



1 Improved retrieval of SO₂ plume height from TROPOMI 2 using an iterative Covariance-Based Retrieval Algorithm

3 Nicolas Theys¹, Christophe Lerot¹, Hugues Brenot¹, Jeroen van Gent¹, Isabelle De Smedt¹,
4 Lieven Clarisse², Mike Burton³, Matthew Varnam³, Michel Van Roozendael¹

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6 ¹ Royal Belgian Institute for Space Aeronomy (BIRA-IASB), Brussels, Belgium.

7 ² Université libre de Bruxelles (ULB), Spectroscopy, Quantum Chemistry and Atmospheric
8 Remote Sensing (SQUARES), C. P. 160/09, Brussels, Belgium.

9 ³ School of Earth and Environmental Sciences, University of Manchester, Oxford Road,
10 Manchester, M139PL, UK

11 *Correspondence to:* N. Theys (theys@aeronomie.be)

12

13 **ABSTRACT**

14 Knowledge of sulfur dioxide layer height (SO₂ LH) is important to understand volcanic eruption
15 processes, the climate impact of SO₂ emissions and to mitigate volcanic risk for civil aviation.
16 However, the estimation of SO₂ LH from ground-based instruments is challenging in particular for
17 rapidly evolving and sustained eruptions. Satellite wide-swath nadir observations have the
18 advantage to cover large-scale plumes and the potential to provide key information on SO₂ LH. In
19 the ultraviolet, SO₂ LH retrievals leverage the fact that, for large SO₂ columns, the light path and
20 its associated air mass factor (AMF) depends on the SO₂ absorption (and therefore on the vertical
21 distribution of SO₂), and SO₂ LH information can be obtained from the analysis of measured back-
22 scattered radiances coupled with radiative transfer simulations. However, existing algorithms are
23 mainly sensitive to SO₂ LH for SO₂ vertical columns of at least 20 DU. Here we develop a new
24 SO₂ LH algorithm and apply it to observations from the high spatial resolution TROPOspheric
25 Monitoring Instrument (TROPOMI). It is based on an SO₂ optical depth look-up-table and an
26 iterative approach. The strength of this scheme lies in the fact that it is a Covariance-Based
27 Retrieval Algorithm (COBRA; Theys et al., 2021). This means that the SO₂-free contribution of
28 the measured optical depth is treated in an optimal way, resulting in an improvement of the SO₂



1 LH sensitivity to SO₂ columns as low as 5 DU, with a precision better than 2km. We demonstrate
2 the value of this new data through a number of examples and comparison with satellite plume
3 height estimates (from IASI and CALIOP), and back trajectory analyses. The comparisons
4 indicates an SO₂ LH accuracy of 1-2 km, expect for some difficult observation conditions.

5 **1. INTRODUCTION**

6 Volcanic eruptions can emit large quantities of rock fragments and fine particles (ash) into the
7 atmosphere as well as several trace gases, such as carbon dioxide (CO₂), sulphur species (SO₂,
8 H₂S), halogens (HCl, HBr, HF), and water vapour. These volcanic ejecta can have a tremendous
9 impact on human health, society and nature, and on air traffic safety. In particular, emission of
10 sulphur dioxide (SO₂) receives considerable attention due to its subsequent conversion into
11 aerosols and potentially strong effect on global climate (Robock, 2000). Among the emitted
12 constituents, SO₂ is also the easiest to detect from ultraviolet (UV) and thermal infrared (TIR)
13 remote-sensing techniques, and is being used for many decades to monitor volcanoes worldwide.
14 In order to understand volcanic processes and assess the impact of eruptions, it is crucial to
15 measure not only the total abundance of SO₂ but also the height of the SO₂ plume. This information
16 is important for (1) aviation actors such as Volcanic Ash Advisory Centres (VAACs) in case ash
17 and SO₂ clouds are collocated; sulfur alone is also becoming increasingly recognized as causing
18 long-term damage to aircraft engines (mainly because of sulfuric acid), (2) volcanology as it
19 informs on eruption rate, eruption type and underlying volcanic processes (e.g., Mastin et al.,
20 2009), (3) atmospheric chemistry and climate research, e.g. to model the impact of volcanic
21 eruptions on air quality (Schmidt et al., 2015) or to study the partly understood role of modest
22 volcanic eruptions on climate forcing (Solomon et al., 2011; Vernier et al., 2011; Santer et al.,
23 2014), and (4) the estimation of SO₂ emissions, as the measured SO₂ abundances are often directly
24 dependent on the knowledge of the SO₂ vertical distribution.

25 Ground-based cameras can be used to routinely monitor plume heights (e.g., Scollo et al., 2014)
26 but these measurements are performed very near-field. For large and sustained volcanic eruptions,
27 estimation of plume heights is very difficult in practice – not to say impossible – and the available
28 measurements generally suffer from poor or infrequent sampling of the volcanic plumes.
29 Moreover, many volcanoes on the globe are not monitored. Consequently, satellite nadir sensors
30 with large swaths and frequent revisiting time offer the best solution to cover completely the



1 emitted volcanic cloud.
2 Space nadir sensors have provided global measurements of SO₂ vertical columns and masses for
3 more than 40 years (Carn et al., 2016, and references therein). However, the retrieval of SO₂ plume
4 height (also referred to SO₂ layer height, SO₂ LH) from satellite hyperspectral measurements is a
5 relatively recent development. In the TIR, global SO₂ LH retrievals from the Infrared Atmospheric
6 Sounding Interferometer (IASI) by Carboni et al. (2012) and Clarisse et al. (2014) and the Cross-
7 track Infrared Sounder (CrIS) by Hyman and Pavolonis (2020) proved to have an excellent
8 sensitivity to the SO₂ height above ~5 km, even for SO₂ columns at 1 DU level (Dobson unit – 1
9 DU: 2.69×10^{16} molecules cm⁻²). In the UV spectral range, the sensitivity to SO₂ is better at lower
10 altitudes and the first studies using full radiative transfer calculation schemes were from Yang et
11 al. (2010) and Nowlan et al. (2011), based on the Ozone Monitoring Instrument (OMI; Levelt et
12 al., 2006) and the Global Ozone Monitoring Experiment-2 (GOME-2; Munro et al., 2006). More
13 recently, new approaches based on Inverse Learning Machine schemes have become available for
14 GOME-2 (Efremenko et al., 2017), TROPOspheric Monitoring Instrument-TROPOMI (Hedelt et
15 al., 2019) and OMI (Fedkin et al., 2021). These algorithms greatly improve the computational
16 performance of the previously published techniques. However, all the UV schemes referenced
17 above have demonstrated sensitivity to SO₂ LH only for SO₂ vertical columns of more than ~ 20
18 DU, which limits their use to relatively large volcanic events. In this paper, we present a new UV
19 spectral fitting algorithm allowing to retrieve SO₂ LH for SO₂ columns as low as 5 DU, and for
20 SO₂ layer heights as low as 1km. This scheme is an extension of our recently published SO₂
21 Covariance-Based Retrieval Algorithm (COBRA; Theys et al., 2021) that enables drastic reduction
22 in spectral interferences and retrieval noise. Here we combine COBRA with an iterative look-up
23 table approach to treat the non-linear SO₂ contribution to the measured signal. This allows joint
24 retrieval of the SO₂ vertical column density (VCD) and SO₂ layer height with improved sensitivity
25 while avoiding time-consuming on-line radiative transfer simulations. We apply this technique to
26 measurements from TROPOMI (Veefkind et al., 2012) aboard the Sentinel-5 Precursor (S-5P)
27 satellite. The motivation is obviously its high spatial resolution of 3.5×5.5 km². TROPOMI
28 resolves locally enhanced SO₂ columns much better than predecessor instruments like OMI (Theys
29 et al., 2019). The retrieval of SO₂ LH is therefore expected to be possible for several degassing
30 volcanoes. This has the potential to enhance our capability of monitoring height-resolved volcanic
31 plumes globally in the troposphere. In addition, for strong eruptions, retrieved SO₂ LH (and SO₂



1 vertical columns) at high spatial resolution can also provide unique insights into volcanic
2 processes, atmosphere-plume interactions and transport (Pardini et al., 2018, references).

3 The paper is structured as follows. Section 2 describes the algorithm in detail and demonstrates
4 the performance of the SO₂ LH retrieval. In Section 3, the results are evaluated against other
5 satellite data sets and dispersion model results. Conclusions and perspectives are given in Section
6 4.

7

8 **2. ALGORITHM DESCRIPTION**

9 The theoretical basis for a joint retrieval of SO₂ column amount and layer altitude from satellite
10 nadir back-scattered UV measurements is described by Yang et al. (2010) and Nowlan et al.
11 (2011). The TROPOMI SO₂ layer height algorithm, outlined in this section, is an iterative retrieval
12 scheme. It is conceptually close to these pioneering algorithm studies in the way the SO₂ absorption
13 is handled but differs in the treatment of the other contributions to the measured signal.

14 We first define the measured top-of-atmosphere total optical depth (OD) by:

$$15 \quad y_{meas} = y_{SO_2} + y_{bckg} + \varepsilon \quad (1)$$

16 All terms of the equation depend on wavelength (not labelled here, for simplicity). $y_{meas} =$
17 $-\log(I/I_0)$ is the logarithmic ratio of the wavelength calibrated measured radiance (I) and
18 irradiance (I_0) over a given wavelength range, y_{SO_2} is the unknown SO₂ optical depth, y_{bckg} is the
19 “background” optical depth, accounting for all contributions to the total OD except that of SO₂,
20 and ε is the measurement error.

21 In case of strong SO₂ absorption, the optical depth y_{SO_2} is fundamentally a non-linear function of
22 the VCD and LH of SO₂, and solving Eq. (1) is non-trivial. However, we assume here that the
23 expression can be linearized using a Taylor expansion:

$$24 \quad y_{meas} - y_{SO_2,i} \approx \Delta VCD \frac{\partial y_{SO_2,i}}{\partial VCD} + \Delta LH \frac{\partial y_{SO_2,i}}{\partial LH} + y_{bckg} + \varepsilon \quad (2)$$

25 $y_{SO_2,i}$ is the SO₂ OD at the linearization point $y_{SO_2,i} = y_{SO_2}(VCD_i, LH_i)$, $\partial y_{SO_2,i}/\partial VCD$ and
26 $\partial y_{SO_2,i}/\partial LH$ are the corresponding partial derivatives with respect to the SO₂ VCD and LH



1 (Jacobians), and ΔVCD and ΔLH are the VCD and LH increments. Index i stands for the i^{th}
2 iteration.

3 To solve Eq. (2), we developed a hybrid method. To model the SO_2 signal, the algorithm makes
4 use of a large look-up-table (LUT) of SO_2 OD. At each iteration, improved estimations of VCD
5 and LH become available. These results are used to update the SO_2 OD and Jacobians for the next
6 calculation, until convergence is reached. This part of the algorithm will be detailed in section 2.1.
7 To treat the background and error terms of Eq. (2), we propose a COBRA method. In brief, instead
8 of fitting the background optical depth, the algorithm considers a representative set of measured
9 spectra uncontaminated by SO_2 , and characterized by a mean optical depth \bar{y} and a covariance
10 matrix S , to represent statistically $y_{bckg} + \varepsilon$. The idea of the method, initially presented by Walker
11 et al. (2011) and further developed in other studies (e.g., Carboni et al., 2012; Clarisse et al., 2014;
12 Theys et al., 2021), is to consider $y_{bckg} + \varepsilon$ as an error term, and to interpret S as a generalized
13 error covariance matrix. Thus, the solution of the inverse problem can be expressed (Rodgers,
14 2000):

$$15 \quad \hat{x}_{i+1} = \hat{x}_i + (k_i^T S^{-1} k_i)^{-1} k_i^T S^{-1} (y_{meas} - y_{SO_2,i} - \bar{y}) \quad (3)$$

16 where \hat{x}_i is the retrieved state vector ($[LH_i, VCD_i]^T$) and k_i is the SO_2 forward model
17 ($[\partial y_{SO_2,i}/\partial LH \quad \partial y_{SO_2,i}/\partial VCD]$).

18 It should be emphasized that the matrix S accounts (if well-constructed) for most atmospheric
19 background and instrumental-related variability of the spectra, including cross-correlations. The
20 strength of the algorithm lies in the fact that only two parameters are retrieved (SO_2 LH and VCD).
21 As will be shown in sections 2.3 and 3.1, this constitutes a significant advance in terms of retrieval
22 sensitivity compared to a classical fitting approach such as the Differential Optical Absorption
23 Spectroscopy (DOAS; Platt and Stutz, 2002), where multiple parameters are fitted in addition to
24 SO_2 LH and VCD.

25 The following two sections describe in more detail the SO_2 LUT approach and the specific
26 algorithm settings.

27



1 2.1 SO₂ optical depth look-up-table: description

2 Forward modeled SO₂ spectra are based on the LInearized Discrete Ordinate Radiative Transfer
3 (LIDORT) model version RRS 2.2 (Spurr et al., 2008). The input data used to set the atmosphere
4 and spectroscopy are detailed in Table A1. Simulations were carried out to cover a large range of
5 possible measurement conditions, using different combinations of LUT entries for the observation
6 geometry, total ozone column, Lambertian Equivalent Reflectivity (LER) and SO₂ vertical profiles
7 (Table 1). More details are given below. The simulation results are the SO₂ slant optical depth
8 spectra over a wavelength range from 309 to 329 nm and at a spectral sampling of 0.05 nm. Note
9 that the spectroscopic input data (absorption cross-sections and solar spectrum) were not pre-
10 convolved with the Instrumental Spectral Response Function (ISRF) of TROPOMI but rather with
11 a box-car function of 0.05 nm width. Therefore, the simulations are not instrument-specific. For
12 application to TROPOMI, the SO₂ OD spectra were convolved with the ISRF, and a specific
13 correction for the so-called solar-I₀ effect (Aliwell et al., 2002) was applied, as it turned out to be
14 important for large SO₂ VCDs (100-1000 DU). It should be noted that the TROPOMI ISRF
15 parameters vary smoothly with the position across-track (450 in total). For practical ease of use,
16 we assumed that the across-track dependence is well encapsulated by the viewing zenith angle
17 (VZA) entry. To represent the full swath, we used the sign convention of negative/positive VZA
18 for W/E, respectively. For each of the VZA grid point (Table 1), a slit function of the TROPOMI
19 detector column was associated with the closest mean VZA. This appears to be a good
20 approximation for TROPOMI and avoids having 450 different LUTs.

21 From the LUT of SO₂ OD spectra, the algorithm extracts a sub-LUT for a given TROPOMI
22 measurement by linear interpolation. To do so, the observation angles at the ground pixel location
23 are used. Input on total ozone is obtained from the TROPOMI off-line total ozone column product
24 (Garane et al., 2019). The latter is well suited for the present application, as it is weakly affected
25 by spectral interferences with SO₂ (bias of only few % in case of strong eruptions, see discussion
26 in Lerot et al., 2014). In addition to the observation geometry and total ozone absorption, the
27 measurement sensitivity to SO₂ is also strongly dependent on the surface reflectance and the
28 presence of clouds or aerosols layer. Here we assume that the radiative transfer in the atmosphere
29 can be sufficiently represented through a lower bound Lambertian Equivalent Reflector. The LER
30 approach works very well in principle when the SO₂ plume is above a cloud or aerosol layer.



1 However, it has limited applicability for cases where SO₂ and aerosols are mixed, especially for
 2 highly absorbing aerosols such as volcanic ash. This aspect will be further discussed in Section 3.
 3 The LER is characterized by effective parameters, height and albedo, that are determined for each
 4 pixel. The LER height is computed as the cloud-fraction weighted mean of the cloud and ground
 5 altitudes. Cloud parameters are from the operational cloud product OCRA/ROCINN CRB (Loyola
 6 et al., 2018). For the LER albedo, it is constrained by TROPOMI measured radiance averaged over
 7 339.5-340.5 nm, a range mostly unaffected by trace gas absorption (O₃ and SO₂). The LER albedo
 8 is retrieved by matching the measured mean radiance to a LUT of radiances (generated in parallel
 9 to the SO₂ OD LUT), and which depends on SZA, VZA, RAA, surface height and albedo, with
 10 the same grid definition as in Table 1. The simulated radiances are convolved and averaged over
 11 the same wavelength range as TROPOMI.

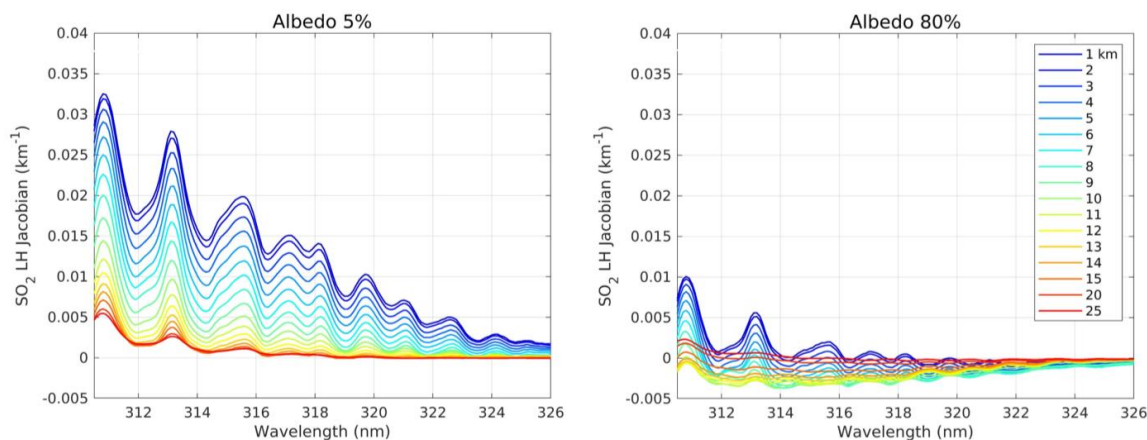
12 Table 1: Physical parameters that define the SO₂ optical depth look-up-table. The total number of
 13 spectra is about 38.5×10^6 . In practice, note that LUT interpolation is performed along the cosine
 14 of SZA and VZA.

Parameter	Grid values	Number of grid points
Solar zenith angle (SZA)	10, 20, 30, 40, 50, 60, 70 (°)	7
Viewing zenith angle (VZA)	-70, -60, -50, -40, -30, -20, -10, 0, 10, 20, 30, 40, 50, 60, 70 (°)	15
Relative azimuth angle (RAA)	0, 45, 90, 135, 180 (°)	5
Total O₃ column	145, 175, 205, 235, 295, 355, 415, 475, 535 (DU)	9
Albedo	0, 5, 10, 20, 40, 60, 80, 100 (%)	8
Surface height	0, 1, 2, 4, 6, 9 (km)	6
SO₂ column	1, 2, 5, 10, 25, 50, 100, 200, 500, 1000 (DU)	10
SO₂ height	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 20, 25 (km)	17
Wavelength	309-329 nm (0.1 nm step, after convolution)	201



1 From the interpolation step, a sub-LUT of SO₂ OD spectra is obtained, which depends only on
2 SO₂ VCD and LH. From this table, the SO₂ VCD and LH Jacobians are derived by simple discrete
3 derivatives. These functions are essential for the retrieval (Eq. 3). Example SO₂ LH Jacobians are
4 presented in Figure 1, for a fixed SO₂ VCD of 25 DU and representative LER albedos of 5% (left)
5 and 80% (right), representative of typical clear-sky and fully cloudy conditions, respectively. For
6 this example, we observe the largest sensitivity to SO₂ LH for low albedo and low SO₂ peak height.
7 This behavior is expected as most of the altitude information comes from the way SO₂ alters the
8 availability of photons to be scattered by air molecules below the SO₂ layer (Yang et al., 2010).
9 Conversely, for high SO₂ height or high albedo (e.g., for an underlying cloud), the scattering
10 weighting functions are weakly dependent on the altitude, and the information on SO₂ LH appears
11 to be less accessible. The performance of the algorithm under various conditions will be discussed
12 further in Section 2.3.

13



14 Figure 1: Examples of SO₂ optical depth Jacobians with respect to LH for different SO₂ peak
15 heights (1-25 km), and LER albedos of 5% (left) and 80% (right). The spectra correspond to SZA:
16 30°, VZA: 0°, RAA: 0°, SO₂ column: 25 DU, ozone column: 295 DU, surface height: 0 km.

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18 2.2 LUT-COBRA settings

19 The retrieval of SO₂ vertical column and height is performed from the analysis of measured
20 radiances in the spectral range from 310.5 to 326 nm (TROPOMI band 3). The algorithm starts
21 from an a priori pair (x_0) of SO₂ VCD and LH. The initial value VCD₀ is taken as the output of the



1 operational TROPOMI SO₂ column product for a plume height of 7 km (Theys et al., 2017). The
2 height z_0 is 7 km, except if the LER height is greater than 5km. In that case, z_0 is equal to the LER
3 height + 2km. First, the SO₂ optical depth and Jacobians spectra for x_0 are derived from the LUT
4 (as described in section 2.1), and interpolated on the wavelength grid of the measurement. Then
5 the results of the fit (Eq. 3) are used to calculate new SO₂ spectra for the next calculation and the
6 retrieval is repeated until the inverted LH and VCD do not change from one iteration to the next
7 by more than 500 m and 10% respectively, or if the number of iterations exceeds a limit value
8 (fixed to 10). Note that for some iterations, the algorithm gives SO₂ LH occasionally outside the
9 SO₂ height grid. For those cases, the SO₂ height is forced to the grid minimum height +1 km or
10 the grid maximum height -1km, depending if the height is below or above the grid
11 minimum/maximum height respectively. More rarely, the same can happen for the retrieved SO₂
12 VCD and then the SO₂ VCD is set to VCD_0 for the next iteration.

13 A key information in the retrieval process is the covariance matrix S (and mean optical depth \bar{y}),
14 as it directly influences the sensitivity of the retrieval (Eq. 3). For the construction of S and \bar{y} we
15 used a set of measured SO₂-free spectra, following an approach analogous to our previous study
16 (Theys et al., 2021). In brief, for each TROPOMI observation for which the SO₂ LH algorithm is
17 applied (see next section), we consider the spectral data of the corresponding orbit and TROPOMI
18 row. To represent best the zonal dependence, we select the radiance spectra of 300 pixels along
19 the flight direction (i.e., ± 150 indices along track), and the pixels with observable SO₂ amounts
20 (with $VCD > 2.5 \times VCD$ retrieval error) are filtered out. To keep a viable number of spectra for the
21 covariance calculation (at least 100), we also allow the number of pixels along the flight direction
22 to increase, if necessary. Note that an upper limit on the SZA is fixed to 65° in order to exclude
23 difficult conditions with high ozone absorption.

24 It should be noted that the quality of the LUT-COBRA results depends strongly on the signal of
25 SO₂. This aspect is addressed in the next section.

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1 **2.3 Performance of the retrievals**

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3 Following Rodgers (2000), the estimated error covariance of the solution (Eq. 3) is given by:

$$4 \quad \hat{S}_i = (k_i^T S^{-1} k_i)^{-1} \quad (4)$$

5 This matrix can be calculated for each TROPOMI pixel and the square root of the diagonal
6 elements of \hat{S}_i provide error estimates on the retrieved SO₂ LH and VCD.

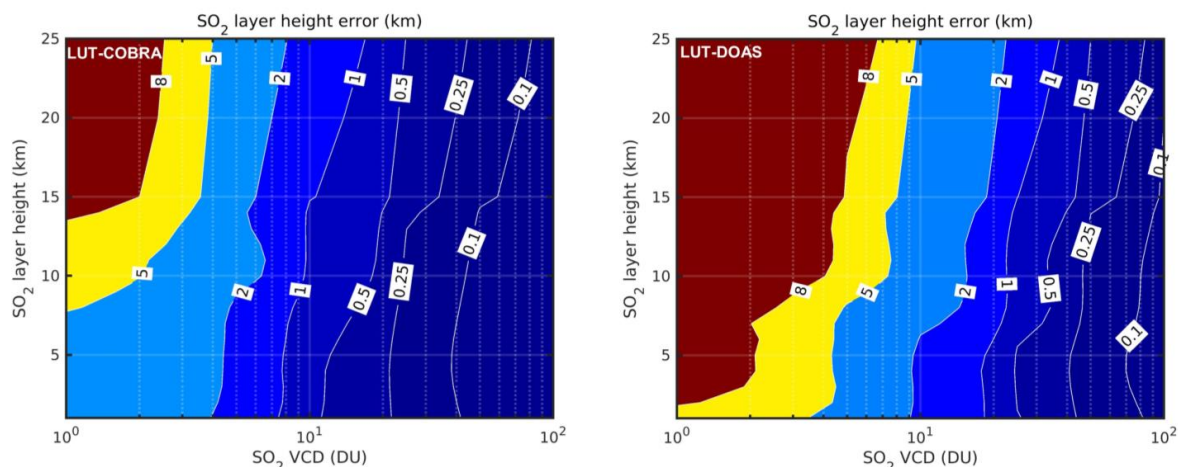
7 To demonstrate the sensitivity of the algorithm for different SO₂ altitudes and vertical columns,
8 the SO₂ LH error was computed for predefined pairs of Jacobians, and a fixed covariance matrix
9 S . The results are summarized in Figure 2 (left) for typical observation conditions in the tropics,
10 over a dark surface. From this example, it is clear the SO₂ LH retrieval uncertainty decreases for
11 high SO₂ columns. This is obvious as the SO₂ signal dominates all variability contributions. The
12 results also suggest that the algorithm performs better for SO₂ at low heights. This behavior is
13 logical, and is in line with the dependence of the SO₂ LH Jacobians with the SO₂ height (see Fig.
14 1 left, and related discussion). It is interesting to note that the observed retrieval performance
15 dependence with SO₂ height is complementary to the one found for thermal infrared nadir
16 sounders, like IASI or CrIS. Indeed, Carboni et al. (2012) and Clarisse et al. (2014) demonstrated
17 (using IASI) that the best SO₂ height retrieval is achieved for SO₂ plumes in the upper
18 troposphere/lower stratosphere (UTLS) while in the lower troposphere (below 3-5 km), the
19 sensitivity to the SO₂ height is strongly reduced, as a result of water vapor absorption. This
20 complementarity is further addressed in Section 3.

21 To compare the performance of the LUT-COBRA with more classical fitting approaches, we have
22 also developed a modified DOAS algorithm, referred as LUT-DOAS in the following. In short,
23 the forward model matrix was expanded to include not only the LH and VCD Jacobians (grouped
24 as k_i) but also other spectra, used to fit the measured OD. Essentially, we implemented a linearized
25 version of the DOAS scheme used in the operational TROPOMI SO₂ algorithm (Theys et al.,
26 2017). More precisely, 13 spectral functions are used to model the ozone absorption, Ring effect,
27 broadband component (in the form of a 3rd order polynomial), spectral shift and squeeze (Beirle et
28 al., 2013), and linear intensity offset. Based on this DOAS-type forward model matrix, the SO₂
29 layer height error for the LUT-DOAS scheme was calculated using Eq. 4, by replacing the
30 covariance matrix S with an identity matrix, divided by the square of the signal-to-noise ratio



1 (SNR). The latter was fixed to 800, a typical SNR of TROPOMI radiances over the fitting window
2 considered. The results are presented in Figure 2 (right).

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5 Figure 2: SO₂ layer height theoretical uncertainty for (left) LUT-COBRA and (right) LUT-DOAS
6 schemes. The results correspond to SZA: 30°, VZA: 0°, RAA: 0°, ozone column: 295 DU, surface
7 height: 0 km, surface albedo: 5%.

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9 This example indicates that, in theory, the LUT-DOAS scheme yields reasonable height retrievals
10 with uncertainty of 1-2 km for SO₂ columns greater than 10-40 DU, depending on the SO₂ height.
11 This finding is mostly consistent with previous studies on UV retrievals of SO₂ plume height (e.g.,
12 Nowlan et al., 2011; Hedelt et al., 2019).

13 However, compared to the LUT-DOAS algorithm, Figure 2 suggests that our LUT-COBRA
14 scheme significantly improves the SO₂ layer height error by a factor of 2-3. This is an appealing
15 aspect of the LUT-COBRA approach, as it enables the application of the SO₂ LH retrievals to SO₂
16 columns as low as 5 DU.

17 The performance results of Figure 2 were repeated for other observation conditions. In particular,
18 a high reflectance scenario (80%) was tested to represent the situation of an SO₂ plume lying above
19 a cloud deck. The results show no significant change in the performance of the retrievals indicating
20 that underlying clouds have little impact on the sensitivity to SO₂ LH. In a way, this is counter-
21 intuitive when looking at Figure 1, but one should keep in mind that the algorithm retrieves both



1 LH and VCD of SO₂. Clearly, high reflectance conditions help better constrain the SO₂ vertical
2 column (especially for low SO₂ heights) which in turn is beneficial to access the spectral
3 information on SO₂ LH. Another interesting result is to test the sensitivity of the retrieval as a
4 function of the observation angles. For instance, increasing the SZA first leads to improved results,
5 because the SO₂ signal increases as the optical path through the SO₂ layer gets longer, but then the
6 performance quickly deteriorates for high angles due to large absorption by ozone. Those
7 conditions are, however, discarded by the algorithm SZA cutoff of 65° (section 2.2).

8 It should be emphasized that the SO₂ layer height error presented here does not account for
9 systematic uncertainties. It is clear that in many circumstances, forward model errors can actually
10 dominate the total error on SO₂ LH. These errors are generally difficult to evaluate and depend on
11 the prevailing conditions. In Section 3, examples of TROPOMI results will be presented with
12 specific attention to possible sources of error. We also refer to Yang et al. (2010) and Nowlan et
13 al. (2011) for a presentation of the various sources of systematic uncertainties.

14 In practice, the SO₂ layer height error (Eq. 4) can be computed for each TROPOMI pixel. This is
15 useful as it helps diagnose the retrieval quality. In what follows, the retrievals are considered only
16 for a SO₂ layer height error lower than 2.5 km and retrieved VCD of at least 5 DU. In order to
17 preselect the spectra that potentially fulfill these criteria (and limit the computational effort), the
18 TROPOMI operational SO₂ product was examined. Only measurements with slant column
19 densities (a quantity independent of SO₂ plume height) larger than 2.5 DU were selected and
20 processed by the SO₂ LH algorithm.

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1 3. RESULTS

2 In this section, we present TROPOMI SO₂ LH data and evaluate the results against independent
3 plume height estimates from satellites and back-trajectory modelling.

4

5 3.1 Comparison with satellite plume height estimates

6 For a selection of eruption events, we performed comparisons with the IASI SO₂ height data of
7 Clarisse et al. (2014), readily available in near real-time in the Support to Aviation Control Service
8 (SACS; Brenot et al., 2014, 2021). Another useful dataset to validate the TROPOMI SO₂ height is
9 from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument onboard the
10 Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO). Here we used the
11 532 nm total backscatter coefficient profiles from the standard CALIOP level-2 v4 product
12 (CAL_LID_L2_05kmAPro-Standard-V4), available from NASA ([https://www-
13 calipso.larc.nasa.gov](https://www-calipso.larc.nasa.gov)). Although those datasets are very useful, we note that these can only be used
14 to validate TROPOMI SO₂ LH in a rather qualitative way because (1) IASI has a different overpass
15 time than TROPOMI, (2) CALIOP has a narrow swath (resulting in limited sampling of what S-
16 5P observes), and measures aerosols rather than SO₂. More validation results are shown in sections
17 3.2 and 3.3.

18 The first example is for the Sierra Negra volcano (0.83°S, 91.17°W, Ecuador) that erupted on 26
19 June 2018 at ~20:00 UTC, according to the Global Volcanism Program (volcano.si.edu).
20 Coincidentally, TROPOMI passed over the region shortly after the start of the eruption at ~20:12
21 UTC, and detected a freshly emitted and nearly undispersed SO₂ plume with heights of 3-5 km, in
22 good agreement with the S-5P FP_ILM results of Hedelt et al. (2019). On 27 June, the TROPOMI
23 overpass (orbit 03652, approximate time 19:50 UTC) revealed an SO₂ plume distributed in
24 multiple layers. Figure 3a shows the results of the LUT-COBRA. The retrieved SO₂ heights are as
25 low as 1-2 km near the vent and up to 18 km further downwind. The characteristic pattern of SO₂
26 height levels observed for the different parts of the plume is consistent with the retrievals of CrIS
27 (Hyman and Pavolonis, 2020). Figure 3 also presents results from IASI on 28 June at ~15:15 UTC
28 (SO₂ height images for other dates and acquisition times are accessible on the SACS webpage;
29 <http://sacs.aeronomie.be>). As can be seen, the TROPOMI LUT-COBRA and IASI SO₂ LH results
30 agree qualitatively well considering the relatively large time difference of nearly 20 hours between
31 the two sensors. A notable difference though is for the SO₂ plume located below 3 km, which is

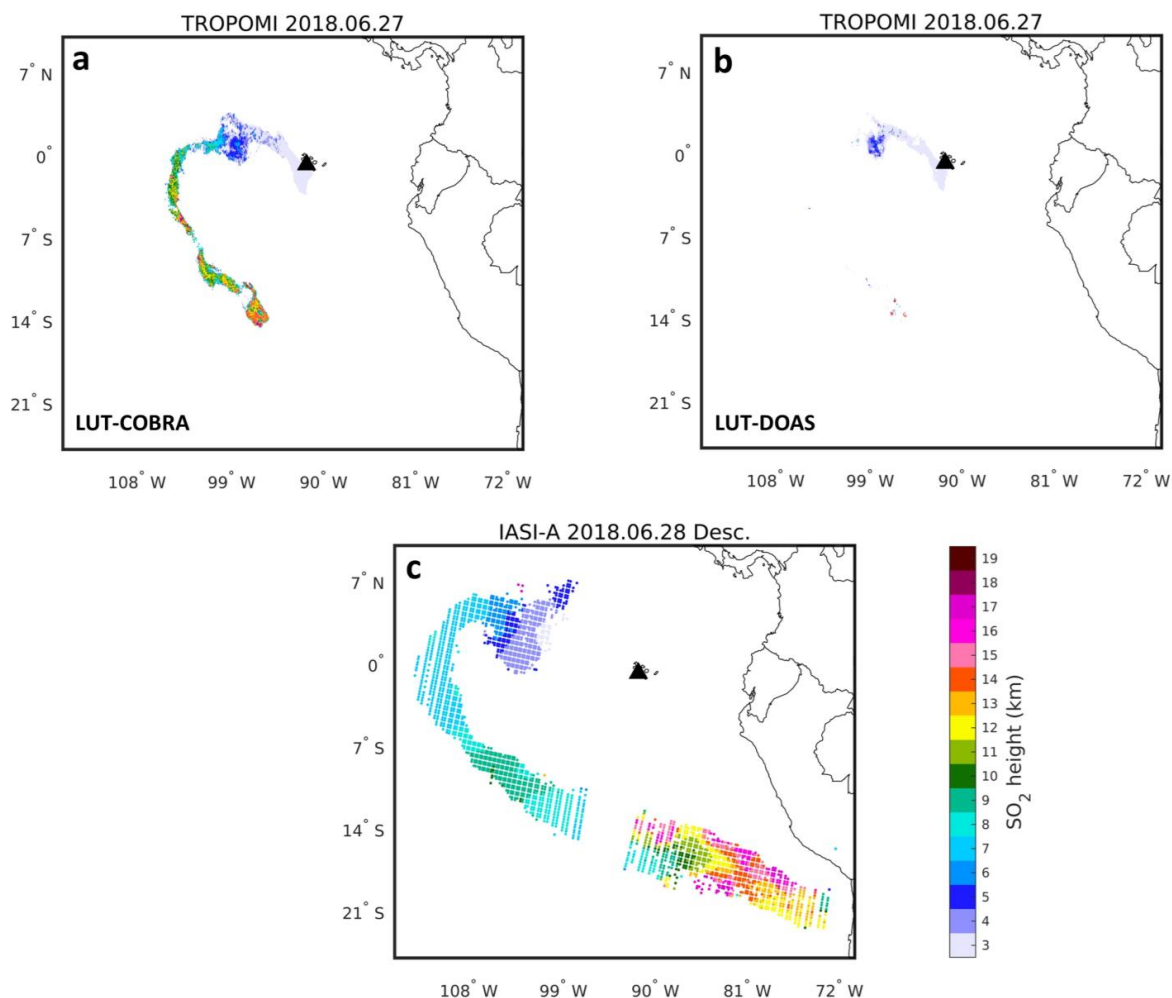


1 barely seen in the IASI data. For this particular example, this is partly because the volcano lies
2 between two orbits but inspection of other SO₂ images does not reveal significant SO₂ detections
3 below 3 km by IASI. The reason is likely the limited sensitivity of IASI in the lowermost
4 troposphere, particularly in the tropics.

5 In addition to the LUT-COBRA results, Figure 3b also presents the corresponding retrievals from
6 the LUT-DOAS implementation (introduced in Section 2.3). Here we show the results for retrieved
7 SO₂ VCDs greater than 20 DU in order to keep the SO₂ height data with retrieval errors better than
8 1- 2 km (Figure 2, right panel). This threshold is the same as in Hedelt et al. (2019). Overall, the
9 SO₂ LHs from LUT-DOAS are in close agreement with the LUT-COBRA results, for the pixels
10 in common. The LUT-DOAS values also match very well the results of S-5P FP_ILM, Fig. 10b
11 of Hedelt et al. (2019). However, this example of Figure 3 clearly demonstrates that the LUT-
12 COBRA is able to retrieve SO₂ LH for many more pixels with greater sensitivity than the LUT-
13 DOAS approach. In the following, only the LUT-COBRA results will be presented and discussed.
14 A second illustration of the LUT-COBRA SO₂ LH results is for the Raikoke volcano (48.29°N,
15 153.25°E, Kuril Islands, Russia) that erupted on 21 June 2019 with multiple explosions that started
16 at ~18:00 UTC and lasted several hours. The eruption emitted enormous amounts of SO₂ in the
17 atmosphere, around 1.5 Tg (e.g., de Leeuw et al., 2021), as well as volcanic ash. Raikoke is
18 therefore a good case to test the SO₂ LH algorithm under extreme conditions. Note that the eruption
19 of Raikoke is also well documented and is the subject of an Atmos. Chem. Phys./Atmos. Meas.
20 Tech./Geosci. Model Dev. special issue.

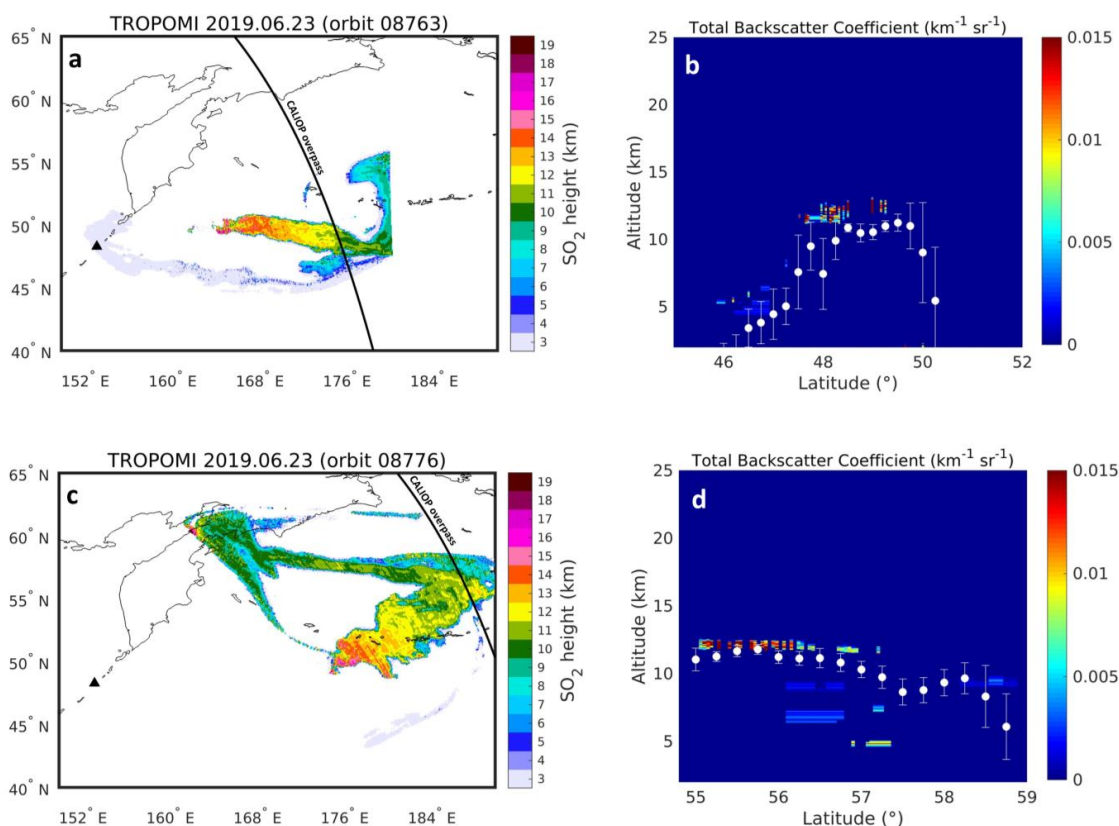
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1
2 Figure 3: SO₂ layer height results for the eruption of Sierra Negra from (a) TROPOMI LUT-
3 COBRA, (b) TROPOMI LUT-DOAS on 27 June 2018, and (c) IASI/MetOp-A on 28 June 2018
4 (descending orbit). The Sierra Negra volcano is marked by a black triangle.

5 Figure 4 a,c present two examples of SO₂ LH results for Raikoke on the 23 June 2019. Most of the
6 SO₂ is found between 5 and 15 km, in agreement with Cai et al. (2021). The SO₂ distribution as a
7 function of height seen in this example is also observed by other satellite data sets. The core of the
8 plume is located in the 8-14 km altitude range similar as Hyman and Pavolonis (2020) and Hedelt
9 et al. (2019), and is consistent with SO₂ profiles from the Microwave Limb Sounder (MLS).



1

2 Figure 4: SO₂ layer height results from TROPOMI observations of the Raikoke plume on 23 June
3 2019, for orbits (a) 08763 and (c) 08776. The retrieved maximum SO₂ VCD is 613 DU and 274
4 DU, respectively. The Raikoke volcano is marked by a black triangle. The black lines indicate the
5 CALIOP overpasses within 1 hour. (b,d) Comparison between CALIOP plume detection from
6 measured 532 nm total backscatter coefficient and TROPOMI SO₂ LH results, for (a,c)
7 respectively. The TROPOMI values (in white) are the mean and standard deviation of the SO₂ LH
8 results within 0.25° × 0.25° boxes, coincident with CALIOP.

9

10 Figure 4a,c also features a plume at much lower altitude, which is consistently observed in other
11 studies (e.g., Hyman and Pavolonis, 2020, de Leeuw et al., 2021, Muser et al., 2020). The
12 TROPOMI results of Figure 4 also agree reasonably well with IASI SO₂ heights. However, the
13 comparison is left out of this study, as it will be covered in a future publication (Vernier et al., in
14 preparation). Instead, we compare the TROPOMI SO₂ LH with nearly coincident CALIOP



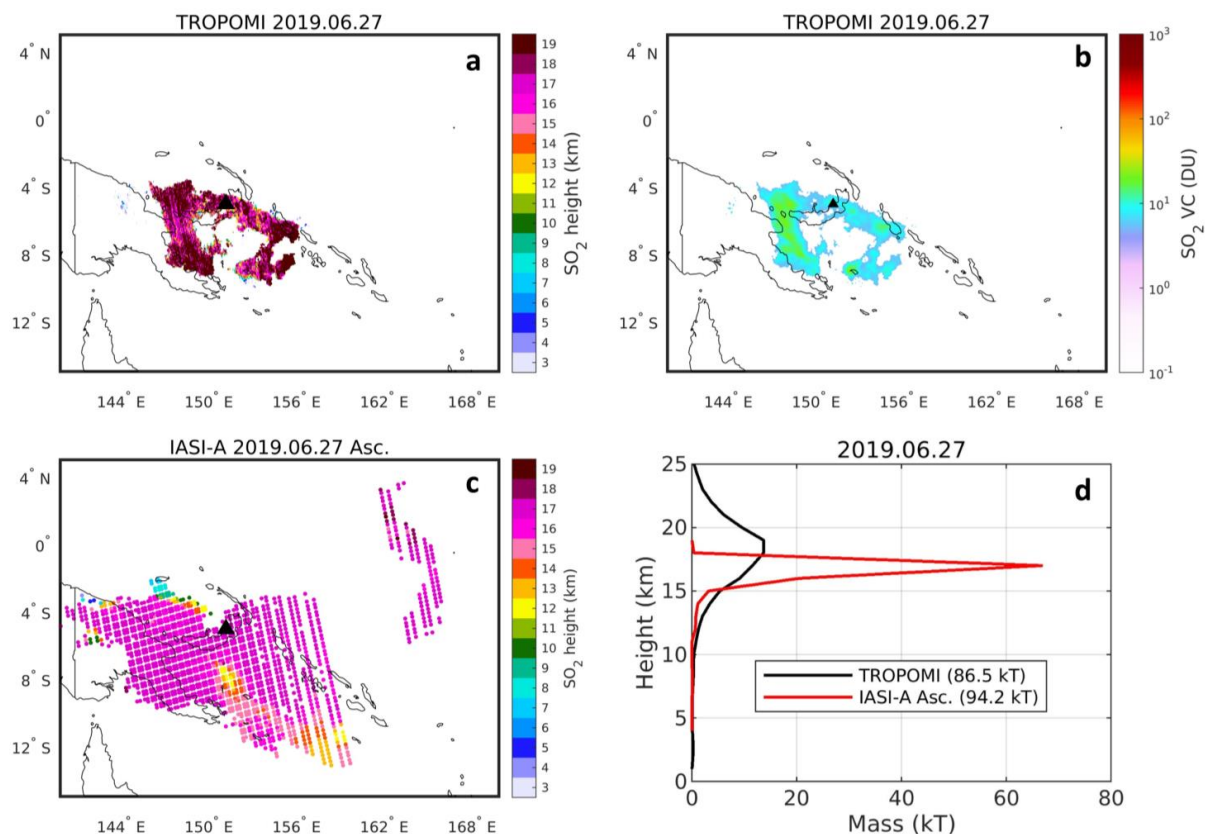
1 observations of the Raikoke plume. Figure 4b,d show the comparison between the measured total
2 backscattered coefficient of CALIOP and collocated TROPOMI SO₂ LH, for the two orbits of Fig.
3 4a and c. Qualitatively, the TROPOMI SO₂ LH is in agreement with the aerosol features detected
4 by CALIOP. For instance, the plume at lower altitude (~46-48°N in Fig. 4b), is well captured by
5 TROPOMI. However, it is clear that overall the retrieved SO₂ heights are systematically lower
6 than CALIOP by 1-3 km. This finding is in line with the results of Koukouli et al. (2021) who
7 found similar low bias of the S-5P FP_ILM SO₂ height product compared to CALIOP. Note also
8 that the first TROPOMI observations of the Raikoke plume were made on 22 June 2019 (orbit
9 08749). For that plume, the algorithm retrieves SO₂ heights of ~8 km (not shown). Unfortunately,
10 there was no CALIOP measurement available on that day, but it appears that these heights are
11 much too low when compared to other data. This is not a problem specific to our algorithm, and it
12 highlights the difficulty to retrieve the SO₂ height in the UV for a scene with a mixture of SO₂ and
13 ash (e.g. Yang et al., 2010; Hedelt et al., 2019). Under these conditions, the LUT and LER
14 approximation fail to reproduce adequately the complex radiative transfer in the volcanic plume,
15 leading to a low bias on the SO₂ height, which can be as high as 5 km for fresh and thick ash
16 plumes.

17 A last test case is for the Ulawun volcano (5.05°S, 151.33°E, Papua New Guinea) that erupted
18 explosively on 26 June 2019 around 04:30 UTC and injected SO₂ at the tropical tropopause level
19 in the form of a well-defined umbrella cloud. On 27 June, TROPOMI (orbit 08821, approximate
20 time 04:00 UTC) observed a SO₂ plume over the region of Ulawun (Figs. 5a,b) with SO₂ LH
21 distributed mainly between 15 and 21 km (Fig. 5d), with a center-of-mass height of 17.7 km.
22 Conversely, the IASI/MetOp-A overpass at ~11:20 UTC on the same day revealed a plume of SO₂
23 injected in a narrower vertical layer, with center-of-mass height of 16.6 km, hence slightly lower
24 than the TROPOMI estimate. It is interesting to note that the total SO₂ mass inferred from
25 TROPOMI is in rather good agreement with the IASI estimate, within 10%. We argue that the
26 difference in the SO₂ mass distributions could actually relate to the limited sensitivity of the
27 TROPOMI retrievals for that plume. Indeed, the SO₂ columns from TROPOMI (Fig. 5b) are
28 modest, smaller than 20 DU for most pixels, and 9 DU on average. At this VCD level, the retrieval
29 error on TROPOMI SO₂ LH (Fig. 2, left panel) is significant for a SO₂ plume in the UTLS, around
30 1.5 km. This is compatible with the observed spread of the TROPOMI SO₂ mass distribution (Fig.
31 5d). Regarding the apparent ~1 km difference between TROPOMI and IASI SO₂ center-of-mass



1 heights, we can of course not completely rule out a systematic error on the IASI retrievals but it is
2 unlikely to explain fully the observed offset. Importantly, for these conditions (low to medium
3 VCD, high LH), the TROPOMI SO₂ height retrieval is exposed to forward model errors that could
4 easily explain a 1 km bias. For example, the LUT uses a simplified representation of the
5 atmosphere in terms of temperature and ozone profiles that can ultimately lead to systematic errors
6 in the Jacobians used for the retrievals.

7 Overall, the examples presented in this section show that there is a general good agreement
8 between TROPOMI SO₂ LH and other satellite heights estimates. Nevertheless, the results also
9 highlight limitations of the retrievals in some (difficult) conditions. More work would be needed
10 to improve these results.



11
12 Figure 5: SO₂ plume on 27 June 2018 after the eruption of Ulawun. (a,b) TROPOMI retrievals of
13 SO₂ LH and VCD, respectively, (c) SO₂ LH from IASI/MetOp-A (ascending orbit), (d)



1 comparison of SO₂ mass histograms between TROPOMI and IASI. Total masses are indicated in
2 the legend. The Ulawun volcano is marked by a black triangle.

3

4 **3.2 Comparison with back trajectory analysis from PlumeTraj: the Etna case**

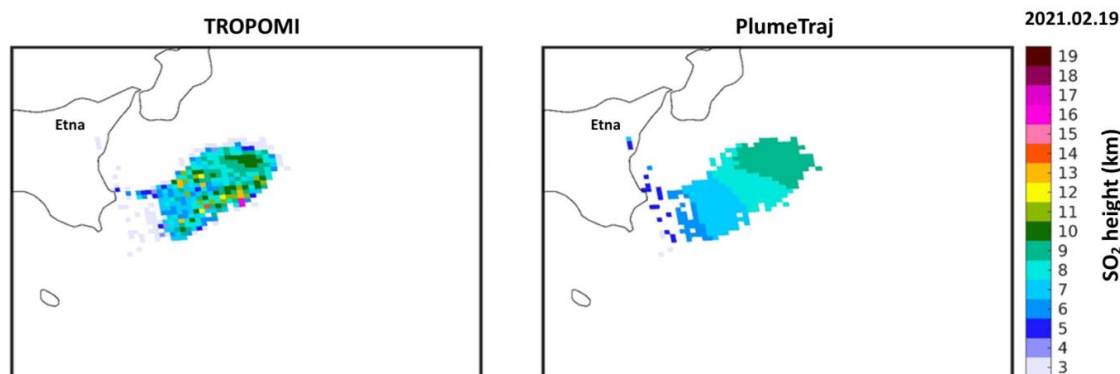
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6 An independent validation of the SO₂ height retrievals of TROPOMI can be obtained from back
7 trajectory analysis. Recently, a general algorithm has been developed called PlumeTraj (Pardini et
8 al., 2017, 2018; Queißer et al, 2019), which allows the height and age of volcanic SO₂ emissions
9 to be quantified for each TROPOMI pixel in a SO₂ image, using HYSPLIT back trajectories (Stein
10 et al., 2015). PlumeTraj leverages the fact that, because of the wind shear in the atmosphere, only
11 a limited range of back-trajectory altitudes connects a SO₂ pixel location with a given volcanic
12 vent. Importantly, when all pixels containing a volcanic plume are considered together, the height
13 and age parameters inferred by this method can be used in combination with the SO₂ column data
14 to reconstruct height- and time-resolved SO₂ emissions. This approach proves to be very powerful,
15 as it provides unique insights into the volcanic processes driving eruptions (Burton et al., 2021).

16 Here we have analyzed and compared the height results of TROPOMI SO₂ LH and PlumeTraj for
17 17 paroxysmal events of Mount Etna, Italy (37.75°N, 15°E), occurring in 2021.

18 Figure 6 presents an example of comparison, for a plume on February 19, 2021. It should be
19 stressed that the SO₂ plume heights, as shown in Fig. 6, are retrieved independently from each
20 other, as PlumeTraj only needs as input the observation time and pixel coordinates. For this event,
21 PlumeTraj derives SO₂ heights typically between 5 and 9 km with the highest values for the eastern
22 part of the plume and lower heights on the western part, and near the vent. Overall, this pattern is
23 well reproduced by our TROPOMI SO₂ LH retrievals - despite a few outliers. For the core of the
24 plume, the agreement between TROPOMI SO₂ LH and PlumeTraj is generally very good, with
25 differences mainly within ± 2 km. However, much larger differences are found for the edges of
26 the plume, where the TROPOMI SO₂ LH algorithm often retrieves SO₂ plume heights much lower
27 than PlumeTraj. We attribute this feature to an effect of the strong SO₂ horizontal inhomogeneity
28 within a TROPOMI pixel, which ultimately causes an underestimation of the retrieved SO₂ height
29 (consistent with Yang et al., 2010). This effect is also visible in Figure 4.

30

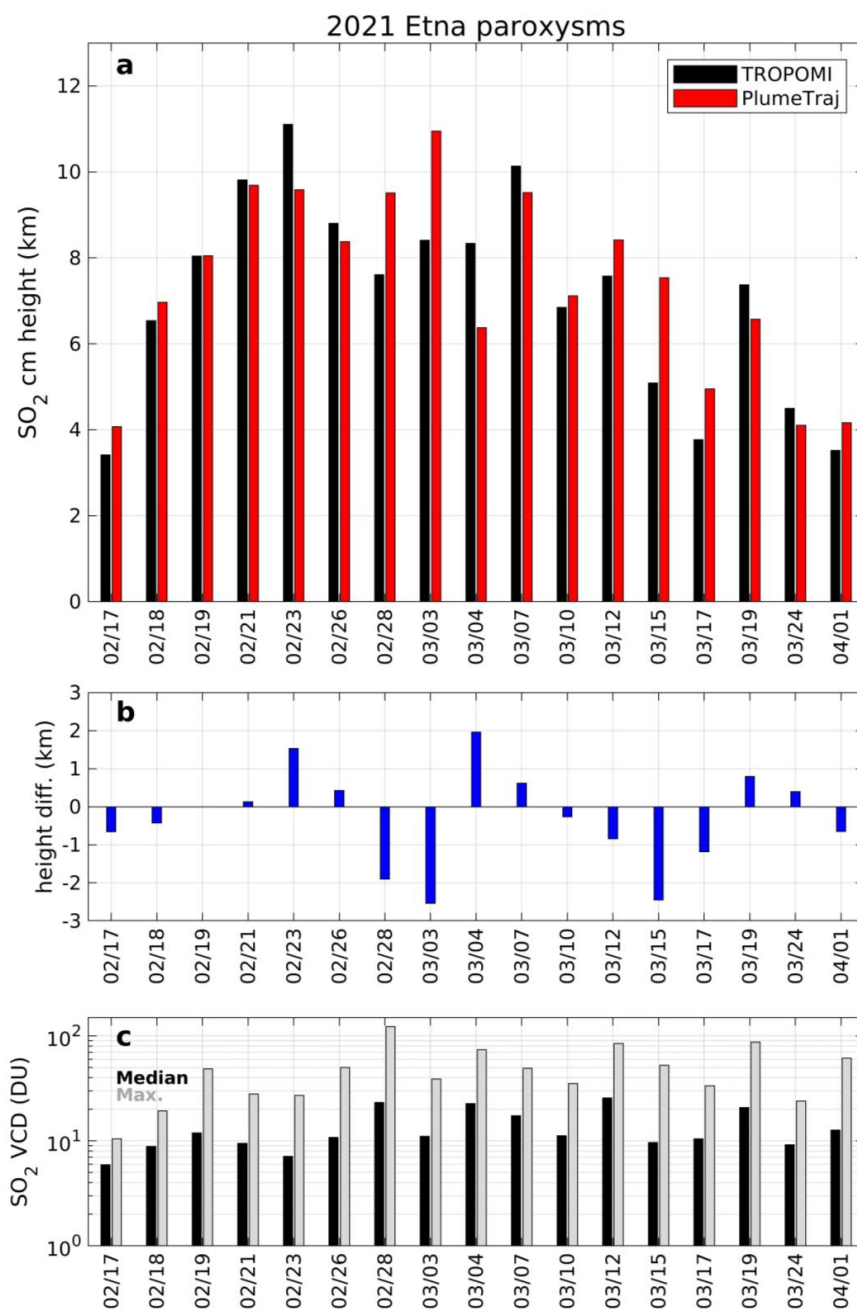


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2 Figure 6: Example of SO₂ height results derived from TROPOMI and PlumeTraj for an Etna plume
3 on 19 February 2021.

4

5 Figure 7a summarizes the TROPOMI and PlumeTraj SO₂ height results, for all selected Etna
6 events. For each SO₂ plume, the center of mass height was calculated by averaging the SO₂ height
7 weighted by the SO₂ column amount in each pixel. For TROPOMI SO₂ LH, this is performed
8 using the retrieved SO₂ column data, while for PlumeTraj the SO₂ column is estimated using linear
9 interpolation of the TROPOMI operational SO₂ column product (given at 1, 7 and 15km; Theys et
10 al., 2017) to the altitude returned from the trajectory analysis. Note that all pixels with retrieved
11 SO₂ heights < 1 km were excluded from the analysis, in an effort to reduce the impact of the pixels
12 near the plume edges affected by pixel under-filling. As can be seen from Fig. 7a, TROPOMI and
13 PlumeTraj capture comparable SO₂ heights (in the range of ~ 4-11 km) and similar variability. The
14 estimated total SO₂ masses are also very consistent (not shown), and are in the range between 3.5
15 to 18.5 kt.





1 Figure 7: (a) Comparison of SO₂ center of mass height (km) from TROPOMI and PlumeTraj for
2 17 Etna paroxysmal events in 2021. (b) Differences of TROPOMI minus PlumeTraj SO₂ height.
3 (c) Median and maximum SO₂ columns retrieved by TROPOMI LUT-COBRA.

4

5 Figure 7b shows the differences in height between TROPOMI and PlumeTraj for all paroxysmal
6 events, and Fig. 7c summarizes the corresponding median and maximum SO₂ column values as
7 retrieved by TROPOMI SO₂ LUT-COBRA.

8 For about two third of the cases, the height estimates agree within ± 1 km. There are only a few
9 instances for which the height difference is higher than 1.5 km (in absolute value). However,
10 further investigation reveals that these cases correspond to particularly difficult conditions either
11 for the satellite retrievals (e.g., due to the presence of volcanic ash or because the plume was
12 narrow) or for PlumeTraj (unfavorable wind shear settings or because the plume was old).
13 Importantly, from Figs 7b and 7c, we cannot find a relation between the height discrepancy and
14 the SO₂ loadings. The median SO₂ VCD lies in the range between 6 and 25.6 DU, and Fig. 7
15 confirms that the TROPOMI SO₂ LH algorithm is able to derive reasonable SO₂ heights, even for
16 modest SO₂ vertical columns.

17

18 **3.3 Temporal analysis over degassing volcanoes**

19

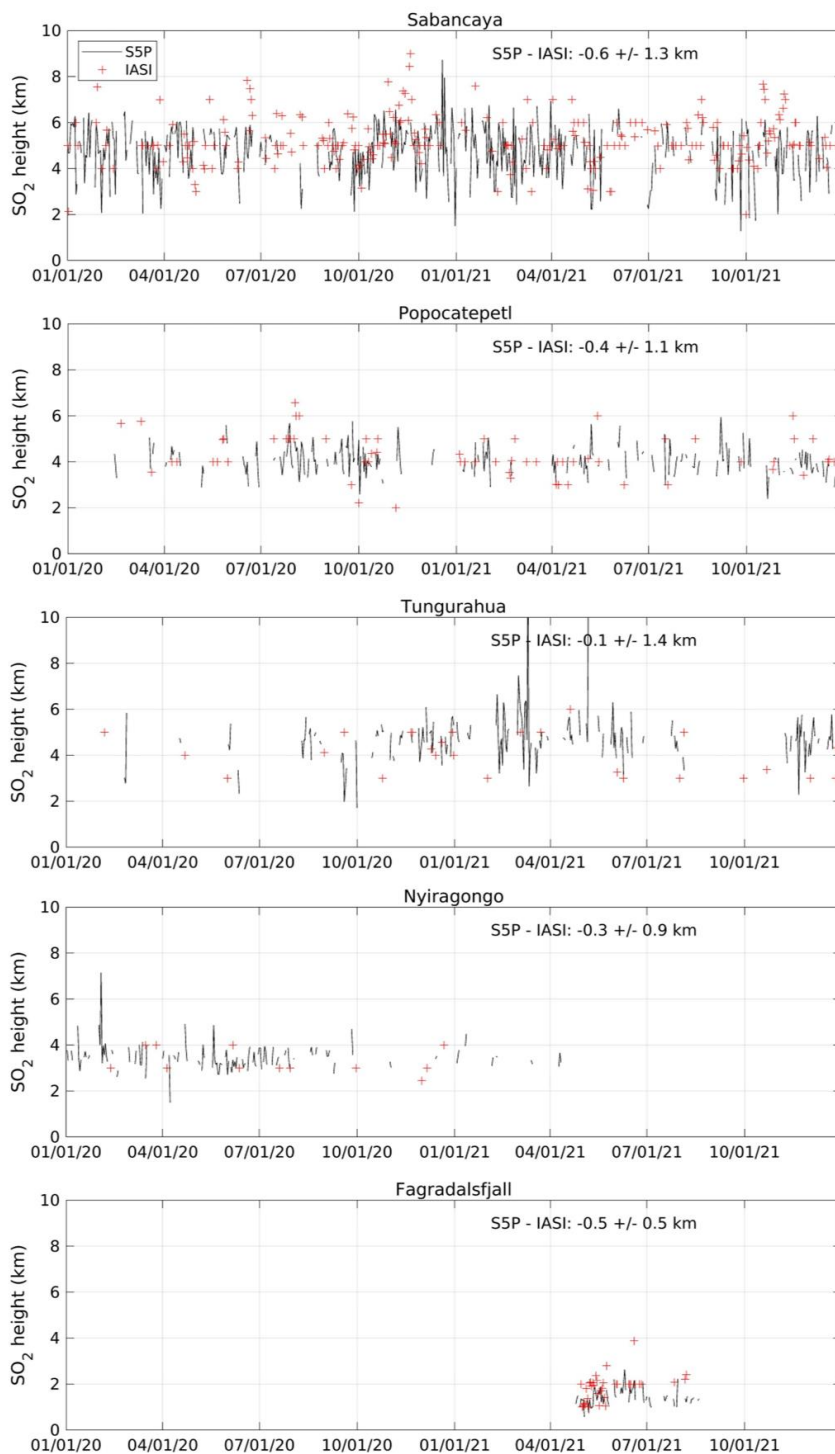
20 Apart from eruptive events, TROPOMI is able to detect SO₂ emissions from degassing volcanoes
21 worldwide (Queißer et al, 2019; Theys et al., 2019, 2021; Fioletov et al., 2020), and it is therefore
22 important to test our SO₂ LH algorithm on some of these volcanic emitters. Previous studies have
23 shown that IASI is sensitive to weaker volcanic emissions as well (Clarisse et al., 2012; Taylor et
24 al., 2018), and thus it is interesting to compare the TROPOMI and IASI height retrievals. For this,
25 we have considered two complete years of data (2020 and 2021) and analyzed time series of daily
26 height estimates from TROPOMI and IASI/MetOp-B over many different volcanic regions.
27 Because of the limited sensitivity of the satellites, it is clear that not all comparisons were
28 meaningful. However, for some volcanoes, the height of SO₂ was regularly retrieved by both
29 instruments over the studied period. Example of results are shown in Figure 8 for five active
30 volcanoes, namely Sabancaya, Peru (15.78°S, 71.85°W, summit elevation: 5967 m), Popocatepetl,
31 Mexico (19.02°N, 98.62°W, 5426 m), Tungurahua, Ecuador (1.47°S, 78.44°W, 5023 m),



1 Nyiragongo, DR Congo (1.52°S, 29.25°E, 3470 m), and Fagradalsfjall, Iceland (63.90°N, 22.27°E,
2 385 m). Note that the IASI height retrievals are the same as already introduced in Section 3.1,
3 except that additional criteria were applied to select the data with sufficient SO₂ signal and to make
4 the results comparable to TROPOMI. In particular, the same lower threshold of 5 DU for the
5 vertical column is applied.

6 As can be seen from Figure 8, a remarkable agreement is found between TROPOMI and IASI SO₂
7 heights. For Sabancaya, it is noticeable that successful SO₂ LH retrievals are frequent for both
8 instruments. The reason is likely due to the relatively high SO₂ columns there but also because
9 Sabancaya is an elevated site characterized by a dry atmosphere. To some extent, this is also true
10 for Popocatepetl and Tungurahua. On the contrary, a site such as Nyiragongo has a summit at
11 lower altitude and a wet atmosphere, resulting in fewer IASI retrievals. Finally, for Fagradalsfjall,
12 SO₂ was emitted much lower in the atmosphere (mainly below 2 km height) than for the other
13 cases. However, the match between TROPOMI and IASI is very good. In this case, IASI seems to
14 be able to retrieve SO₂ LH below 2 km. This is the result of dry conditions over Iceland for the
15 studied period. Note that the time series for Fagradalsfjall covers only a few months, after its 2021
16 fissure eruption.

17 Figure 8 indicates that TROPOMI tends to retrieve slightly lower SO₂ heights than IASI by ~ 0.5
18 km, although there is significant scatter in the height differences (standard deviation of about 1
19 km). The nature of this small systematic difference is unknown. However, this result nicely
20 demonstrates the value of the LUT-COBRA approach to infer the height of SO₂ for degassing
21 volcanoes or modest eruptions. For plumes in the lower troposphere, more frequent SO₂ heights
22 are retrieved with TROPOMI than IASI, which is an appealing aspect of the algorithm.





1

2 Figure 8: Time series of SO₂ height over five volcanic regions from TROPOMI and IASI/MetOp-
3 B (daytime observations) for January 2020 to December 2021. Daily estimates of SO₂ center of
4 mass height were calculated from quality-filtered data using fixed latitude-longitude boxes (from
5 top to bottom) for Sabancaya (12-20°S, 70-78°W), Popocatepetl (15-22°N, 96.5-102°W),
6 Tungurahua (5°S-1°N, 77-83°W), Nyiragongo (5°S-3°N, 25-32°E), and Fagradalsfjall (60-70°N,
7 10-32°W). For IASI, the same VCD lower threshold of 5 DU as TROPOMI is applied to select the
8 data. The calculated SO₂ heights are shown only for days with at least 2 pixels. The mean and
9 standard deviation of the differences between TROPOMI and IASI SO₂ heights are given as an
10 inset for each plot.

11

12 4. CONCLUSIONS

13 We have presented a new algorithm to retrieve the SO₂ layer height and vertical column from
14 TROPOMI UV observations. The retrieval scheme combines a large look-up-table to model the
15 SO₂ signal and an error covariance-based approach, to represent the SO₂-free contribution of the
16 spectrum. The method minimizes atmospheric or instrumental-related spectral interferences and
17 reduces the SO₂ layer height error by a factor of 2 to 3 compared to a DOAS-fashioned
18 implementation of the algorithm. This enables derivation of the SO₂ layer height with a precision
19 better than 2 km for SO₂ columns as low as 5 DU and for a wide range of conditions. This is a
20 significant improvement compared to other existing UV retrievals, which are limited to scenes of
21 at least 20 DU of SO₂ columns.

22 We have demonstrated this approach on a number of eruptive events. Comparison with satellite
23 IASI and CALIOP measurements and back-trajectory analyses indicate an agreement within 1-2
24 km, except for specific observations conditions. The presence of ash, in large amounts and at the
25 same altitude as SO₂, causes the retrieval to underestimate the SO₂ height by several kilometers,
26 in line with previous studies. Moreover, partially SO₂-filled scenes underestimate the SO₂ layer
27 height, and this is mostly seen at plume edges. Despite these limitations, the performance of the
28 algorithm is particularly good, especially for plumes below 10-12 km. We investigated the results
29 against back trajectory analysis from the PlumeTraj toolkit, for relatively modest eruptions of
30 Mount Etna in 2021. Using column-weighted average heights of SO₂, we found a very good
31 agreement with PlumeTraj, even for total SO₂ masses of a few kt. Capitalizing on this, the temporal



1 evolution of TROPOMI SO₂ plume height was studied over some of the largest degassing
2 volcanoes, for a period of two years. An excellent correspondence is found between TROPOMI
3 and IASI with a mean difference of -0.5 km. This highlights the high sensitivity of the proposed
4 technique for the determination of plume height.

5 The algorithm is fast and could be adapted for near real-time implementation, and used e.g. in the
6 Support to Aviation Control Service, or other volcanic monitoring applications. The SO₂ height
7 results could also be helpful as a constraint for atmospheric dispersion modeling.

8 Future developments will focus on possibly enhancing the algorithm sensitivity, improving the
9 retrievals in the presence of aerosols, expanding the algorithm to stratospheric injection heights
10 (e.g., the 2022 eruption of Hunga Tonga–Hunga Ha’apai), and producing a proper quality
11 assurance flag.

12

13 **DATA AVAILABILITY**

14

15 The TROPOMI COBRA SO₂ layer height dataset is available from the corresponding author on
16 request. The ULB IASI SO₂ dataset is available from Dr. Lieven Clarisse on request. The output
17 of the PlumeTraj tool is available from Dr. Mike Burton on request. NASA CALIPSO data can be
18 downloaded from <https://www-calipso.larc.nasa.gov/> (last access: 30 March 2022).

19

20 **AUTHOR CONTRIBUTIONS**

21

22 N.T. prepared the manuscript and figures with contributions from all the coauthors. N.T., C.L.,
23 J.v.G., H.B., I.D.S., M.V.R. contributed to the development of the LUT-COBRA, processing of
24 the data and satellite comparison. L.C. analyzed and provided IASI data. M.B. and M.V. analyzed
25 and provided PlumeTraj data. All authors contributed to the interpretation of the results and
26 improvement of the manuscript.

27

28 **COMPETING INTERESTS**

29

30 The authors declare that they have no conflict of interest.

31

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33



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1 Appendix

2 Table A1. Input settings used to generate radiative transfer simulation.

Model	Lidort RRS; Raman scattering switched off.
Wavelength range	309-329 nm, 0.05 nm spectral sampling. All spectroscopic data are pre-convolved using 0.05 nm box-car function.
Solar spectrum	Chance and Kurucz (2010).
Cross-sections	Ozone: Serdyuchencko et al. (2014), SO ₂ : Bogumil et al. (2003) Temperature dependence of the cross-sections are accounted for in the simulations.
Atmosphere	Ozone and temperature profiles: total ozone column classified profiles from Lamsal et al. (2004). All available climatological profiles are averaged for each total ozone column value of 145, 175, 205, 235, 295, 355, 415, 475, 535 DU. Pressure profile (US Standard).
SO₂ profiles	Gaussian profiles with full width at half maximum of 500 m, peaking at SO ₂ height and scaled to VCD as in Table 1.
Aerosols and clouds	Not included in the simulations (treated as LER by the algorithm).
Output	Radiance and SO ₂ slant optical depth (log ratio of radiance simulations including/not-including SO ₂ absorption).

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