes [middle row] and subsequently no collocations with the SO_2 LH product [bottom row.] As the S5P/TROPOMI Aerosol Layer Height and Aerosol Index products mature we will revisit them in order to examine possible collocations and comparisons.



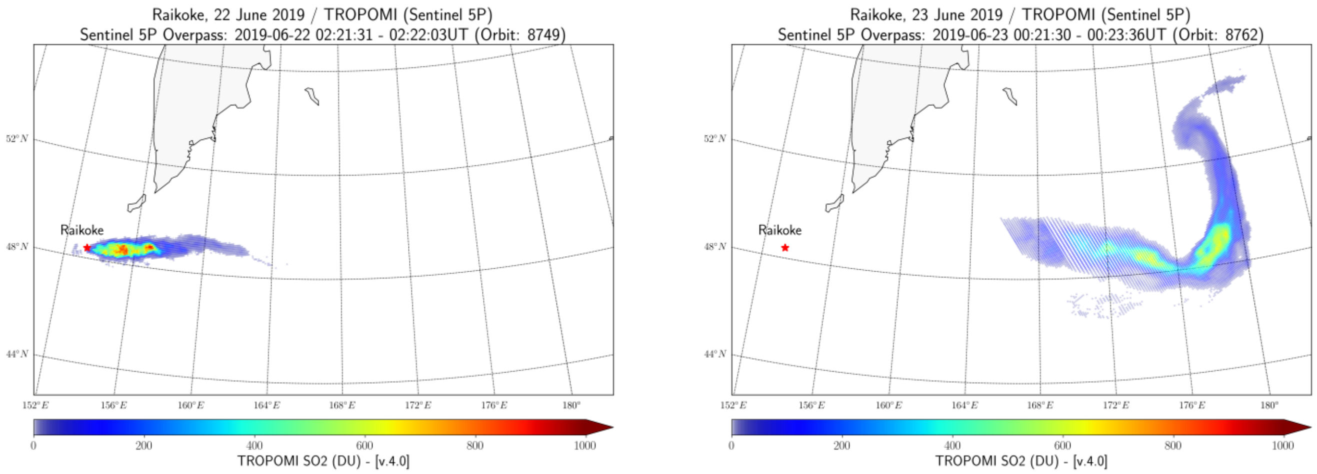


Figure 21. *S5P/TROPOMI Aerosol Index [upper row], Aerosol Layer Height [middle row] and SO_2 Layer Height [bottom row] for the 22nd of June 2019 [left column] and the 23rd of June 2019 [right column.]*

6.2.3 Summary of the comparisons with the CALIPSO observations

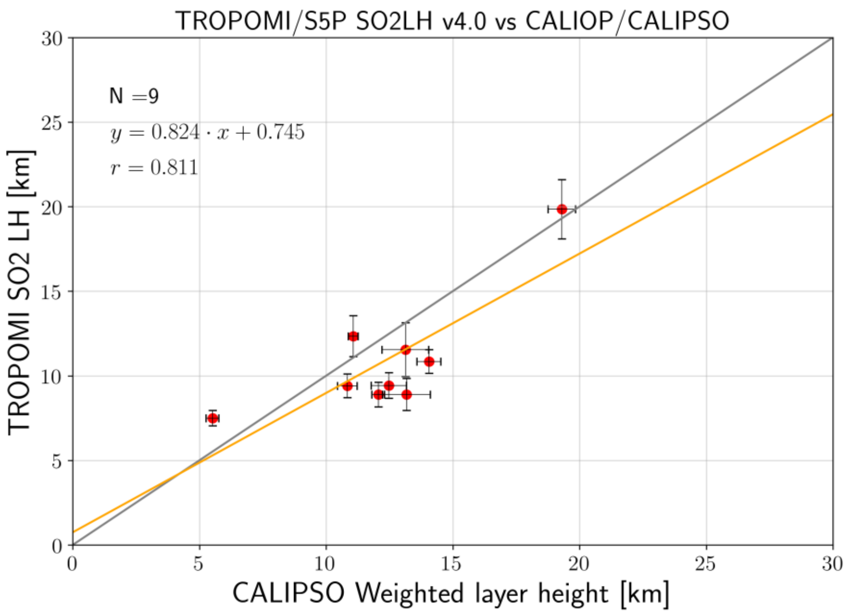


Figure 22. *Scatter plot of the mean daily average reported SO_2 LHs by TROPOMI/S5P and CALIOP/CALIPSO for all available collocated eruptive days.*

- The S5P/TROPOMI SO_2 layer height product has been compared against independent CALIOP satellite instrument ash layer height observations. In general, the SO_2 LH satellite products show good agreement, within 2-3km, with the calculated weighted extinction (532nm) height product from CALIOP demonstrating that the volcanic plumes can be detected and characterized. For multilayer cases with mixed aerosol plumes and presence of clouds the retrieval is challenging, as well as for optically very thin plumes.

- In the quantitative assessment of the performance of TROPOMI SO_2 LH, 241 collocated points between CALIOP and TROPOMI for Raikoke case, were identified, offering a unique opportunity for the validation of the SO_2 LH product. On average, the mean values of the data products reveal similar height ranges between TROPOMI and CALIOP with a mean difference of ~-2.4±1.7km, the differences ranging between ~-6km and 0km. Furthermore, two additional cases from Nishinoshima and La Soufriere eruptions, showed good agreements, with the height difference being within ~1.5km.

- A summary plot of the comparisons between S5P SO_2 LHs and CALIOP/CALIPSO weighted ALH is shown in *Figure 22*, where the mean plume height reported for each of the 9 days of collocations is shown as a scatter plot. The mean SO_2 LHs, as expected, follow quite closely a straight line, with slope of 0.82 and y-intercept of ~0.74km, with the correlation coefficient is also excellent, at 0.81. 7 days belong to the Raikoke eruptive period, 1 case to the La Soufriere comparison on the 11th of April 2021 and 1 case belong to Nishinoshima for the 1st of August 2020.

- For the Sinabung eruption, the comparison with CALIPSO satellite data shows that TROPOMI SO_2 layer height data provide volcanic ash cloud heights comparable to heights measured by CALIPSO for optically thick volcanic ash clouds. The CALIPSO measurements clearly show an attenuation by ash or aerosols at altitudes around 15–18 km, however the layers below are classified as over-saturated and the spread of SO_2 layer heights sensed by TROPOMI are not available by the CALIPSO database. Fresh volcanic plumes are typically rich in water vapor, and also the volcanic clouds contain high concentrations of water droplets. For this reason, the classification in the CALIPSO vertical feature mask sometimes fails to pick up the volcanic ash or sulfate aerosol because of competing clouds (Hedelt et al. 2019). Nevertheless the altitudes of the identified features are likely those of the volcanic plumes themselves, given the collocations in time and space.

- The features identified as volcanic ash by the CALIOP aerosol subtype mask are captured by the TROPOMI algorithm, but the surrounding clouds often affect the retrieval. The comparison of the TROPOMI SO_2 LH product within this project shows promising capability in detecting volcanic presence, with some limitations related to the eventual clouds. It is important to note that the CALIOP lidar only indicates the height of the ash plume and not the SO_2 height. Although ash and SO_2 plumes are often collocated, this is not always the case, making direct comparisons challenging.

7 Conclusions

In this validation report, the SO_2 layer heights reported by the FP_ILM SO_2 LH algorithm applied on S5P/TROPOMI observations as part of the European Space Agency S5P+ Innovations project have been compared to independent observations for four major volcanic eruptions since the beginning of S5P/TROPOMI operations. Summaries of the verification/validation activity, separated for the IASI/Metop and the CALIOP/CALIPSO correlative datasets, can be read in Sections 6.1.3 and 6.2.3. We note in summary here the following:

The S5P/TROPOMI SO_2 layer height product has been compared against IASI/Metop SO_2 layer height for the eruptive periods of the Raikoke volcano, 22 June to 30 July 2019, the Taal volcano, 13 January 2020, the Nishinoshima eruptive period, July 2020, and the La Soufriere eruptive days, April 10th to 11th, 2021. Two different algorithms were examined, the official EUMETSAT ACSAF algorithm, labeled ULB/LATMOS, and the University of Oxford, AOPP, algorithm.

Considering the comparisons of the mean SO_2 plume height reported for each of the 27 days of collocations between S5P and IASI/AOPP, the mean SO_2 LHs are shown to follow quite closely a straight line, with slope of 0.9 and y-intercept of ~1.2km, with the correlation coefficient is also very promising, at 0.66. 24 days belong to the Raikoke eruptive period, 2 days to La Soufriere, while the Taal comparison for the 13th of January 2020 appears as the outlier point, with low IASI/AOPP SO_2 LH (~6km) while S5P reports a high LH at ~10km. Similarly, for the S5P to IASI ULB/LATMOS comparisons, the co-variability is excellent between height, with a with slope of ~1 and y-intercept of ~0.8km, and correlation coefficient of 0.73

Considering the comparisons of the S5P SO_2 plume height to the CALIOp/CALIPSO ash plume, 241 collocated points between CALIOP and TROPOMI for Raikoke case, were identified, offering a unique opportunity for the validation of the SO_2 LH product. On average, the mean values of the data products reveal similar height ranges between TROPOMI and CALIOP with a mean difference of ~-2.4±1.7km, the differences ranging between ~-6km and 0km. Furthermore, two additional cases from Nishinoshima and La Soufriere eruptions, showed good agreements, with the height difference being within ~1.5km. The case of the Sinabung eruption in February 2018 was also extensively examined, however the CALIOP instrument was unable to sense ash below cloud layers, rendering this case study as quite challenging as the clear separation of cloud, ash and SO_2 layers is impossible in this case.

Overall, we report an excellent agreement between the IASI/Metop and TROPOMI/S5P SO_2 plume heights, within 2km for S5P SO_2 loads greater than 20 D.U., well within the user requirements, and typical mean differences of less than 1km. Furthermore, we find a very good agreement with the CALIOP/CALIPSO ash plume heights, with typical differences of 2km.

We recommend that this product is used for volcanic plume studies following the filtering criteria of qa_value > 0.5, SO_2 optimized column > 20 D.U. and LH flag < 16.

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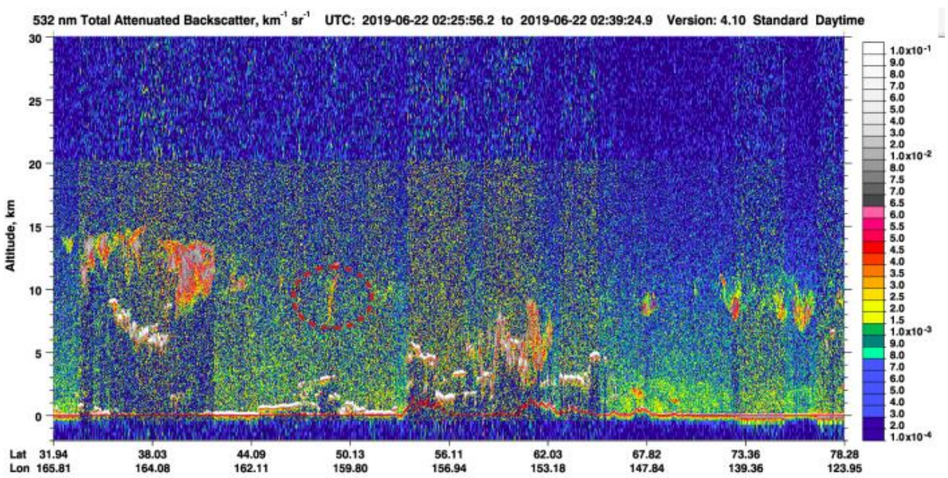
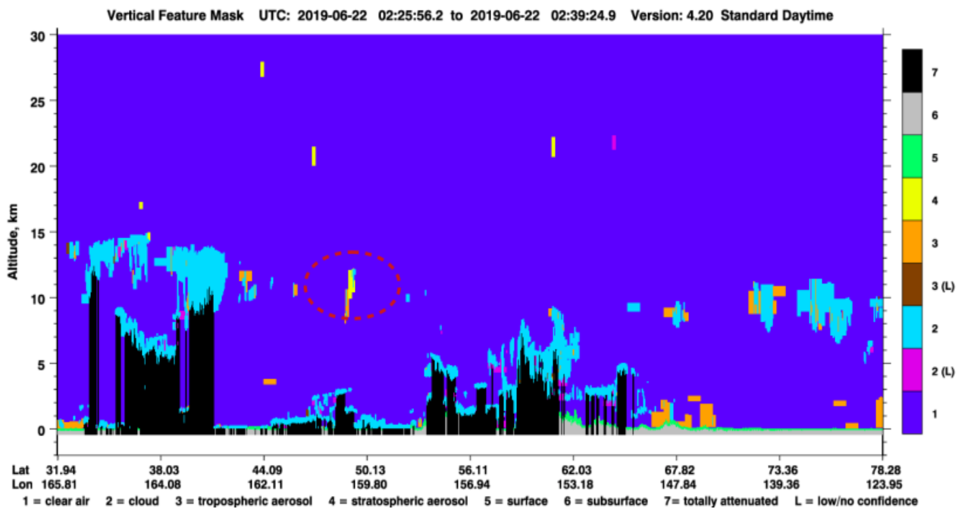
APPENDIX

In the following section, we present triplet plots for all comparative cases between the CALIOP/CALIPSO observations and collocated TROPOMI/S5P measurements for all Raikoke 2019 eruptive days studied. The first plot shows the total attenuated backscatter field at 532nm at the top, the vertical feature mask in the middle and the aerosol subtype at the bottom. The second plot shows the two datasets superimposed in space and time. The third plot shows the geographic location of the collocative data points on the left and the scatter plot of the comparisons, including statistics, on the right.





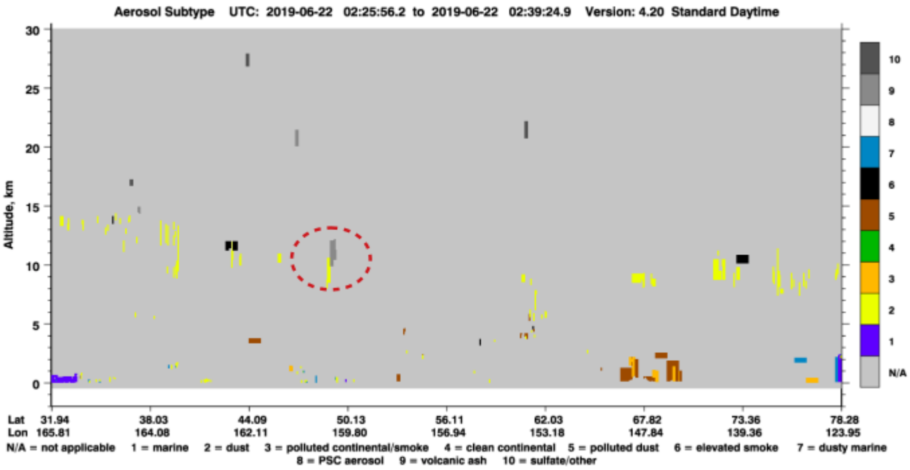


Figure 23. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 22 June 2019 (middle) Vertical feature mask image showing the location of all layers detected and (bottom panel) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

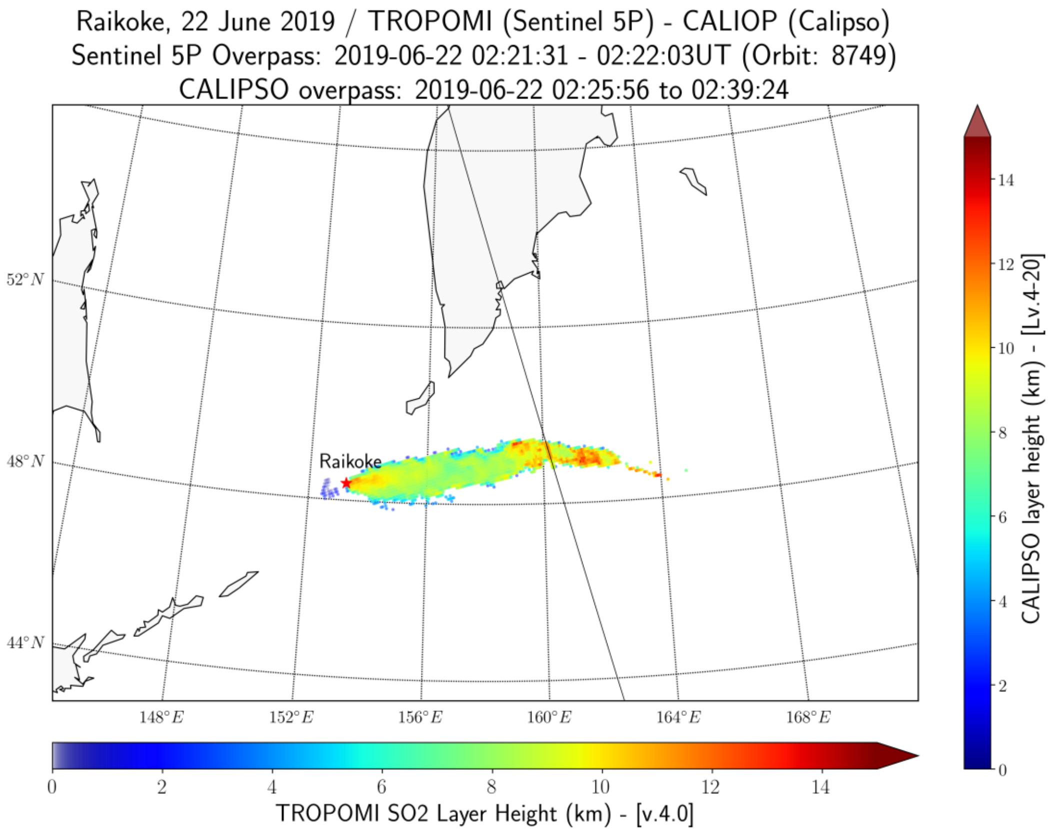
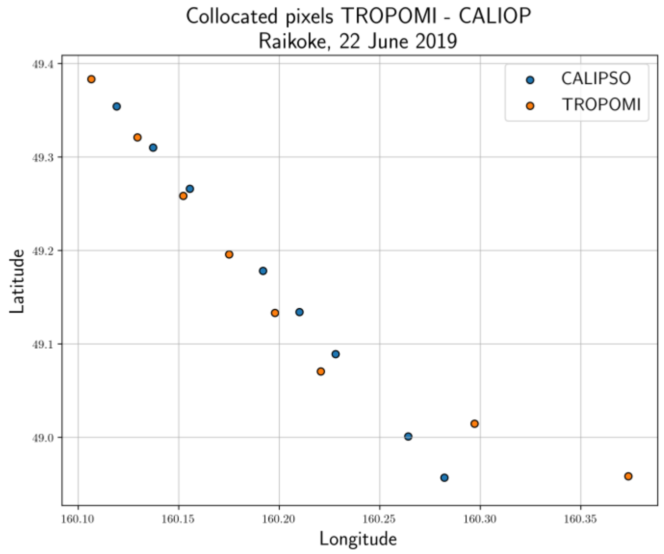


Figure 24. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 22 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicates the CALIPSO ground track and the coloured circles along the line indicate the weighted extinction height values (in km), for the results shown in* ***Figure 23****.*



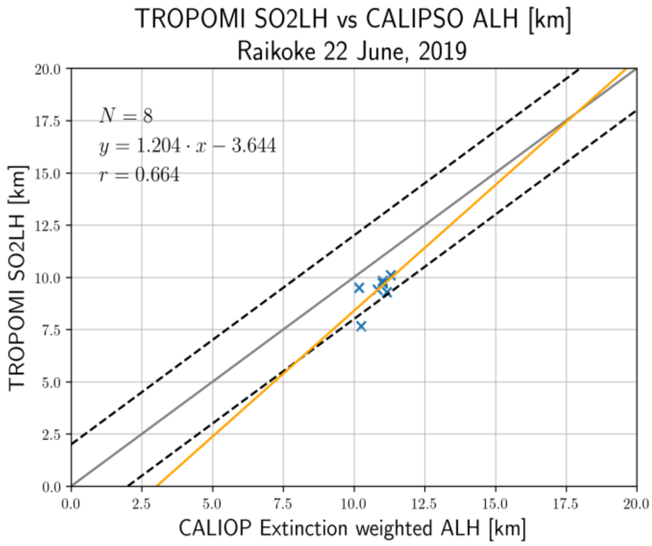


Figure 25. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 22 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line.*

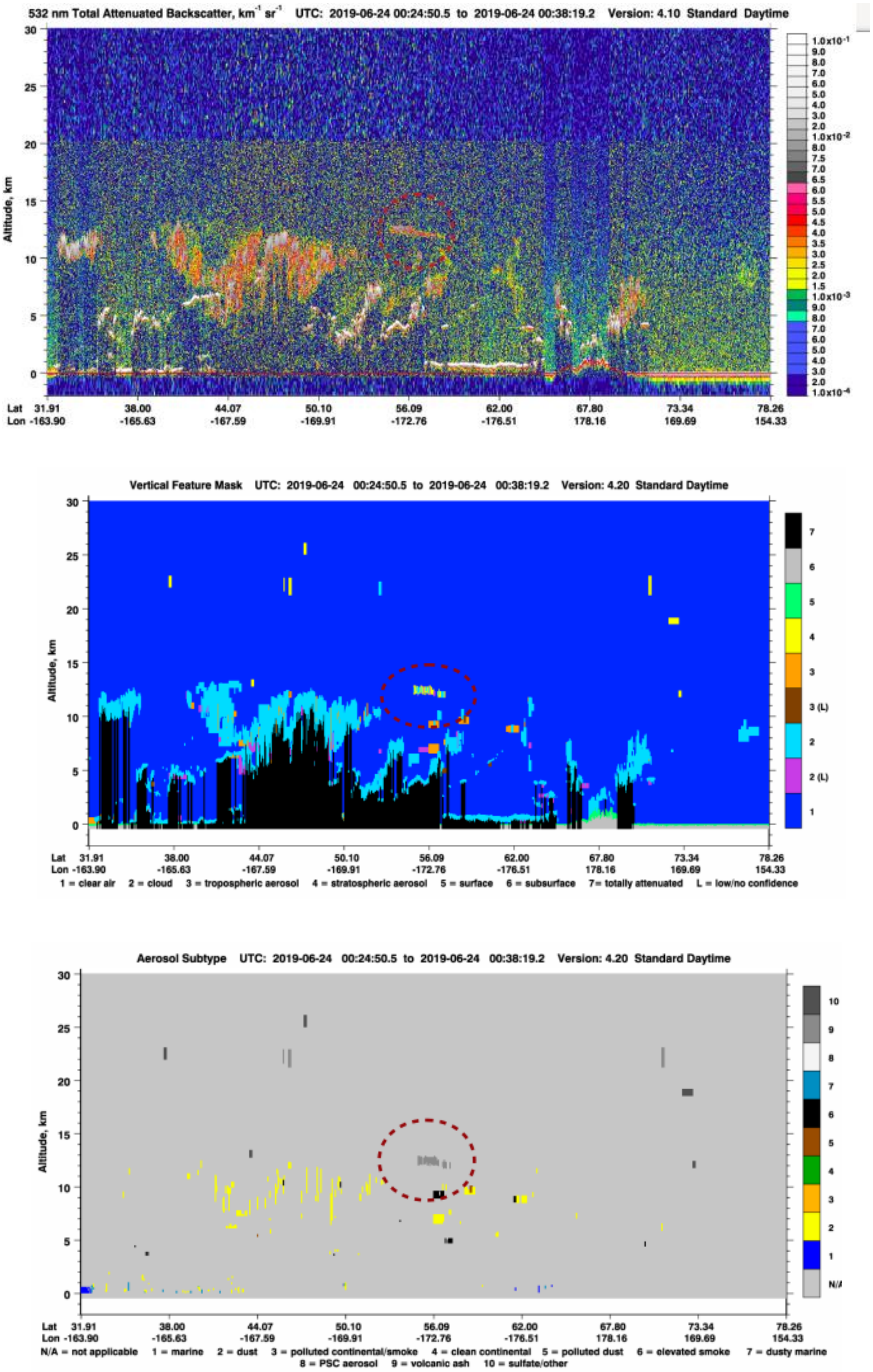


Figure 26. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 24 June 2019 (middle) Vertical feature mask image showing the location of all layers detected and (bottom panel) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

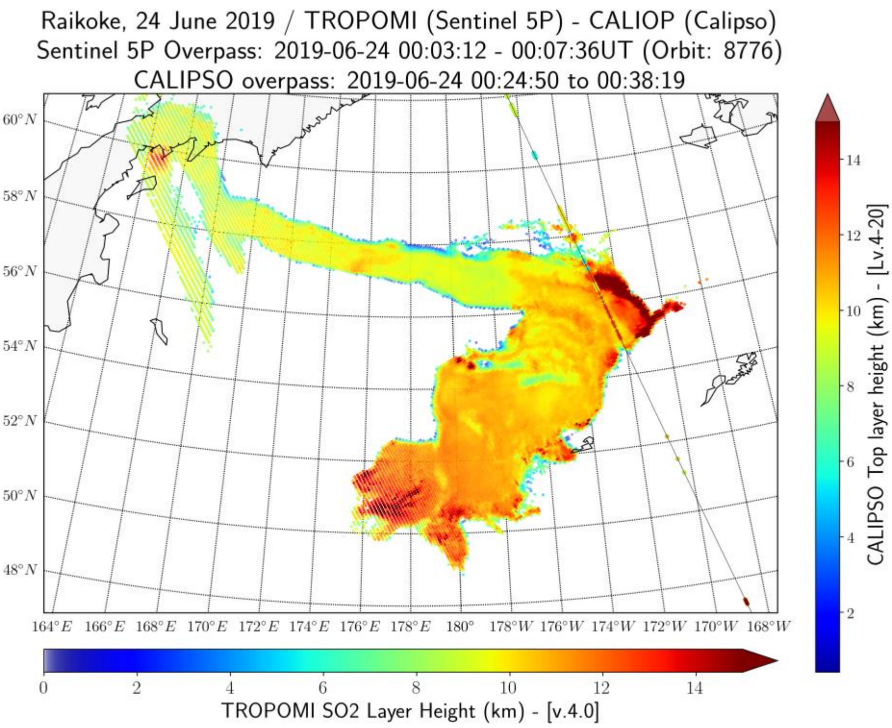


Figure 27. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 24 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicates the CALIPSO ground track and the coloured circles along the line indicate the weighted extinction height product values (in km), for the results shown Figure 26.*

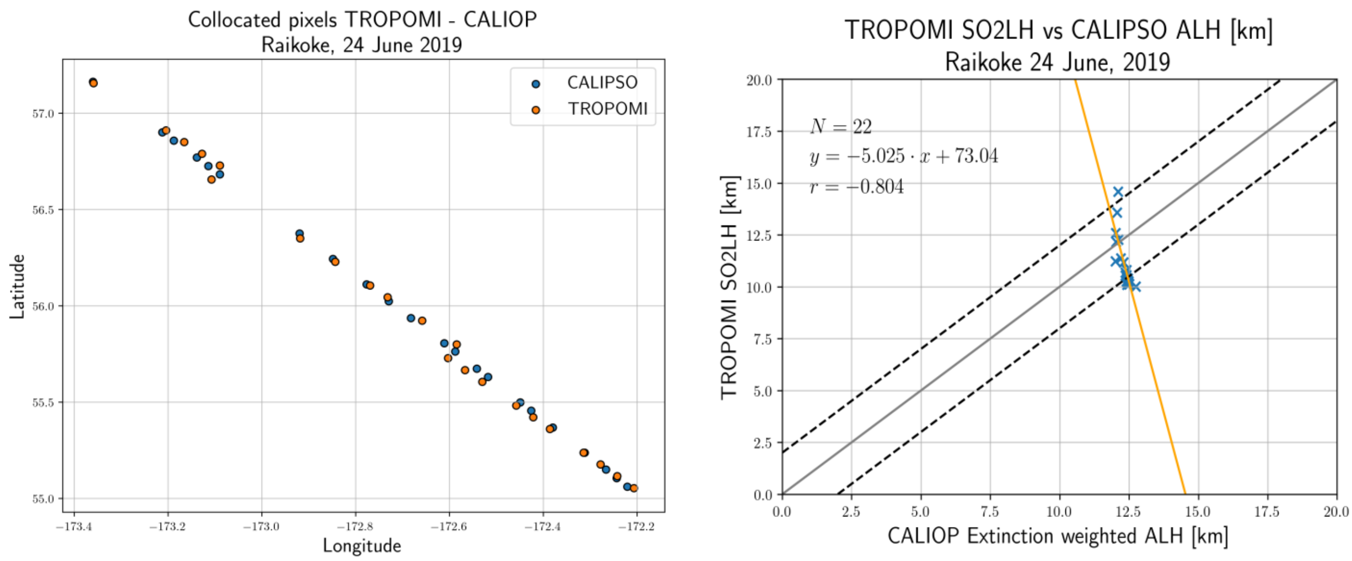
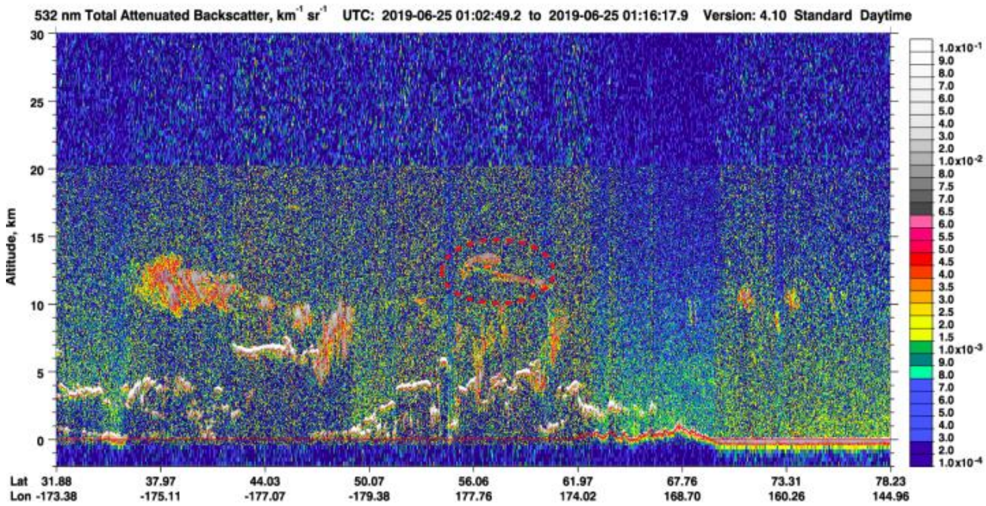
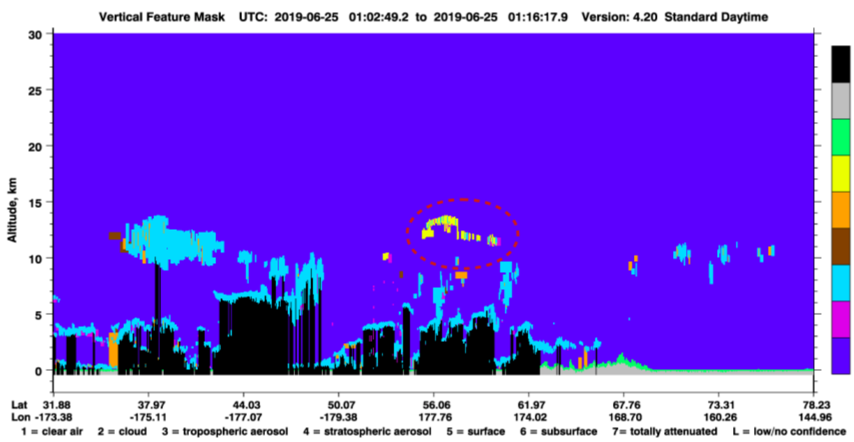


Figure 28. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 24 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line.*


532 nm Total Attenuated Backscatter, km-1 sr-1 UTC: 2019-06-25 01:02:49.2 to 2019-06-25 01:16:17.9 Version: 4.10 Standard Daytime
Altitude, km
Lat 31.88 37.97 44.03 50.07 56.06 61.97 67.76 73.31 78.23
Lon -173.38 -175.11 -177.07 -179.38 177.76 174.02 168.70 160.26 144.96


Vertical Feature Mask UTC: 2019-06-25 01:02:49.2 to 2019-06-25 01:16:17.9 Version: 4.20 Standard Daytime
Altitude, km
Lat 31.88 37.97 44.03 50.07 56.06 61.97 67.76 73.31 78.23
Lon -173.38 -175.11 -177.07 -179.38 177.76 174.02 168.70 160.26 144.96
1 = clear air 2 = cloud 3 = tropospheric aerosol 4 = stratospheric aerosol 5 = surface 6 = subsurface 7= totally attenuated L = low/no confidence

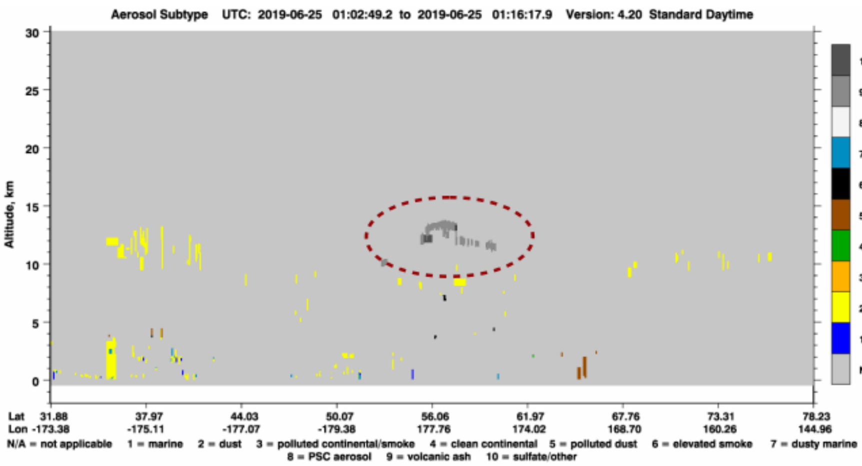

Aerosol Subtype UTC: 2019-06-25 01:02:49.2 to 2019-06-25 01:16:17.9 Version: 4.20 Standard Daytime
Altitude, km
Lat 31.88 37.97 44.03 50.07 56.06 61.97 67.76 73.31 78.23
Lon -173.38 -175.11 -177.07 -179.38 177.76 174.02 168.70 160.26 144.96
N/A = not applicable 1 = marine 2 = dust 3 = polluted continental/smoke 4 = clean continental 5 = polluted dust 6 = elevated smoke 7 = dusty marine
8 = PSC aerosol 9 = volcanic ash 10 = sulfate/other

Figure 29. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 25 June 2019, (middle) Vertical feature mask image showing the location of all layers detected and (bottom) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

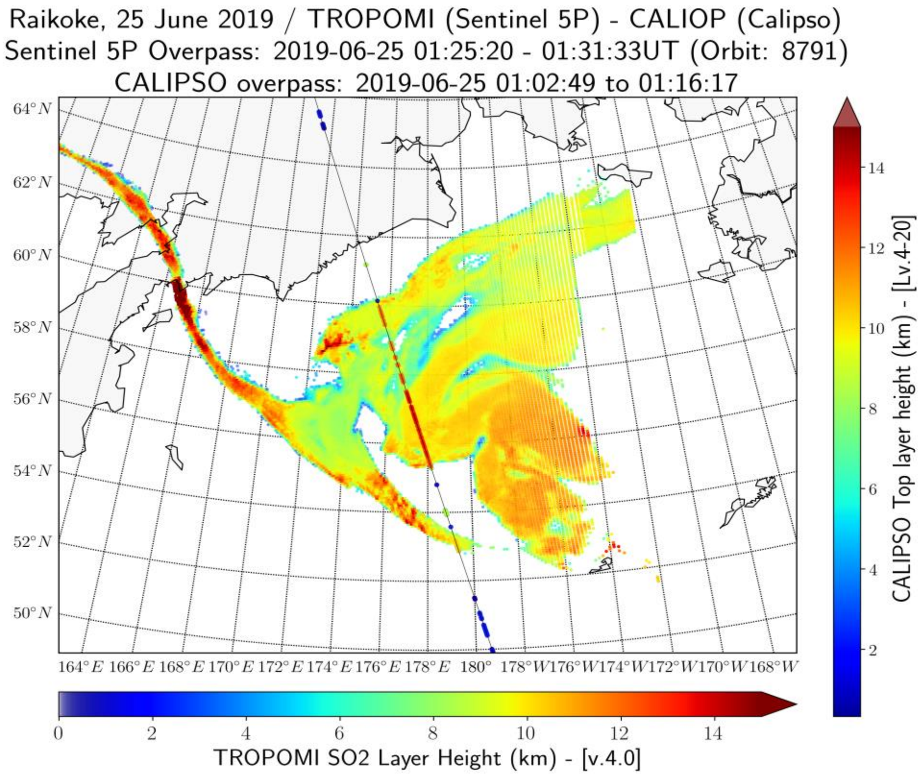


Figure 30. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 25 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicate the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height product values (in km), for the results shown in Figure 29.*

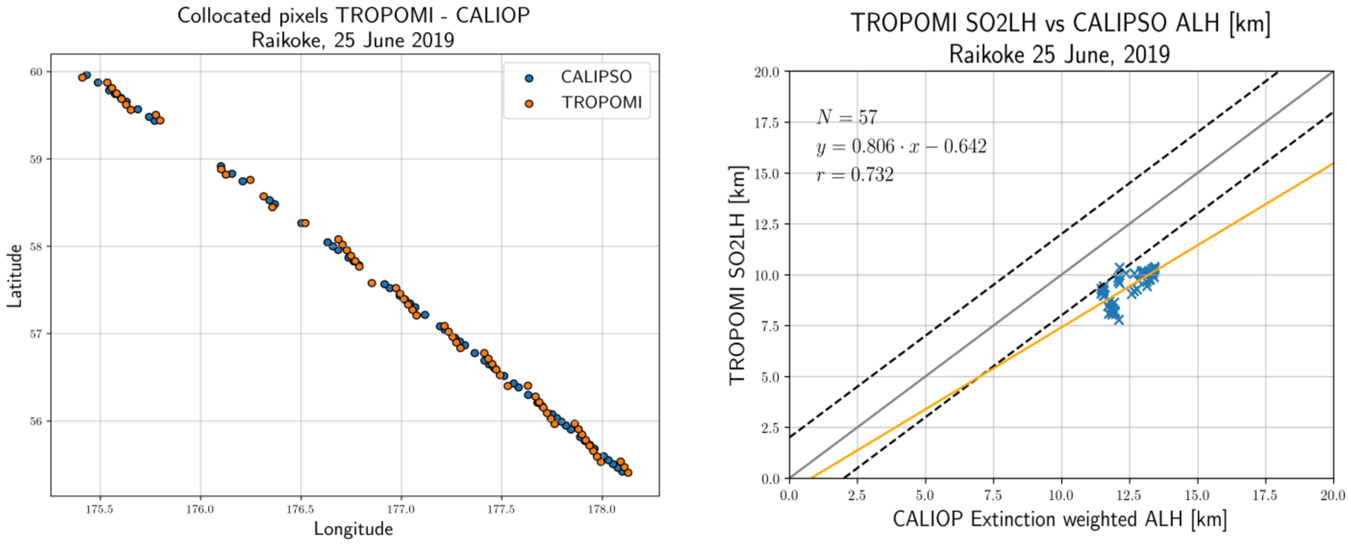
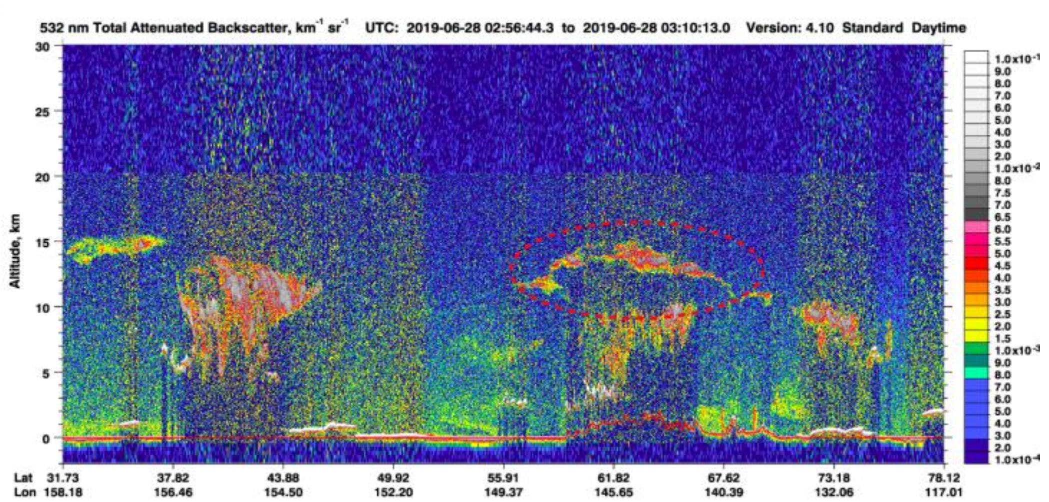


Figure 31. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO2 LH and CALIPSO weighted extinction height for the 25 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations; the grey line is the 1:1 line.*



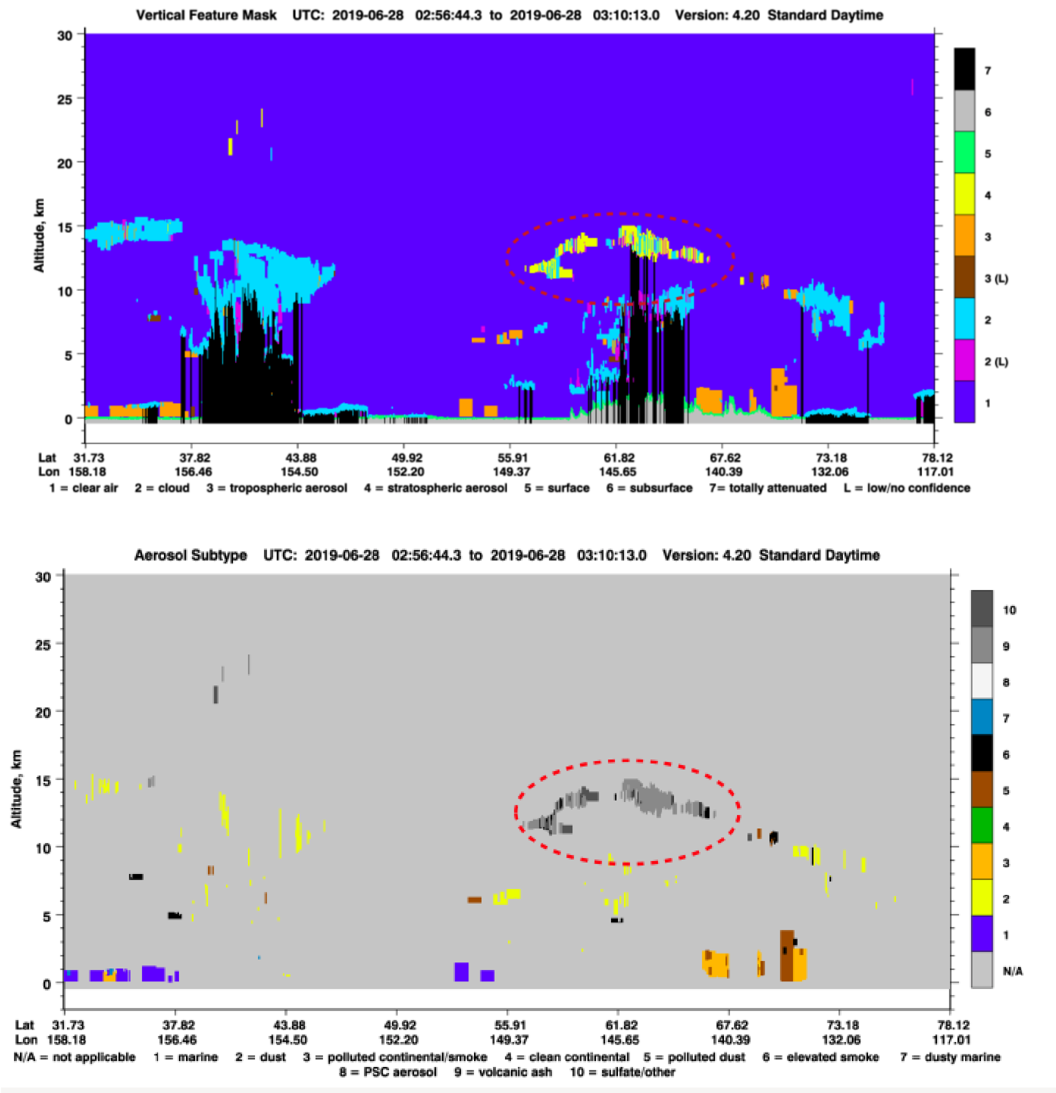


Figure 32. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 28 June 2019, (middle) Vertical feature mask image showing the location of all layers detected and (bottom) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

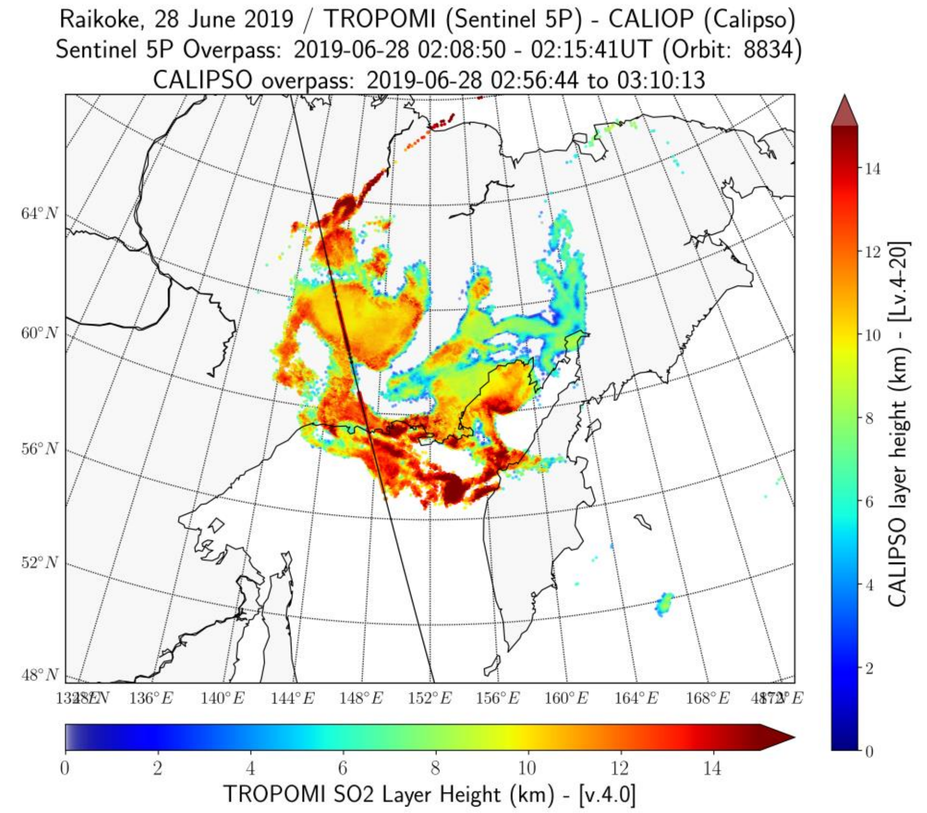
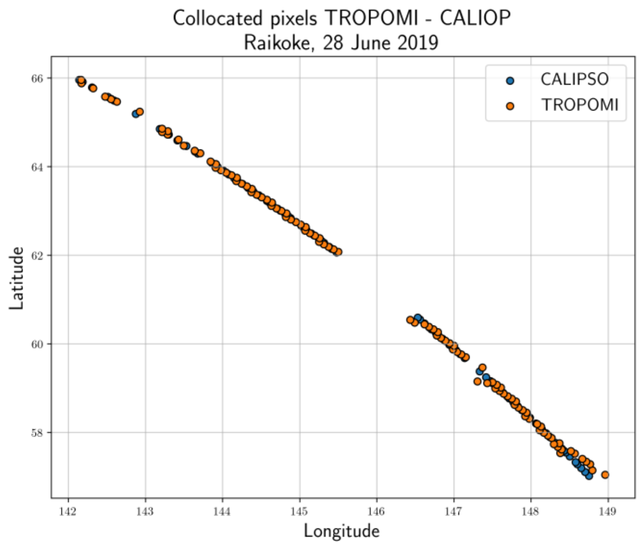


Figure 33. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 28 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicate the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height values (in km), for the results shown in Figure 32.*



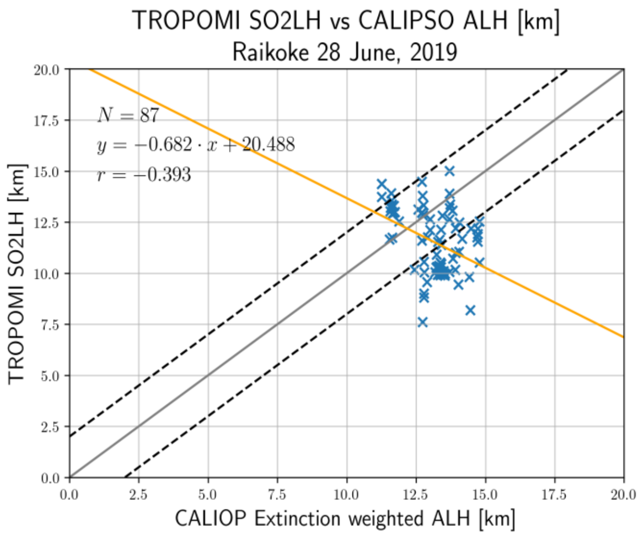
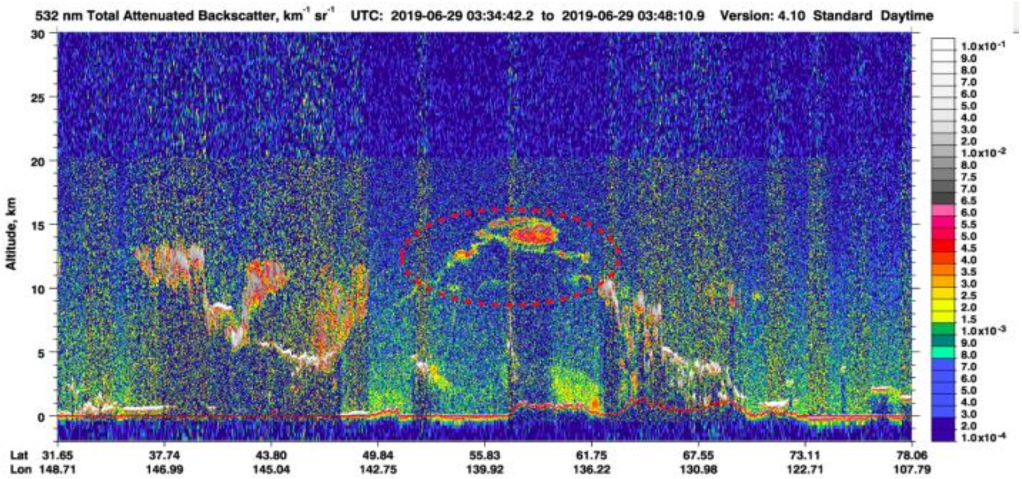
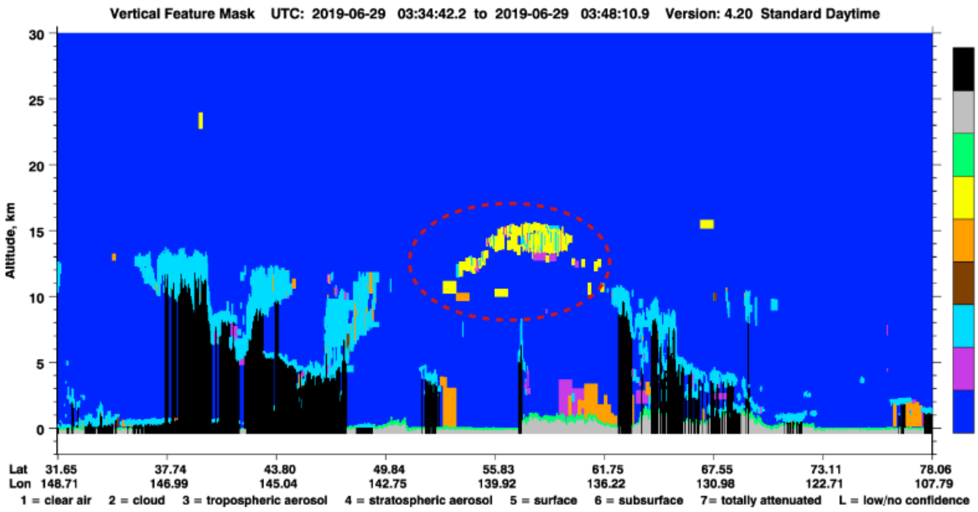


Figure 34. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 28 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line.*





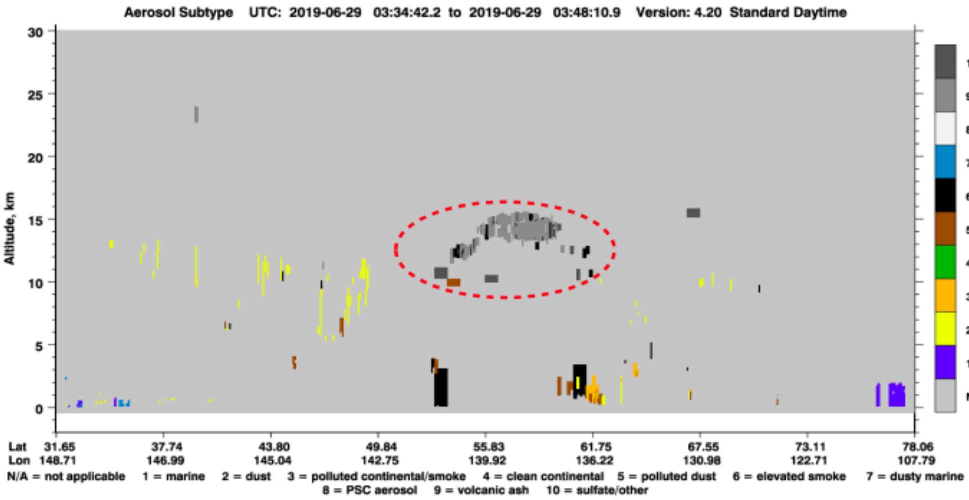


Figure 35. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 29 June 2019, (middle) Vertical feature mask image showing the location of all layers detected and (bottom) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

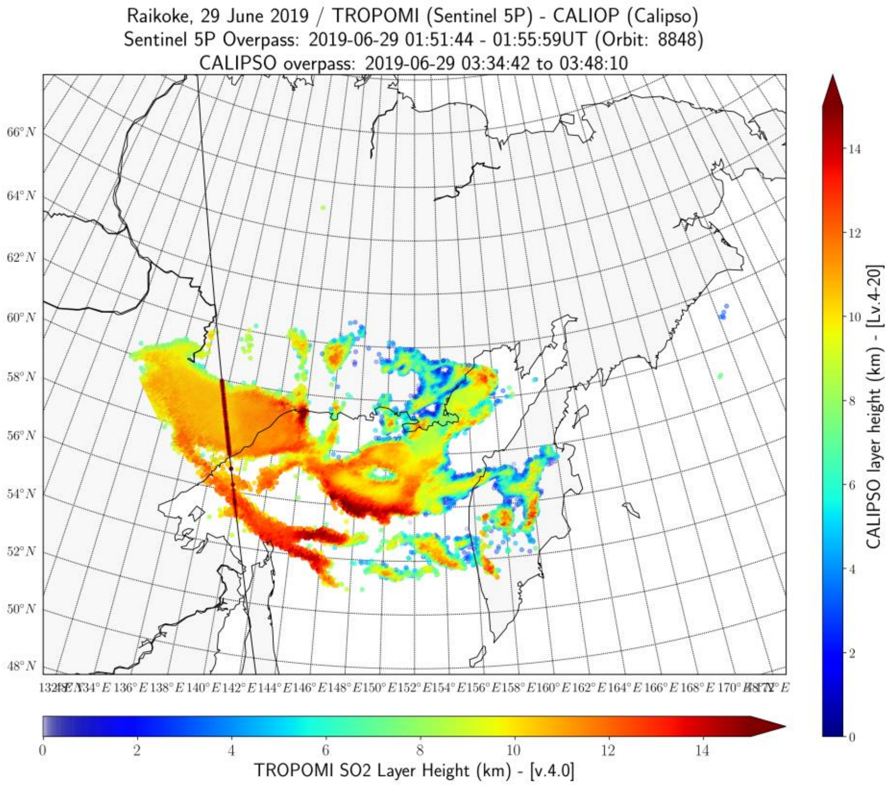


Figure 36. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 29 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicate the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height values (in km), for the results shown in Figure 35.*

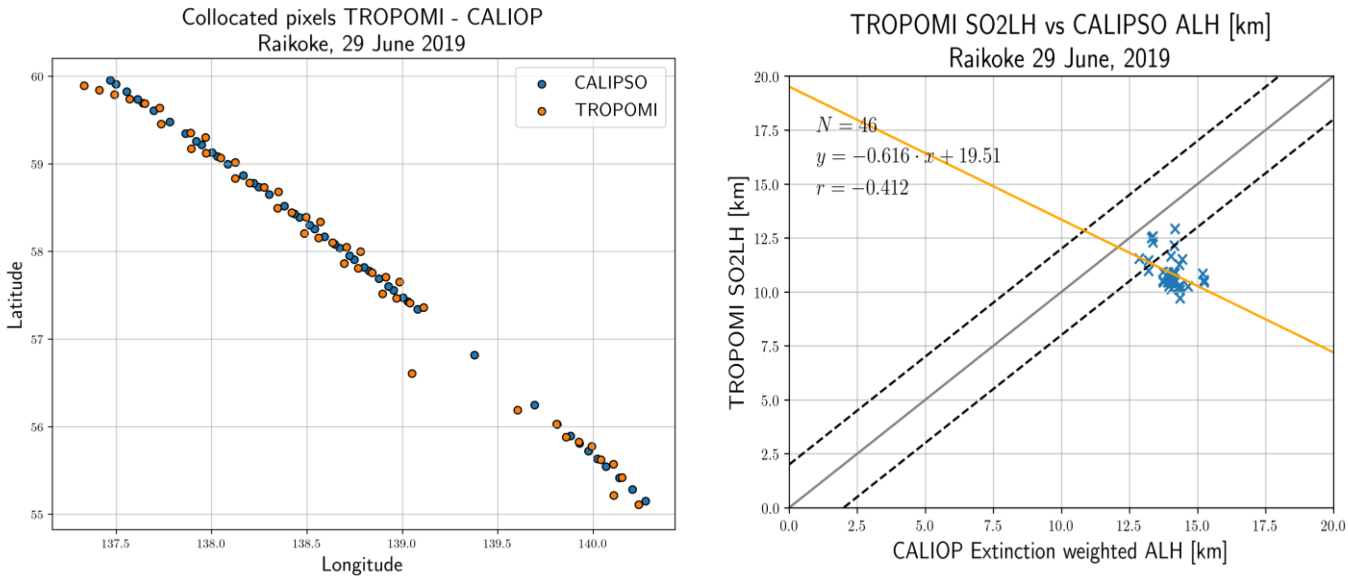
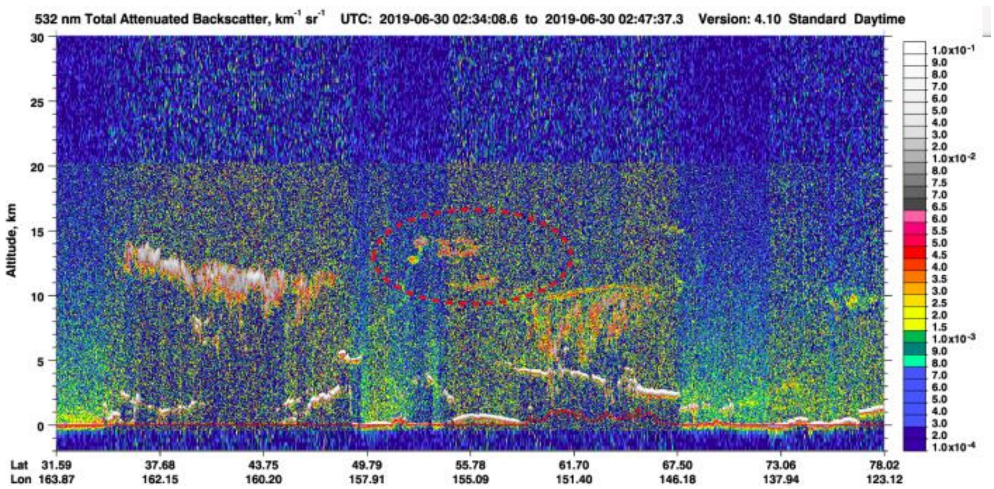


Figure 37. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI SO_2 LH and CALIPSO weighted extinction height for the 29 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line.*



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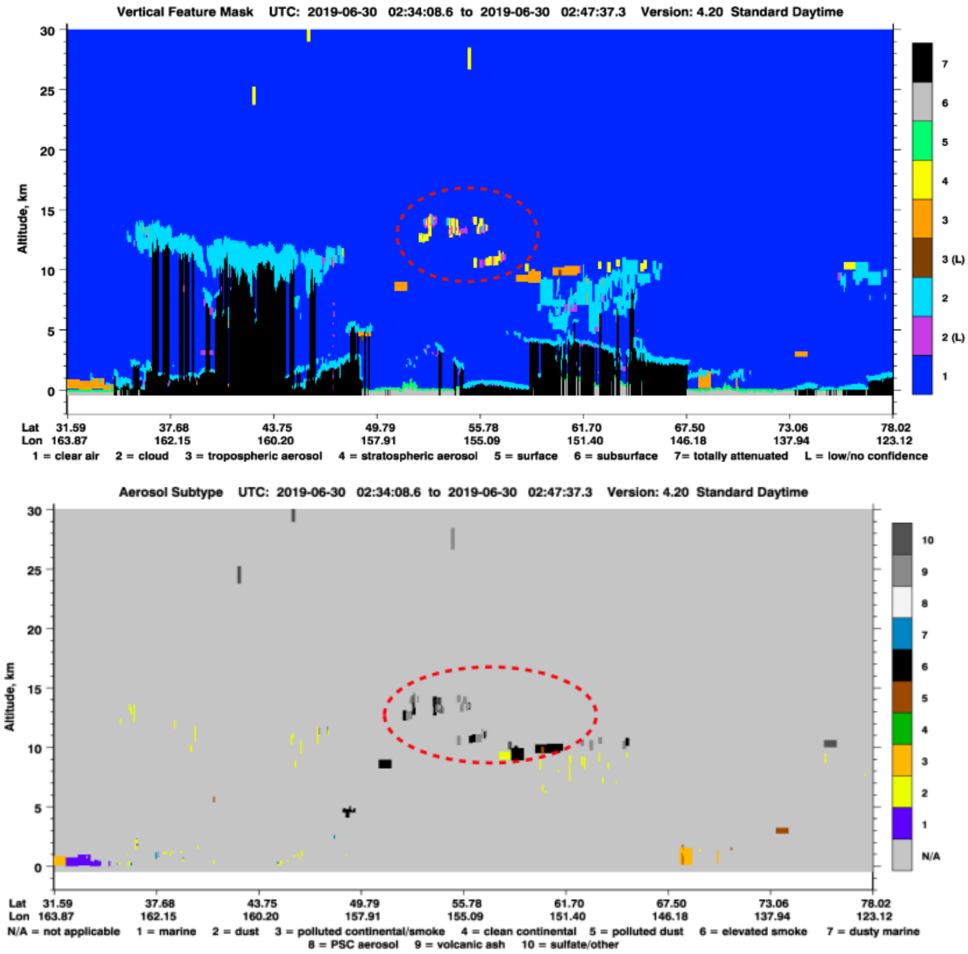

Figure 38. *(Top) CALIOP total attenuated backscatter profile for the Raikoke eruption on 30 June 2019, (middle) Vertical feature mask image showing the location of all layers detected and (bottom) aerosol subtype. The red dashed circles denote the volcanic feature detected from CALIOP. (***https://www-calipso.larc.nasa.gov/products/***)*

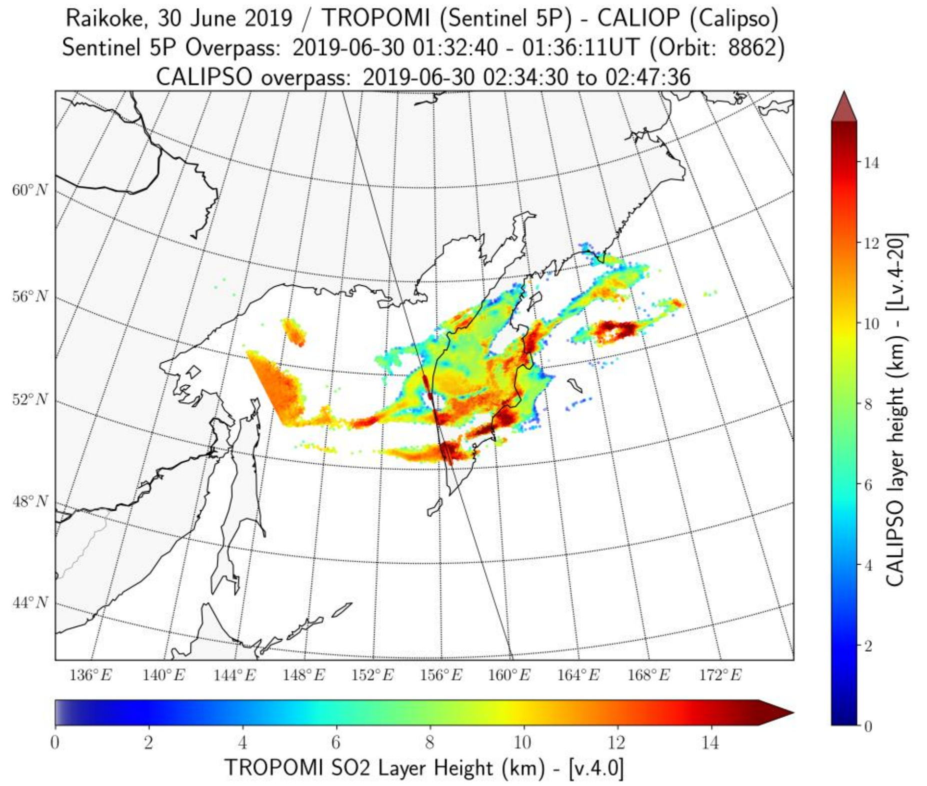

Figure 39. *TROPOMI SO_2 layer height for the Raikoke volcanic eruption, measured on the 30 June 2019. Only pixels with SO_2 VCDs greater than or equal to 20 DU are shown. The black line indicate the CALIPSO ground track and the coloured circles along the line indicate weighted extinction height values (in km), for the results shown in Figure 38.*

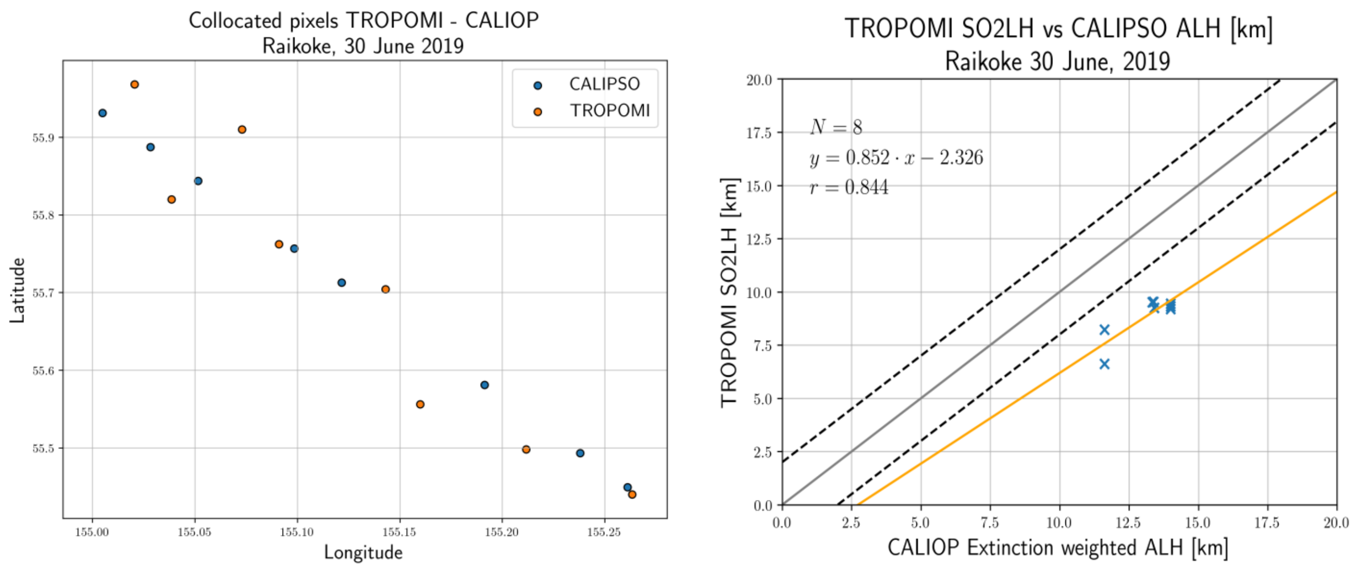

Figure 40. *Left. The latitude/longitudes of the collocated pixels. Right. Comparison between TROPOMI $S0_2$ LH and CALIPSO weighted extinction height for the 30 June 2019. The orange line is the regression line of the TROPOMI-CALIPSO observations, the grey line is the 1:1 line.*