Automatic estimation of daily volcanic sulfur dioxide gas flux from TROPOMI satellite observations: application to Etna and Piton de la Fournaise

Raphael Grandin\textsuperscript{1}, Marie Boichu\textsuperscript{2}, Théo Mathurin\textsuperscript{3}, and Nicolas Pascal\textsuperscript{3}

\textsuperscript{1}Université de Paris, Institut de physique du globe de Paris, CNRS
\textsuperscript{2}CNRS/Université de Lille, Laboratoire d’Optique Atmosphérique, CNRS
\textsuperscript{3}University of Lille, CNRS, CNES, UMS 2877 – ICARE Data and Services Center

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Raphaël Grandin¹, Marie Boichu², Théo Mathurin¹, and Nicolas Pascal³

¹Institut de Physique du Globe de Paris, Université Paris Cité, ForM@Ter
²Laboratoire d’Optique Atmosphérique, Université de Lille, CNRS
³AERIS/ICARE, Université de Lille, CNRS

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Key Points:
• Daily volcanic SO₂ flux is deduced from TROPOMI satellite imagery by mass-to-distance regression, including a noise estimation procedure
• SO₂ emission rates at Etna, during episodes of passive and eruptive degassing, demonstrate a good correlation with seismic energy
• The algorithm is made available to all as an open-source Python package and on the interactive web application “SO₂ Flux Calculator”

Corresponding author: Raphael Grandin, grandin@ipgp.fr
Abstract

Understanding the dynamics of sulfur dioxide (SO$_2$) degassing is of primary importance for tracking temporal variations in volcanic activity. Here we introduce the novel “disk method”, which aims at estimating the daily volcanic SO$_2$ mass flux from satellite images (such as those provided by Sentinel-5P/TROPOMI). The method calculates a “proto-flux” using a regression, as a function of distance, of SO$_2$ mass integrated in a series of nested circular domains centered on a volcano. After regression, a single multiplication by plume speed suffices to deduce the SO$_2$ mass flux, without requiring a subsequent regression. This way, a range of plume speed and plume altitude scenarios can be easily explored. Noise level in the image is simultaneously evaluated by the regression, which allows for estimating posterior uncertainties on SO$_2$ flux and improving the level of detection for weak sources in noisy environments. A statistical test is also introduced to automatically detect occurrences of volcanic degassing, lowering the risk of false positives.

Application to multi-year time-series at Etna (2021) and Piton de la Fournaise (2021–2023) demonstrates (a) a reliable quantification of SO$_2$ emissions across a broad range of degassing styles (from passive degassing to effusive or paroxysmal events), and (b) a reasonable day-to-day correlation between SO$_2$ flux and seismic energy. The method is distributed as an open-source software, and is implemented in an interactive web application within the “Volcano Space Observatory Portal”, facilitating near-real-time exploitation of the TROPOMI archive for both volcano monitoring and assessment of volcanic atmospheric hazards.
Plain Language Summary

Volcanic eruptions emit sulfur dioxide gas (SO$_2$) into the atmosphere, which may cause harm to populations and the environment, and need to be monitored. Tracking volcanic emissions is also important for volcanologists to detect changes on a given volcano, and anticipate eruptions. SO$_2$ can be observed by satellites every day, but exploitation of satellite imagery requires complex procedures. Wind speed is a crucial ingredient, but it is often poorly known, leading to large uncertainties in estimated SO$_2$ mass. Here, a simple algorithm is proposed for analyzing SO$_2$ images provided by satellites. The mass of SO$_2$ is extracted in an area surrounding a volcano (typically 500 km) to estimate the quantity of SO$_2$ released, as well as associated uncertainties. Plume speed information can be incorporated after running the algorithm, which facilitates testing different plume speed scenarios. Application to Etna and Piton de la Fournaise volcanoes shows that temporal variations of SO$_2$ emissions follow the same pattern as seismic energy recorded by ground seismometers, which gives confidence in the results. The algorithm is made available to all as open-source code and in an open-access interactive web application within the framework of the “Volcano Space Observatory Portal”.

1 Introduction

The release of volcanic material into the atmosphere, in the form of lava, tephra, aerosols and gas, represents a major source of hazard for populations living in the vicinity of volcanoes (Loughlin et al., 2015). Volcanic emissions can also put aircrafts at risk (Prata & Rose, 2015) and lead to a deterioration of air quality (Stewart et al., 2022). Among the various species of volcanic gases, sulfur dioxide ($\text{SO}_2$) is of primary importance, since it is the main precursor of sulfate aerosols, which can affect climate (e.g. Marshall et al., 2022, and references therein). Furthermore, since it separates from magma at shallow depth, $\text{SO}_2$ is the most effective gas species for tracking the dynamics of volcanic activity using remote sensing techniques, a task facilitated by its characteristic spectral signature and low background concentration in the atmosphere (Oppenheimer et al., 2011). Alongside measurements of ground deformation, estimating the $\text{SO}_2$ budget of a given volcano places constraints on the architecture of magma reservoirs, especially on the volume of magma stored or transported during periods of passive degassing, unrest, or during effusive or explosive eruptions (Girona et al., 2014; Kilbride et al., 2016; Shreve et al., 2022).

For any given eruption, the primary $\text{SO}_2$ emission parameters that need to be constrained are (i) the emission rate, or mass flux, and (ii) the altitude of emissions, or injection height. Emission rate is indicative of the dynamics of the eruption, and may change by orders of magnitude over time scales as short as a few hours, and is therefore a prime target for volcano monitoring (e.g. Aiuppa et al., 2015). On the other hand, constraining emission height is critical for Volcanic Ash Advisory Centers (VAAC), as it is essential for initializing simulations of atmospheric transport and accurately predicting the trajectory and lifetime of volcanic parcels (e.g. Stohl et al., 2011; Brenot et al., 2014; Boichu et al., 2015). In theory, plume injection height scales with the fourth root of the eruptive mass flux (Morton et al., 1956; Sparks et al., 1997). However, the relationship is in reality subject to substantial variability (e.g. see the compilations by Mastin et al., 2009; Aubry et al., 2023), due to the dependency of the dynamics of plume ascent upon external factors (e.g. atmospheric conditions, Tupper et al., 2009) or internal factors (e.g. particle grain-size distribution, Girault et al., 2014). Hence, deriving the emission rate directly from the plume height, or vice-versa, is not straightforward. When possible, the two quantities need to be constrained independently.
SO$_2$ emissions can be tracked from the ground using networks of UV-DOAS instruments installed near the volcanic source (e.g. Arellano et al., 2021), which are capable of measuring weakly- to moderately-degassing sources, such as events of transient pre-eruption degassing, or continuous, passive degassing. Unfortunately, a minority of active volcanoes are monitored from the ground, mainly due to a lack of resources (Loughlin et al., 2015). Furthermore, the viewing geometry from the ground means there is a limited view of the plume. Hence, when activity escalates, observation from ground sensors may become less reliable, or even fail entirely, especially near the source where a high abundance of ash and aerosols may lead to plume opacification (e.g. Andres & Schmid, 2001; Kern et al., 2012; Boichu et al., 2015; Kern et al., 2020). During large eruptions, personnel safety may also be threatened (e.g. at Merapi in 2010 or Soufrière Saint Vincent in 2021, see Surono et al., 2012; Joseph et al., 2022, respectively). As a consequence, satellite remote sensing is currently being considered as a cost-efficient way to complement ground-based observation systems and lessen the risk of observational gaps and biases (National Academies of Sciences, Engineering, and Medicine, 2017). In this context, it is essential to develop efficient, automatic and portable algorithms to leverage the wealth of satellite data currently available. This will better assist local observatories, decision-makers and the atmospheric and volcanology science communities in their respective tasks (Pritchard et al., 2022).

Today, SO$_2$ abundance in the atmosphere can be mapped by various imaging hyperspectral sensors onboard low-Earth orbit platforms (see Carn et al., 2016; Theys et al., 2019; Hyman & Pavolonis, 2020, and references therein), either operating in the ultraviolet (e.g. Sentinel-5 Precursor/TROPOspheric Monitoring Instrument, hereafter referred to as “TROPOMI”, Aura/OMI, Suomi NPP/OMPS) or infrared domains (e.g. MetOp/IASI, JPSS/CrIS, Aqua/AIRS). These systems provide near-global coverage every 24 h for UV, or 12 h for IR (IR sensors being capable of both daytime and nighttime acquisitions), with a spatial resolution ranging from 5 km to 50 km, achieving variable levels of sensitivity with altitude (UV sensors being more sensitive to low-altitude SO$_2$).

SO$_2$ detection can also be achieved at higher temporal resolution by geostationary sensors (every 10–15 minutes for MSG/SEVIRI, GOES/ABI or HIMAWARI/AHI, and exceptionally down to 30 seconds in on-demand zoom mode for GOES/ABI), or at higher spatial resolution by multispectral sensors in low-Earth orbit (≈ 1 km pixel size for Aqua/MODIS, Suomi NPP/VIIRS, or 90 m for Terra/ASTER). However, these
specifications are obtained at the expense of the spectral resolution, which leads to a curtailment of the detection capability due to the presence of water vapor, ash or meteorological clouds (Thomas et al., 2011; Theys et al., 2019; Corradini et al., 2021).

Currently, TROPOMI provides the best spatial resolution among all hyperspectral sensors capable of daily near-global coverage (Theys et al., 2019; Fioletov et al., 2020), and will be the primary focus of this study.

To derive source terms (mass flux and altitude) from satellite images, it is necessary to account for the spatio-temporal evolution of gas parcels in the atmosphere, from their injection point to their observation location. Several approaches have been proposed to estimate volcanic flux from satellite images of \( \text{SO}_2 \) column amount (see also Theys et al., 2013, for a summary of the different methods):

1. the “Delta-M method” and “Box method” calculate the mass emitted in a known time interval (computed either from the mass burden in a single image, or by differencing between successive images), divided by the time span, correcting for an empirical gas loss rate (Krueger et al., 1996; Lopez et al., 2013; Theys et al., 2013; Coppola et al., 2019; Carboni et al., 2019).

2. “Plume traverses” consist of computing plume cross-sections (defined as the integral of column amounts on a transect perpendicular to the plume), followed by multiplication by plume speed (Carn et al., 2003; Merucci et al., 2011).

3. “Wind-rotation” methods apply a correction to compensate changing day-to-day plume directions and speeds, which makes it possible to fit a simplified model of gas transport, loss rate and dispersion, either on daily observations, or on stacked measurements providing monthly- or annually-averaged emission budgets released by “hotspots” (Beirle et al., 2014; Fioletov et al., 2016; Carn et al., 2017; Hyman et al., 2021; Fioletov et al., 2023).

4. “Inverse modelling” attempts to match the observed spatial distribution of vertical column densities against simulations from a numerical (chemistry-)transport model, initialized with a weather model, thereby incorporating potentially complex atmospheric processes such as diffusion, deposition and/or chemical conversion (Eckhardt et al., 2008; Kristiansen et al., 2010; Boichu et al., 2013; Theys et al., 2013; Flemming & Inness, 2013; Moxnes et al., 2014; Boichu et al., 2014, 2015; Vira et al., 2017; Heng et al., 2016; Cai et al., 2021; Behera et al., 2023).
5. the “Back-trajectory” approach estimates the time-of-flight of gas parcels associated with each pixel in a satellite image, and deduces time and altitude of emissions by back-projecting these individual parcels into the emission parameter space, using a back-trajectory model (Hughes et al., 2012; Queißer et al., 2019; Hayer et al., 2023; Markus et al., 2023; Esse et al., 2024).

Overall, these approaches all require knowledge of the plume direction (except for the simple “Delta-M method” of Krueger et al., 1996), plume speed, and often plume altitude. Unfortunately, these quantities can be uncertain, as they are derived from indirect information (e.g. meteorological reanalysis, radiosonde data, meteorological radar, or advanced satellite retrieval). Secondly, in order to mitigate the impact of background noise, these methods all apply a form of prior selection of points believed to be representative of volcanic emissions. This is achieved by outlining the plume boundary or by removing soundings with a low column amount, which introduces a bias whose impact is seldom quantified. Finally, none of the aforementioned methods is distributed as open-source code (except for the source separation algorithm of Markus et al., 2023) and none is associated with a publicly accessible web application.

Here, we introduce a novel method, hereafter designated as the “disk method”, to estimate SO$_2$ flux released by a volcanic point source. The method starts with the computation of the SO$_2$ mass burdens integrated in a series of nested circular domains centered on a volcano (Figure 1a, Step 1). A regression is then performed to estimate the volcanic flux, which is predicted to behave as a linear term with distance, according to a “Gaussian plume” model, under the “slender plume approximation” (i.e. assuming that along-plume diffusion is negligible compared to advection speed). On the other hand, background noise is modeled as a “truncated normal distribution”, acting as a quadratic term in the regression. The regression provides an estimation of the SO$_2$ flux and its associated uncertainty, together with a characterization of spatially-averaged noise in the input satellite data (Figure 1a, Step 2). The regression is wind-agnostic, rotation invariant, so that knowledge of plume speed or altitude can be accounted for at the post-inversion stage (Figure 1a, Step 3). Based on a statistical significance test, the method also allows for automatically detecting “true” SO$_2$ emissions sourced from the volcanic target and separating them from external perturbations.
In the next section, we present the gas and seismicity datasets analyzed in the paper. In Section 3 we describe the theoretical model and its algorithmic implementation. In Section 4 we assess the sensitivity of the inversion to free and internal parameters. In Section 5 we apply the method to a real dataset, computing long time-series with daily resolution on two volcanoes with contrasting dynamics (Etna, 2021 and Piton de la Fournaise, 2021–2023), examining the relationship between degassing and seismic energy. Finally, in Section 6 we provide recommendations on how to adjust the free-parameters of the method, and provide a few perspectives.

2 Data

2.1 SO$_2$ satellite imagery

TROPOMI provides daily near-global observations of trace gas and aerosols around 13:30 local solar time, with an approximate spatial resolution of 3.5×5.5 km at nadir (Veefkind et al., 2012). We use the TROPOMI Level-2 (L2) SO$_2$ product, with SO$_2$ retrieved at 1 km and 7 km altitude (Theys et al., 2022). The SO$_2$ retrieval in the L2 product is based on the DOAS technique (Theys et al., 2017).

For practical exploitation of TROPOMI data, an optional data preselection step may be applied. First, a preselection based on the column amount value may be performed, as retrievals may be considered as dominated by noise when the column amount is below a certain cutoff threshold (an operation hereafter referred to as “truncation”). The implications of truncation will be discussed specifically in the following sections. Furthermore, a certain number of swath-edge rows may be discarded, as SNR degrades close to the edge of the TROPOMI swath. For example, Fioletov et al. (2020) remove 20 swath-edge rows, but strictly following this criterion produces periodic observation gaps at low latitudes (within ±30°N). Hence, when applying the algorithm to real data in Section 5, only 7 columns will be removed to prevent gaps at Piton de la Fournaise (21.24°S), whereas 22 columns will be discarded at Etna (37.75°N). No further preselection is applied, in particular depending on sounding quality. Removal of dates based on spatially-averaged solar zenith angle or cloud fraction may be performed a posteriori, as discussed in Section 6.

All selected soundings acquired within a 24 hour time window are concatenated and resampled to a regular 0.05° × 0.05° grid (which corresponds roughly to a 5 km × 5 km
pixel size at the Equator). In the interior of the convex hull, we use a linear interpolation, with gap-filling up to a maximum distance of 1° from the closest valid pixel. No extrapolation is made outside the convex hull.

The resulting SO$_2$ column amount for each resampled pixel is noted $p_i$ (expressed in Dobson Units or DU, where 1 DU = $2.69 \times 10^{16}$ molecules.cm$^{-2}$). Each pixel is converted to an SO$_2$ columnar mass, noted $x_i$ (expressed in kton) via a conversion formula

$$x_i = \kappa \cdot p_i,$$ with $\kappa = 2.69 \times 10^{16} \frac{M_{SO_2}}{N_{Avo}} A,$ where $N_{Avo}$ is the Avogadro number, $M_{SO_2}$ the molar mass of SO$_2$, and $A$ the resampled pixel area. Hereafter, we use $A = 25$ km$^2$ (pixel size after regridding), such that $\kappa \approx 7 \times 10^{-4}$ kton.DU$^{-1}$.

Finally, SO$_2$ mass is integrated in circular regions centered on a volcano, by summation of the pixels located in the interior of a disk of radius $r_n$:

$$M(r_n) = y_n = \sum_{i=1}^{n} x_i$$ (1)

where $i$ is the pixel index, $x_i$ the pixel SO$_2$ mass, $n$ the number of summed pixels, and $M(r_n)$ the integrated SO$_2$ mass. Thanks to the regridding step, the number of summed pixels in the summation domain, $n$, can be directly deduced from the radius $r_n$ of the disk according to:

$$n = \frac{\pi r_n^2}{A}$$ (2)

2.2 Seismic data

Real-time seismic amplitude (RSAM, Endo & Murray, 1991) is often interpreted as a proxy for the lava discharge rate (e.g. Battaglia et al., 2005; Ichihara, 2016). In the absence of influences such as excess degassing or gas scrubbing, the mass of SO$_2$ emitted during an eruption is often considered proportional to the volume of erupted lava (e.g. Nadeau et al., 2011; Hibert et al., 2015). Hence, at first approximation, a direct comparison of RSAM and SO$_2$ flux can be used to assess the reliability of satellite-based estimations of SO$_2$ flux (e.g. Boichu et al., 2015; Hayer et al., 2023).

To compute the RSAM, raw seismic data is first converted to ground velocity by applying an instrument response correction. Data is then filtered between 1 Hz and 5 Hz, and RSAM is calculated over 60-seconds time windows. Comparison of RSAM with daily SO$_2$ time-series requires a specific procedure to correct for the time lag between the seismic measurement (which is synchronous with the emission of lava or gas) and the satellite
overpass (which measures the gas mass after its emission). We synchronize the seismic record to the temporal sampling of the satellite products by applying a causal rolling average filter to the RSAM time-series, with a window length $\theta$ (i.e. we replace each RSAM record at $t$ by its mean in the preceding time interval $[t-\theta, t]$). Considering a characteristic length of the plume $L_{\text{plume}}$, and a plume speed $u$, we can deduce the appropriate delay using $\theta = L_{\text{plume}}/u$.

3 Methodology

3.1 Theoretical foundation for the “disk method”

3.1.1 Gaussian plume model

The advection-diffusion equation describes the distribution of mass concentration $C$ for a gas during its transport and diffusion in the atmosphere. In Cartesian coordinates, and assuming an incompressible flow, this equation is expressed:

$$\frac{\partial C}{\partial t} + u_x \frac{\partial C}{\partial x} + u_y \frac{\partial C}{\partial y} + u_z \frac{\partial C}{\partial z} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - kC + S \quad (3)$$

where emissions are described by the source term $S$, whereas gas loss is modeled by a sink term with first-order decay at constant rate $k$. The $x, y, z$ components of advection velocity are $u_x, u_y, u_z$, the corresponding coefficients of diffusion are $D_x, D_y, D_z$, and time is noted $t$ (Equation 18.11, p. 768 in Seinfeld & Pandis, 2016).

In order to simplify Equation 3, several assumptions are made:

- we assume steady-state ($\partial/\partial t = 0$), thus the source term $S$ is constant.
- we consider a transport taking place in the $x$-direction (such that $u_y = u_z = 0$ and $u_x \neq 0$), which can be accommodated by a rotation of the coordinate system. Hereafter, we will note $u = u_x$.
- we use the “slender plume” approximation, which assumes that advection dominates over along-plume diffusion. This assumption corresponds to a large Péclet number $P_e$, i.e. $P_e = L u / D_x \gg 1$, with $L$ a characteristic length. Taking the “e-folding distance” $u/k$ as the characteristic length $L$ (e.g. as in Hyman et al., 2021) translates the slender plume approximation into: $P_e = u^2 / D_x k \gg 1$. The extent of the $P_e \gg 1$ domain, as a function of $u$, $k$ and $D_x$ is represented in Figure S1.
These simplifications allow for rewriting Equation 3 as:

\[
\frac{u \partial C}{\partial x} = D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - kC + S \tag{4}
\]

The solution of this equation for a point source at \(x = 0, y = 0\) and \(z = 0\) releasing mass at a constant rate \(\dot{m}\) (mass flux rate) can be written as:

\[
C(x, y, z) = \frac{\dot{m}}{4\pi x \sqrt{D_y D_z}} \exp \left\{ \frac{-u}{4x} \left( \frac{y^2}{D_y} + \frac{z^2}{D_z} \right) \right\} \exp \left\{ \frac{-kx}{u} \right\} \tag{5}
\]

where the classical Gaussian solution without gas loss (Equation 18.63, p. 777 in Seinfeld & Pandis, 2016) is multiplied by an exponential depletion factor (see e.g. Overcamp, 1982).

Satellite sensors provide \(\text{SO}_2\) column amounts, which correspond to a mass per unit area integrated over a vertical column. In our plume model, vertical integration of concentration \(C\) in Equation 5 gives the following expression for the column amount \(D\), which becomes independent of the vertical diffusion term:

\[
D(x, y) = \int C(x, y, z) \, dz = \frac{\dot{m}/u}{\sqrt{4\pi D_y (x/u)}} \exp \left\{ \frac{-uy^2}{4D_y x} \right\} \exp \left\{ \frac{-kx}{u} \right\} \tag{6}
\]

Besides wind speed \(u\), Equation 6 depends on two atmospheric parameters. First, the gas loss rate \(k\), which can span many orders of magnitude, depending on plume properties and atmospheric conditions, in particular plume injection height (e.g. \(k = 10^{-7} - 10^{-3} \text{ s}^{-1}\), according to Carn et al., 2016; Pattantyus et al., 2018). Hence, its reciprocal \(\tau = 1/k\), the “e-folding time of gas loss”, varies from tens of minutes to several days. In addition, in Equation 6, the horizontal cross-plume diffusion coefficient \(D_y\) (also known as the “cross-wind eddy diffusivity”) describes the progressive cross-plume spreading of \(C\) with time, hence with distance from the source. Typical values for tropospheric volcanic plumes in the range of \(D_y = 0.5 - 3 \times 10^4 \text{ m}^2 \text{ s}^{-1}\) have been reported by Barr and Gifford (1987), whereas a lower \(D_y = 0.1 \times 10^4 \text{ m}^2 \text{ s}^{-1}\) was estimated for a 6 km-high plume at Etna by Tiesi et al. (2006). An example of a modeled column amount distribution is represented in Figure 2a, showing a Gaussian shape in any downwind cross-plume profile (yellow dots in Figure 2e). The validity of the plume model, in particular the slender plume approximation, as a function of \(D_y, k\) and \(u\) will be addressed in Section 4.
3.1.2 Relationship between integrated mass and mass flux

Integration of the column amount $D$ of Equation 6 in a 2D circular domain of radius $r$ centered on the volcano provides the total mass of $SO_2$, $M_{volc}$, released in the time interval $t \in [-T,0]$, with $T = r/u$ (taking $t = 0$ for the acquisition time). To a first approximation, this circular integration is equivalent to integration over a semi-infinite rectangular domain perpendicular to the direction of transport, in the interval $x = [0,r]$:

$$M_{volc}(r) = \int_{R=0}^{R=r} D(R) dS \approx \int_{x=0}^{x=r} \int_{y=-\infty}^{y=+\infty} D(x,y) \, dx \, dy = \frac{\dot{m} u}{k} \left(1 - \exp \left\{ -\frac{kr}{u} \right\} \right) \tag{7}$$

Note that the equation remains valid if advection occurs in a direction different from the $x$-axis (rotation invariance). Furthermore, this expression becomes independent from cross-plume diffusivity $D_y$, and only depends on gas loss rate $k$.

A first-order expansion of Equation 7 yields a linear evolution of integrated mass as a function of distance $r$ (yellow dots in Figure 2f):

$$M_{volc}(r) \approx \frac{\dot{m} u}{u} r \tag{8}$$

The proportionality coefficient, $\left\{ \frac{\dot{m}}{u} \right\}$, hereafter named “proto-flux”, is a lumped quantity that condenses the advection speed $u$ and proportional to the mass flux $\dot{m}$ averaged over the time interval $T = r/u$ before the image acquisition. The “proto-flux” can be estimated by linear regression of the integrated mass $M(r)$ versus distance $r$, where $M(r)$ is directly calculated from real data using Equation 1. In practice, the range of distances used in the regression needs to be optimized, depending on gas loss rate $k$.

The approximation of Equation 8 entails an underestimation of the “true” flux by a maximum 37% at $r = u/k$ (or, equivalently, at $T = \tau$). Thus, depending on the tolerance of the downstream application, Equation 8 may remain valid up to $r \approx u/k$. The effect of the maximum distance $r_{max}$ used for fitting Equation 8 will be specifically discussed in Section 4, and recommendations for defining $r_{max}$ will be provided in Section 6.1.

3.2 Pixel noise characterization

3.2.1 Spatially-averaged background noise

The value of an individual pixel in a TROPOMI image is the result of a retrieval algorithm, which translates raw radiance measurements into vertical column densities, or column amounts. In the following, we treat the mass $x_i$ for pixel $i$ as the realization of a
random variable $X_i$ with expectation $E[X_i]$ and variance $\text{Var}(X_i)$. In what follows, we temporarily ignore the presence of volcanogenic or anthropogenic SO$_2$, to focus only on stochastic properties of the “background noise”.

Any retrieval algorithm aims at optimizing the accuracy of the reported column amounts, which implies keeping $E[X_i]$ as close to zero as possible when the true gas concentration is zero. On the other hand, the pixel variance $\text{Var}(X_i)$ can be understood as the precision of the retrieval algorithm, and should be as small as possible.

The quality of the retrieval mainly depends on variations in surface reflectivity, solar zenith angle, atmospheric conditions, proximity to swath edge, 3D effects, while also depending on the retrieval algorithm itself (McCormick et al., 2013; Fioletov et al., 2020; Wagner et al., 2023). In the literature, the “uncertainty of the retrieval” is generally reported in the form of a single aggregated standard deviation “$\sigma$” (after appropriate unit conversion, $\sigma = \kappa \cdot \sigma_{\text{CA}}$, where pixel mass standard deviation $\sigma$ is expressed in kton and pixel column amount standard deviation $\sigma_{\text{CA}}$ is expressed in Dobson Units). For TROPOMI SO$_2$, a standard deviation $\sigma_{\text{CA}} = 0.3$ DU is reported for the 7-km altitude product (Theys et al., 2019), and 1–1.5 DU for ground-level products (respectively, 1 DU over the tropics and 1.5 DU at high latitudes, Fioletov et al., 2020).

In practice, the actual value of the pixel standard deviation is variable in time and space. In the context of this study, we do not consider the spatial variability of noise, which is levelled out by the spatial integration of the SO$_2$ mass (Equation 1), such that the pixel index $i$ can be dropped. Thus, $\sigma_{\text{CA}}$ represents the level of noise averaged over a large number of pixels. On the other hand, temporal variations of the image quality are of primary importance, as they impact the uncertainty on the day-to-day integrated SO$_2$ mass (and thus SO$_2$ flux). Hereafter, the spatially-averaged standard deviation $\sigma_{\text{CA}}$ is considered as an unknown that we estimate independently for each TROPOMI image.

### 3.2.2 Quadratic dependence of mass with distance in the presence of noise

In the presence of noise in an SO$_2$ satellite image, care should be taken in the spatial integration of pixel mass over a circular domain (Equation 7). Ignoring the influence of the volcanic plume, the summation formula for independent random variables gives an expression for the expectation of the random variable $Y_n$ representing the integrated mass.
\[
M_n:\quad E[Y_n] = \sum_{i=1}^{n} E[X_i] = n.E[X] = \frac{\pi r^2}{A}.E[X]
\]  
where \(i\) is the pixel index and \(n\) is number of pixels in the summation (Equation 2). In Equation 9, since \(n\) is large, and following the same argument as for \(\sigma_{CA}\) in Section 3.2.1, pixel expectations \(E[X_i]\) are replaced by a single spatially-averaged quantity \(E[X]\).

Equation 9 indicates that the estimated volcanic flux will be **biased** by the presence of noise. Noise amplification will be proportional to both \(r_n^2\) and \(E[X]\), except in the ideal case where \(E[X] = 0\).

### 3.2.3 Effect of truncation

In addition to intrinsic pixel noise, **truncation** represents a distinct, but potentially dominant contribution to a non-zero pixel expectation \(E[X]\). Indeed, to mitigate the effect of noise when analyzing SO\(_2\) satellite images, a common practice consists in masking pixels with low column amounts. For instance, Theys et al. (2019) recommend discarding pixels with values smaller than \(3 \times \sigma_{CA}\) for volcanic applications, in order to keep only values that are well above the noise. As a consequence of truncation of the lowest values, the mean pixel value will increase on average, henceforth biasing the expectation of the integrated pixel mass \(E[Y_n]\) according to Equation 9.

For the sake of illustration, Figure 2 shows a synthetic TROPOMI image consisting of a superposition of (a) a plume and (b) random noise. The plume of Figure 2a is modeled using Equation 6, with parameters reported in the caption of Figure 2. Figure 2b represents noise only, assuming that individual pixel mass follows a zero-mean normal distribution. Figure 2c shows the result of summation of the plume and noise (Figure 2a + 2b), followed by truncation of pixels with column amounts below a cutoff threshold, hereafter noted \(CA_{min}\) (in Figure 2 we use \(CA_{min} = 0.3\) DU). Ignoring the contribution of noise, integration of SO\(_2\) mass in circular domains produces a nearly linear evolution with distance, as predicted from Equation 8 (yellow symbols in Figure 2f).

However, addition of noise, combined with truncation, gives rise to a quadratic behaviour that progressively outweighs the contribution of the plume with distance from the source (green symbols in Figure 2f).

In what follows, we derive an expression for the curvature of the quadratic contribution of noise in presence of truncation (see Supporting Text S1 for full details). Following the
notations of Section 3.2.1, the pixel mass prior to truncation, noted $X$, is assumed

normally-distributed, with expectation $E[X] = \kappa \mu_{CA}$ and variance $Var(X) = \kappa^2 \sigma^2_{CA}$,

where $\kappa$ is the unit conversion factor defined above. Truncation corresponds to replacing

pixel values $x$ by a new value $x'$, defined by: $x' = x$ if $x \geq \kappa CA_{min}$, else $x' = 0$. The

random variable describing pixel mass after truncation is noted $X'$, and follows a

truncated normal distribution (represented by the red part of the histogram in Figure 2b).

Its expectation and variance, noted $E[X']$ and $Var(X')$, can be expressed analytically as

a function of (i) the expectation $E[X]$ and variance $Var(X)$ of $X$ prior to cutoff, and (ii)

the truncation cutoff $CA_{min}$ (e.g. Johnson et al., 1994, Chapter 13, Section 10.1). The

expressions for $E[X']$ and $Var(X')$ are given in Supporting Text S1, Section S1.1, and the

corresponding moments for integrated mass $Y_n$ are provided in Section S1.2. In the

particular case where $E[X] \approx 0$, the leading term in the expectation $E[Y_n]$ takes a simple

Gaussian form, which allows for rewriting Equation 9 as:

$$ E[Y_n] = \sqrt{\frac{\pi}{2}} \cdot \frac{\kappa}{A} \cdot \sigma_{CA} \cdot \exp \left\{ -\frac{1}{2} \left( \frac{CA_{min}}{\sigma_{CA}} \right)^2 \right\} \cdot r_n^2 $$

(10)

Equation 10 provides a closed-form expression for the quadratic bias, as a function of the

data noise standard deviation $\sigma_{CA}$ and truncation threshold $CA_{min}$. This expression is

validated in an experiment with real TROPOMI data in Supporting Text S1, Section S1.3.

3.3 Flux estimation procedure

3.3.1 Forward problem formulation

Combining the plume model (Section 3.1, Equation 8: $M_{volc} \propto r_n$) and the noise model

(Section 3.2, Equation 10: $M_{noise} \propto r_n^2$), the integrated mass in a disk of radius $r_n$

centered on a volcano is expected to follow the regression model:

$$ M(r_n) = M_{volc}(r_n) + M_{noise}(r_n) + \epsilon_n $$

$$ = a \cdot r_n + b \cdot r_n^2 + \epsilon_n $$

(11)

where we need to solve for $a$ and $b$ given $M(r_n)$ for a list of radii $r_n$. The linear term $a$

represents the “proto-flux” $\left( \frac{\dot{m}}{u} \right)$ (Equation 8). The quadratic term $b$ absorbs the bias

inherited from the combination of pixel noise $\sigma_{CA}$ and cutoff threshold $CA_{min}$

(Equation 10). The cutoff $CA_{min}$ may be adjusted on a case-by-case basis, and should be

considered as a user-defined parameter, known a priori.
To complete the definition of the regression model, the error term $\epsilon_n$ should rigorously describe the uncertainties affecting observations $M(r_n)$, ideally in the form of a probability density function. Observations $M(r_n)$ are obtained by summation of the column amount for $n$ pixels, where $n$ is proportional to $r_n^2$ (Equation 2). The summed pixels have individual variances $Var(X_i)$, themselves proportional to $\sigma^2_{CA}$, and are assumed to be uncorrelated for simplicity. Hence, the variance formula for a linear combination of uncorrelated random variables implies that the variance of $\epsilon_n$ is proportional to both $r_n^2$ and $\sigma^2_{CA}$. Furthermore, $n$ being large (typically $>1000$), $\epsilon_n$ converges to a normal distribution. Since all biases are supposed to be already absorbed by the linear term $a_r n$ (volcanic) and the quadratic term $b_r^2 n$ (noise), the residual (error) term $\epsilon_n$ can be considered zero-mean.

In summary, using the standard notation for a normal random variable:

$$\epsilon_n \sim N\left(0, \gamma^2 \sigma^2_{CA} r_n^2\right)$$

where the factor $\gamma$ is an unknown proportionality factor that accommodates the linearity of aggregation of individual variances in the summation.

### 3.3.2 Inverse problem resolution

An inversion of the forward model of Equation 11 aims to provide estimates of the “proto-flux” $\hat{a}$ and noise strength $\hat{b}$, along with their associated posterior uncertainties, respectively $\hat{\sigma}_a$ and $\hat{\sigma}_b$ (where the “hat” symbol refers to estimated values). Full details of the inversion procedure are provided in Supporting Text S2. For brevity, only the key aspects are summarized below.

Importantly, Equation 12 entails that the standard deviation of observational uncertainties (error term $\epsilon_n$) depends on $r_n$, hence is non-constant, which precludes using Ordinary Least-Squares. This issue can be tackled by weighting observations proportionally to the inverse of their standard deviation, i.e. by $1/(\gamma \sigma_{CA} r_n)$ (e.g. see Sen & Srivastava, 2012, Chapter 6). As demonstrated in Supporting Text S2, Section S2.1, application of weights proportional to $1/r_n$ suffices to linearize the problem. As a result, a closed-form solution for $\hat{a}$ and $\hat{b}$ can be expressed using Weighted Least-Squares (Equation S15).
Estimating posterior uncertainties $\hat{\sigma}_a$ and $\hat{\sigma}_b$ requires a prior information on the pixel
noise $\sigma_{CA}$. However, since pixel noise is variable in time and space, using a hard-coded
(fixed) prior value for $\sigma_{CA}$ will yield poorly representative results for the posterior
uncertainties $\hat{\sigma}_a$ and $\hat{\sigma}_b$. Fortunately, as described in Supporting Text S2, Section S2.2, we
can estimate the "true" spatially-averaged pixel noise $\hat{\sigma}_{CA}$ directly from the quadratic
term of the regression $\hat{b}$, by reversing Equation 10. The estimated pixel noise
standard-deviation is expressed as:

$$\hat{\sigma}_{CA} = \frac{CA_{\text{min}}}{\sqrt{W_0 \left( \frac{\pi \kappa CA_{\text{min}}}{2} \right)^2}}$$

where $W_0$ is the first branch of the real-valued Lambert function (Equation S18). This
expression is evaluated after the inversion, and the resulting pixel noise $\hat{\sigma}_{CA}$ is used for
estimating realistic posterior uncertainties $\hat{\sigma}_a$ and $\hat{\sigma}_b$ (Equation S16).

Here, instead of the simple linear solution described above, we use a more robust inversion
procedure by further imposing prior bounds on $a$ and $b$ (which turns the problem into a
non-linear one), and by including an additional constant term $c$ in Equation 11
(intercept). Full details of the resolution of this generalized inverse problem are provided
in Supporting Text S2, Section S2.3, and the numerical stability of the inversion
procedure is demonstrated in Supporting Text S3.

### 3.3.3 Plume speed normalization

After inversion and estimation of posterior uncertainties, the final estimation of the mass
flux $\hat{m}$ (and its uncertainty $\hat{\sigma}_m$) is deduced from the "proto-flux" $\hat{a}$ (and its uncertainty
$\hat{a}$) using Equation 8, i.e. by a simple multiplication by plume speed $u$:

$$\begin{bmatrix} \hat{m} \\ \hat{\sigma}_m \end{bmatrix} = u \begin{bmatrix} \hat{a} \\ \hat{\sigma}_a \end{bmatrix}$$

This step is performed after the inversion, which makes it possible to adjust the plume
speed, without necessitating a second inversion. A custom plume speed can be chosen, or,
optionally in our implementation, the ECMWF ERA-5 wind fields can be queried
(Hersbach et al., 2020; Copernicus Climate Change Service Climate Data Store (CDS),
2023) to deduce the appropriate plume speed based on a choice of plume altitude.
3.3.4 Statistical test for automatic detection of volcanic degassing

The inversion provides an estimate of the posterior uncertainty $\hat{\sigma}_a$ on retrieved “proto-flux” $\hat{a}$, which allows for testing the statistical significance of a detection of volcanic degassing. The null hypothesis is stated as $H_0 : \hat{a} = 0$ (i.e. volcanic flux is insignificantly different from zero), whereas the alternative hypothesis is $H_1 : \hat{a} > 0$ (i.e. volcanic flux is significantly greater than zero). For a given confidence level (or probability $p$), testing the null hypothesis corresponds to evaluating the inequality:

$$\frac{\hat{a}}{\hat{\sigma}_a} > F^{-1}(p)$$

(15)

where $F^{-1}$ is the inverse of the cumulative density function of the standard normal (Gaussian) distribution, and $p \in [0.5 - 1.0]$ is the probability. If the inequality is satisfied, the null hypothesis is rejected, i.e. degassing is considered significant at the prescribed confidence level. As shown in Section 5.2, this statistical test allows for counterbalancing a temporary elevation of the noise level, for instance due to overpass by an external volcanic plume or anthropogenic SO$_2$-rich pollution, without raising a false positive.

3.4 Summary: inputs, outputs and internal parameters of the “disk method”

The “disk method” aims to estimate the volcanic SO$_2$ flux from the spatial distribution of SO$_2$ column amounts in a single TROPOMI image. The outputs of the method are (i) the SO$_2$ flux $\hat{\dot{m}}$, (ii) its uncertainty $\hat{\sigma}_{\dot{m}}$, and (iii) the characterization of noise in the image, in the form of a spatially-averaged pixel standard-deviation, $\hat{\sigma}_{CA}$.

The first step consists in calculating the mass of SO$_2$ integrated in a series of circular domains centered on a volcanic target (Equation 1). From this input dataset, a second-order polynomial regression is applied to the vector of masses (one mass per disk radius), as a function of disk radius (Equation 11).

Based upon a Gaussian plume model (Equation 6), the linear term of the regression is shown to represent a “proto-flux”, defined as a lumped quantity proportional to mass flux $\hat{\dot{m}}$ (Equation 8). This plume model depends on three atmospheric parameters (the wind speed $u$, the cross-wind diffusivity $D_y$ and the gas loss rate $k$) which are not retrieved by the inversion (unlike, e.g. Hyman et al., 2021). Indeed, the outputs are independent of the actual values of $k$, $D_y$ and $u$, so long as they remain within certain ranges of validity (see Section 4).
The quadratic term absorbs the contribution of noise in the image (Equation 10). Injecting noise into the regression corresponds to taking an opposite approach to previous methods aiming at reconstructing daily \( \text{SO}_2 \) emission rates, which all apply a relatively conservative truncation to the data prior to processing (typically, three times the standard deviation on background column amount noise, e.g., Theys et al., 2019). Instead, we purposely apply a permissive (low) truncation threshold \( CA_{\text{min}} \), which allows for lowering the overall level of detection on \( \text{SO}_2 \) flux. Furthermore, exploiting a bijective relationship with the noise standard deviation \( \hat{\sigma}_{CA} \) (Equation 13), this quadratic term is translated into a posterior uncertainty on the estimated \( \text{SO}_2 \) flux. This uncertainty (along with the estimated “proto-flux”) make it possible to devise a simple statistical test for automatically flagging positive detections of an \( \text{SO}_2 \) emission from the volcanic target (Equation 15). The maximum distance of integration \( r_{\text{max}} \) and the threshold \( CA_{\text{min}} \) are the two free input parameters of the method. Their effect on the regression is described in Section 4, and recommendations for setting them are provided in Section 6.

Finally, the determination of the mass flux requires multiplying the “proto-flux” by an estimate of the plume speed \( u \) (Equation 14). This last step is carried out a posteriori, which facilitates exploration of a range of wind speed scenarios.

4 Sensitivity analysis and detection threshold

4.1 Theoretical detection threshold without gas loss and noise

The regression model in Equation 11 ignores the effect of truncation on the apparent linear term \( (\hat{a}) \). Yet, since pixels with a low column amount are masked prior to integration, truncation inevitably leads to an underestimation of the integrated mass \( M(r_n) \), and with that, the retrieved \( \text{SO}_2 \) flux is expected to be affected too. As demonstrated in Supporting Text S4, in the simplifying case where \( k=0 \) (no gas loss) and \( \sigma_{CA}=0 \) (no noise), as long as the integration is limited to a maximum distance \( r_{\text{max}} \), it is possible to derive a closed-form expression for the fraction of mass flux that is underestimated by the inversion as a result of truncation (here, a fixed fraction of 25% is chosen). This expression can be reformulated to define a lower bound for the detectable \( \text{SO}_2 \) mass flux:

\[
\dot{m}_{\text{min}} = \frac{\kappa CA_{\text{min}}}{A} \sqrt{\frac{4\pi D_y}{r_{\text{max}} u}} \quad (16)
\]
Figure 3a displays the detection threshold $\dot{m}_{min}$ (x-axis) depending on the maximum distance $r_{max}$ (y-axis), for a scenario with $u = 10 \text{ m.s}^{-1}$ and $D_y = 10^4 \text{ m}^2 \text{s}^{-1}$, and for a range of cutoff values $CA_{min}$. A higher cutoff threshold $CA_{min}$ limits the possibility to detect low SO$_2$ fluxes, as gas concentration in the plume rapidly falls below the detection level beyond a certain distance $r_{max}$. For practical purposes, Figure 3a can be used as a reference chart to jointly adjust $r_{max}$ and $CA_{min}$ to a targeted detection threshold $\dot{m}_{min}$, given a certain wind speed $u$ and reasonable bounds on $D_y$.

4.2 Valid ranges of gas loss rate ($k$) and diffusivity ($D_y$)

Next, we conduct an experiment to assess the sensitivity of the method to other internal parameters of the forward model, starting with the diffusivity $D_y$ and gas loss rate $k$. We define a series of scenarios with variable levels of cutoff $CA_{min}$, ranging from 0.1 DU to 1.2 DU (Figure 3b). For each scenario, considering a fixed mass flux ($\dot{m} = 1 \text{ kton.day}^{-1}$) and a fixed wind speed ($u = 10 \text{ m.s}^{-1}$), we compute 2,000 simulated TROPOMI images, constructed from the superposition of a synthetic plume with random diffusivity $D_y$ and random gas loss rate $k$ (using Equation 6) and a synthetic noise with $\sigma_{CA} = 0.3 \text{ DU}$ (corresponding to a “moderate noise” case). After application of the threshold $CA_{min}$, we compute the spatial integration of these synthetic TROPOMI images for a series of circular domains, up to $r_{max} = 500 \text{ km}$. Finally, the synthetic data vector of integrated masses is fed into the inversion, and we compute the ratio $R$ between the reconstructed mass flux $\hat{\dot{m}}$ and the “true” SO$_2$ mass flux $\dot{m}_{true}$ (i.e. $R = \dot{m}/\dot{m}_{true}$). An exact reconstruction corresponds to $R = 1$, whereas $R = 0$ means a complete loss of sensitivity.

Figure 3b shows the domain of sensitivity of the inversion (defined as $R > 0.75$, i.e. a reconstructed mass flux no smaller than 75% of the “true” mass flux) as a function of $k$ (y-axis) and $D_y$ (x-axis). We observe that sensitivity is confined to a domain in the lower left quadrant of the graph, bounded by a maximum gas loss $k_{max}$ and a maximum diffusivity $D_{y,max}$ (respectively, upper and right limits of the lower left quadrant in Figure 3b). When $k$ or $D_y$ exceed these critical values, the reconstructed SO$_2$ flux substantially underestimates the “true” value (hatched area in Figure 3b). As expected from Section 4.1, the sensitivity improves when $CA_{min}$ is decreased.
We verify that the domain of sensitivity remains in the interior of the high Péclet number domain (i.e. \( u^2 \gg D_x k \), assuming that \( D_x \approx D_y \) for simplicity, double hatched area in the upper right corner of Figure 3b), consistent with the “slender plume approximation”.

In terms of maximum gas loss, the inversion performs well up to a maximum \( k_{\text{max}} \approx 2-5 \times 10^{-5} \, \text{s}^{-1} \), equivalent to an e-folding time of \( \tau = 6-14 \) hours (Figure 3b).

This bound materializes the limit of validity of the linear approximation of Equation 8: a high gas loss leads to an underestimation of the \( \text{SO}_2 \) flux by the inversion. The limit appears to be well approximated by the inverse of the characteristic time \( T \) defined in Section 3.1.2 (i.e. \( k = 1/T = u/r \), setting \( r \) to the maximum radius \( r_{\text{max}} = 500 \) km used in the synthetic tests). Two factors likely contribute to stabilizing the linear term near \( r \sim u/k \): (i) weighting by \( 1/r_n \) (Section 3.3.2), which counterbalances the influence of data points at large \( r \), which are most affected by the exponential gas loss, and (ii) the beneficial side effect introduced by the quadratic term, which probably absorbs a fraction of the bias generated by the exponential decay. Thereafter, the criterion \( r_{\text{max}} \lesssim u/k_{\text{max}} \) will be retained to define the maximum distance that may be used for the input dataset in the inversion (horizontal dotted lines in Figure S2).

The maximum diffusivity \( D_{y,\text{max}} \) is consistent with the value obtained for \( D_y \) from Equation 16, replacing \( u, r_{\text{max}}, \dot{m} \) and \( CA_{\text{min}} \) by the values used in the synthetic tests (vertical dashed lines in Figure S2). This observation confirms that the theoretical bound defined in Equation 16 can be effectively used to predict the maximum diffusivity allowed by the “disk method”.

### 4.3 Influence of pixel noise (\( \sigma_{CA} \)) and wind speed (\( u \))

Using the same approach as in Section 4.2, we now assess the sensitivity of the method in three cases considered representative of three noise scenarios (Figure 3c). Each scenario is empirically defined by a single pair of values for pixel noise \( \sigma_{CA} \) and cutoff threshold \( CA_{\text{min}} \): (i) “low noise scenario”: \( (\sigma_{CA}, CA_{\text{min}}) = (0.1, 0.2) \) DU; (ii) “moderate noise scenario”: \( (\sigma_{CA}, CA_{\text{min}}) = (0.3, 0.9) \) DU; (iii) “high noise scenario”: \( (\sigma_{CA}, CA_{\text{min}}) = (1.0, 3.0) \) DU. Contrary to the previous exploration where mass flux was held fixed and diffusivity was variable, here, we compute synthetic plumes with random mass fluxes \( \dot{m} \) and a fixed diffusivity \( D_y = 10^4 \) m².s⁻¹ (a value representative of tropospheric plumes, see Section 3.1). The gas loss rate \( k \) remains random.
In Figure 3c, the domain of stability is displayed as a function of the mass flux \( \dot{m} \) (x-axis) and gas loss rate \( k \) (y-axis). The boundary of the sensitivity domain for \( \dot{m} \) (left limit of the lower right quadrant) allows for defining a detection threshold, or minimum detectable SO\(_2\) mass flux. The exploration shows that the detection threshold increases from \( \approx 0.5 \) to \( \approx 5 \) kton.day\(^{-1}\) from low to high noise scenario.

Decreasing the distance of integration \( r_{max} \) improves the sensitivity (upper limit of the sensitivity domain). However, reducing \( r_{max} \) has a negative side effect on the ability of the inversion to correctly estimate the intensity of noise (not shown in Figure 3c, see Supporting Figure S5). We also note that decreasing the integration distance \( r_{max} \) improves the detection level for high gas loss scenarios \( (k) \). Nevertheless, the improvement is marginal, and in fact, wind speed \( u \) has a dominant effect.

In Figure 3d, we explore the primary effect exerted by wind speed \( u \), holding all other parameters fixed according to the “low noise / short distance” scenario defined in the previous exploration \((\sigma_{CA}, CA_{min}) = (0.1, 0.2)\) DU and \( r_{max} = 250 \) km). We observe that a low wind speed improves the detection threshold due to an overall increase of gas concentration, as gas accumulates near the source, enhancing the sensitivity to weak fluxes (left limit of sensitivity domain bounded by the dashed yellow curve in Figure 3d). Nevertheless, a low wind speed also generates an adverse effect: as plume age increases at any given distance, a higher proportion of gas is degraded and lost in the area of integration. This leads to an underestimation of the SO\(_2\) flux (upper limit of sensitivity domain in Figure 3d). Conversely, the effects are reversed for a high wind speed (dashed blue curve in Figure 3d): detection capability is slightly weakened (gas concentration is everywhere lower), but the inversion is much more tolerant to a high gas loss rate (gas parcels are “younger” at any distance). In summary, a low wind speed leads to a substantial underestimation of the SO\(_2\) flux when gas loss rate is high.

5 Results: application to volcanic case-studies

5.1 Etna (January – December 2021)

5.1.1 Volcanic context

In order to investigate the capacity of the method to retrieve SO\(_2\) emissions over long time-intervals, we analyze TROPOMI SO\(_2\) data acquired at Etna volcano (elevation: 3,350 m asl) over a one-year-long period (1 Jan 2021 – 31 Dec 2021). Etna’s volcanic
activity is characterized by a broad spectrum of eruption types and degassing activity, alternating between passive outgassing, effusive eruptions, and occasional Strombolian explosions (Giuffrida et al., 2023).

Etna is equipped with permanent ground monitoring networks of UV-DOAS instruments installed on the flanks of the volcano (FLAME network, Salerno et al., 2009), and with short-range UV cameras near the summit (Delle Donne et al., 2019). However, these data may be affected by temporal gaps and uncertainty due to changing atmospheric conditions, or the presence of ash (Boichu et al., 2015). The annually-averaged daily SO$_2$ mass flux has been estimated from satellite data to 2–3 kton.day$^{-1}$ during passive degassing phases (Carn et al., 2016, 2017; Coppola et al., 2019; Fioletov et al., 2023), increasing to $\sim$ 4 kton.day$^{-1}$ during effusive phases (Coppola et al., 2019; Queißer et al., 2019), with individual paroxysmal events typically releasing 5–20 kton SO$_2$ over time intervals of 3–12 hours (Boichu et al., 2015; Corradini et al., 2020; Sellitto et al., 2023).

Etna’s activity has been particularly intense in 2021, fueled by two episodes of mafic recharge in late 2020 and mid-2021 (Giuffrida et al., 2023). A sequence of 62 intense explosions originating from the South East Crater, associated with lava fountains lasting from a few hours to a couple of days, are concentrated in two paroxysmal sequences (Aiuppa et al., 2015), which are mostly covered by our dataset: (a) between 13 December 2020 and 2 April 2021 and (b) between 19 May and 23 October 2021 (Figure 4).

5.1.2 Daily SO$_2$ flux for the year 2021

In a first analysis, the TROPOMI 7 km altitude product is used, removing 22 rows to reduce the impact of noise from track edges. The cutoff threshold is set to $CA_{\text{min}} = 0.0$ DU, and the maximum distance of integration to $r_{\text{max}} = 1000$ km. Cumulative masses are calculated for radii 25, 50, 75, 100, 150, 200, 250, 300, 400, 500 and 1000 km, and provided as input data for the inversion. We discard acquisitions with a mean cloud fraction $> 75\%$ within 200 km of the volcano (removing 35 dates out of 365). After inversion, plume speed is assumed equal to wind speed from the ERA-5 ECMWF product at a pressure level of 600 hPa (equivalent to an altitude of $\approx 4.2$ km), which provides the best coherence between observed plume direction in the image and predicted wind direction over this one-year-long interval (Figure S6a).
Figure 4a shows the SO$_2$ daily flux estimated by the “disk method” for a 1-year-long time interval spanning the year 2021. Estimated SO$_2$ emission rates are highly variable in time, with isolated bursts exceeding 10 kton.day$^{-1}$, mainly clustered in February-March, May–July and October 2021 (Figure 4a). The largest SO$_2$ peaks reach $\sim 20$ kton.day$^{-1}$, which is comparable in magnitude with (yet larger than) values estimated from ground observations (10–15 kton.day$^{-1}$, according to Aiuppa et al., 2023). These periods of intense degassing alternate with weeks-long intervals of lower emission rates, below 1 kton.day$^{-1}$, especially in April, October and November 2021. The “background” emission rate in these relatively quiet time intervals is estimated to 0.3–0.8 kton.day$^{-1}$ from TROPOMI, commensurate with the 1 kton.day$^{-1}$ reported by Aiuppa et al. (2023).

By integrating daily flux estimates over the full length of the 2021 time-series, we estimate a cumulative SO$_2$ mass of 443 kton for the year 2021 using the 7 km product. An alternative estimate obtained by performing a linear interpolation at 4.5 km (assumed plume height) between fluxes computed using the 1 km and 7 km products (analyzed independently with identical settings, both with CA$_{\text{min}}$=0.0 DU) yields a total mass of 915 kton (Figure S7). These values are reasonably consistent with the total annual emission budget of 600 kton for the same year, as reported by Fioletov et al. (2023, 2022) using an independent method for analysing TROPOMI data (keeping in mind that Fioletov et al. (2023, 2022) excluded days with large SO$_2$ mass burdens). We note that both the pixel noise and the flux estimated over Etna for 2021 from the TROPOMI 1 km product are 2–4 times larger than that from the 7 km product (Figure S7).

5.1.3 Comparison of degassing with RSAM

Further comparison with a ground-based dataset acquired at higher temporal resolution, such as seismicity, provides insights on the ability of satellite-based observations to capture temporal variations of volcanic activity. In Figure 4, the 1-year-long time-series of estimated SO$_2$ flux is compared to the seismic energy (RSAM) recorded continuously at seismic station ESLN, situated 4 km south of Etna’s summit (Figure 4b). The temporal shift that needs to be applied to the RSAM data (Section 2.2) is estimated to $\sim 8$ hours, considering a typical length of the plume of $\sim 500$ km and a mean wind speed of $\sim 15$ m.s$^{-1}$ at 4 km asl.
Bursts of large gas emissions (flux > 7 kton.day\(^{-1}\)) all coincide with peaks of seismic energy (Figure 4b). On the other hand, periods of reduced degassing (April–May, October and December 2021) consistently match with seismically quiescent time intervals. Figure 4c shows a zoom spanning the May–August 2021 paroxysmal sequence, where volcanic activity at Etna was characterized by the occurrence of > 20 lava fountain events, lasting between 2 hours and > 24 hours, with a recurrence interval ranging from a few days to less than 24 hours on 21–27 June 2021 (INGV, 2021b). The day-to-day pattern of seismic energy release variations during this period is closely reproduced in the TROPOMI-derived SO\(_2\) flux history (Figure 4c).

Quantitative comparison of the SO\(_2\) emissions and seismic energy (RSAM) demonstrates a reasonable correlation between the two observables. A power-law fit indicates that the ground velocity is proportional to \(\dot{m}^{\beta}\), with \(\beta = 0.7 - 1.3\) (depending on the points selected, see Figure 4d). This relation is consistent with the near-proportionality between seismic energy and magma discharge rate reported in previous studies (e.g. see Ichihara, 2016, and references therein). Nevertheless, rapid intra-day fluctuations of volcanic activity can be aliased or even missed by our analysis, since we estimate a flux averaged over the time interval between gas emission and satellite acquisition. In addition, our assumption of a steady flux and constant emission height (here, 4.2 km asl) may be overly simplistic for the description of energetic events. For example, on 19 February 2021, the eruption lasted less than 3 hours, and the plume rose up to 10 km (INGV, 2021a; Global Volcanism Program, 2021). Detailed estimates of SO\(_2\) flux for such short-lived events would require a case-by-case analysis.

5.2 Piton de la Fournaise (September 2021 – September 2023)

5.2.1 Volcanic context

In order to assess the capability of the method to constrain smaller emission rates, we now turn to Piton de la Fournaise (elevation: 2,600 m asl), one of the most active volcanoes in the world, producing an average of 2–3 eruptions per year for the past 30 years (Roult et al., 2012; Dumont et al., 2022). Eruptions are generally preceded by a months- to days-long period of pre-eruptive seismicity and inflation sourced from a reservoir ∼ 2 km below the summit (Peltier et al., 2009). Vertical migration of seismicity and deformation over time scales of days to hours mark the ascent of magma toward the
surface (Roult et al., 2012; Smittarello et al., 2019). In the few hundred meters below the summit, the direction of magma migration often shifts, either toward the southern or northern rift zones, eventually feeding an effusive eruption that may last from a few hours to several weeks (Dumont et al., 2022; Journeau et al., 2023). More rarely, explosive eruptions, caldera collapse and lateral flank motion can be triggered. At the time of writing, the last such event occurred in 2007 (Michon et al., 2013).

Contrary to Etna, the SO$_2$ budget of Piton de la Fournaise is modest. SO$_2$ emissions are monitored by a network of three ground-based UV-DOAS instruments (NOVAC network). However, these measurements often substantially underestimate the SO$_2$ budget of the volcano, due to unfavorable atmospheric and geometric conditions (Arellano et al., 2021; Verdurme et al., 2022). Satellite observations of syn-eruptive SO$_2$ emissions of Piton de la Fournaise have also been analyzed (e.g. Khokhar et al., 2005; Carn et al., 2016; Verdurme et al., 2022). A SO$_2$ mass of 230 kton was released during the reservoir collapse of 2007 (Tulet & Villeneuve, 2011), but smaller eruptions generally release 10–35 kton of SO$_2$ (Carn et al., 2016; Verdurme et al., 2022; Hayer et al., 2023), consistent with the release of $<$ 20–30 Mm$^3$ of bulk lava reported from field and satellite observations (Roult et al., 2012; Verdurme et al., 2022). To date, no inter-eruptive satellite detection of SO$_2$ has been reported.

5.2.2 Comparison of SO$_2$ flux and RSAM for three eruptions

Here, we analyze the three latest eruptions of Piton de la Fournaise (at the time of writing): December 2021 – January 2022 (Figure 5a), September – October 2022 (Figure 5b) and July – August 2023 (Figure 5c). The three eruptions have similar duration (several weeks), style (effusive) and volume of extruded lava ($\sim$ 10 Mm$^3$).

To retrieve SO$_2$ fluxes from TROPOMI, we use the 7 km altitude product, integrated up to a maximum distance of 500 km, as the plume rarely extends beyond this distance. To prevent gaps due to increased spacing between TROPOMI tracks at low latitude, we only mask 7 swath-edge rows. As a result, the SO$_2$ column amount maps include more noise from swath-edge rows than at Etna, and the progressive longitudinal drift of the swath during Sentinel-5P’s orbital cycle generates periodic modulations of the ambient noise. Finally, we assume that plume speed is equal to wind speed at the ERA-5 700 hPa
pressure level (≈ 3 km). This altitude best matches with the plume transport direction visible in syn-eruptive TROPOMI images (Figure S6b).

Comparison between SO$_2$ and RSAM is displayed in Figure 5. For RSAM, we use data from three seismic stations selected for their short distance from the active vents of each eruption (respectively, FOR, RVA and PVD). We decrease the time lag for the seismic-to-satellite synchronization down to 5 hours, because volcanic plumes are typically shorter at Piton de la Fournaise than at Etna.

The September – October 2022 eruption (Figure 5b) is characterized by an initial pulse of SO$_2$ on 20 September reaching 3 kton.day$^{-1}$, followed by a week-long period of weaker emissions at 1 kton.day$^{-1}$. The last 5 days of the eruption are marked by an increase of degassing, reaching a maximum of 5 kton.day$^{-1}$ on the last day of the eruption. This progressive increase in SO$_2$ flux coincides with a coeval rise in seismic energy, until both signals drop abruptly on 5 October 2022, when the eruption ceases. The same pattern is also apparent in time-averaged discharge rates reported independently by the MIROVA and HOTVOLC services using MODIS, VIIRS and MSG-SEVIRI data (see Figure S9, adapted from Chevrel et al., 2023). This eruption occurred during a period of exceptionally dry weather, with a cloud cover < 25% for most of the eruption (blue symbols in Figure 5b), October 2022 being the driest October since the first measurements at La Réunion in 1972 (Météo-France, 2022b). This favorable situation facilitates the agreement between RSAM and satellite-based estimations of emission rates.

The July – August 2023 eruption (Figure 5c), in spite of a longer duration (38 days), displays a similarly consistent evolution between degassing and seismicity. The eruption started on 2 July 2023 with a one-week-long phase of intense seismic energy release, followed by a temporary lull, and a resumption of activity on 8 July 2023. After that, a continuous exponential-like decay is observed until the eruption end one month later. During the decay phase, in spite of the low SO$_2$ fluxes involved (less than 0.6 kton.day$^{-1}$), the “disk method” consistently tracks the progressive decrease of SO$_2$ emission rate, and successfully detects surges coinciding with temporary increases in RSAM on 26–27 July and 8 August (last day of the eruption). Remarkably, during this decay phase, in spite of substantial day-to-day fluctuations of wind speed (between 1 m.s$^{-1}$ and 10 m.s$^{-1}$, green symbols at the bottom of Figure 5c), the estimated SO$_2$ flux remains relatively stable. This suggests that the method correctly compensates for the dilution (respectively, the...
accumulation) of SO$_2$ resulting from an increase (respectively, a decrease) of transport speed.

Remarkably, in addition to smooth fluctuations of activity, the two eruptions of September–October 2022 and July–August 2023 are both characterized by an initial large pulse of degassing at the onset of the eruption, greater than in the following days by a factor $\sim 3$. Simultaneously, a spike of seismic energy is detected. These observations may be interpreted as “uncorking” events, where a pressurized batch of gas is suddenly released when the dike connects to the surface.

In contrast with the two 2022 and 2023 eruptions, comparison between SO$_2$ emissions and RSAM during the December 2021 – January 2022 eruption (Figure 5a) is not straightforward. The RSAM displays a progressive increase of seismic energy over the full duration of the eruption, punctuated by quasi-periodic fluctuations in the first two weeks. These two features (progressive rise and fluctuations) are not visible in the SO$_2$ flux.

At least part of this apparent disagreement may be attributed to meteorological conditions affecting the quality of SO$_2$ measurements. A clear and systematic decrease in the apparent SO$_2$ flux is observed when the cloud fraction is high (blue symbols at the bottom of Figure 5a). In fact, windy and cloudy weather was reported during most of the December 2021 – January 2022 eruption, including an exceptionally intense rain episode on 22–23 December 2021, and stormy rains on 8–15 January 2022 (Météo-France, 2021, 2022a). On the other hand, during this eruption, variations in RSAM have probably been influenced by small-scale processes taking place around the vent, such as phases of cone construction and collapse, as well as channelling of lava into lava tunnels or cone overflow (as described in the eruption report of Observatoire volcanologique du Piton de la Fournaise, 2022). These surface processes modulate the relationship between the lava and gas discharge rate and the amplitude of seismic tremor (e.g. Battaglia et al., 2005; Journeau et al., 2023), hence complicating direct comparison. We acknowledge that both factors (cloud cover and small-scale processes at the vent) are not mutually exclusive. A systematic analysis of day-to-day observations would be necessary to quantify the influence of these different factors.

Eventually, by summing daily-averaged SO$_2$ fluxes over the duration of each eruption, the total SO$_2$ mass budget can be estimated. The three eruptions released 19.1 kton SO$_2$ (December 2021 – January 2022 eruption), 23.1 kton SO$_2$ (September – October 2022
eruption) and 17.7 kton SO$_2$ (July – August 2023 eruption), with a $\sim 15\%$ relative 1-$\sigma$
formal uncertainty. These estimates are increased by a factor $\sim 3$ when the 1 km altitude
product is used, instead of the 7 km product (Figure S8). Due to the cloudy conditions
prevailing at that time, estimates for the December 2021 – January 2022 eruption likely
represent an underestimation of the SO$_2$ budget, perhaps by a factor of two or more.

5.2.3 Automatic detection of degassing for a 2-year-long time-series

In order to assess the stability of the results and the capability of the statistical test of
Equation 15 to provide reliable detections of volcanic degassing, we analyze a complete,
two-year-long time series of TROPOMI data over Piton de la Fournaise (Figure 6). The
period includes the three aforementioned eruptions, and is analyzed using the same
parameters (7 km product; maximum distance 500 km; $CA_{\text{min}} = 0.0$ DU; masking 7
swath-edge rows). With a probability threshold fixed to $p = 99\%$, the statistical test of
Equation 15 successfully detects all three eruptions, without any false positives
(Figure 6a). We note that the detection is not directly related to the retrieved value of
the SO$_2$ flux, nor to the mass at any single distance from the volcano (masses are plotted
for $r_n = 25$, 150 and 500 km in Figure 6e). For instance, positive detections are reported
for the January 2022 eruption with emission rates as low as 0.4 kton.day$^{-1}$ (2 January
2022), whereas SO$_2$ rates during non-eruptive periods often exceed this value, but do not
lead to any false positives.

Immunity to false positives depends on the robustness of uncertainty estimation. Indeed,
in the two repose intervals separating the three eruptions, the mean background SO$_2$ flux
is 0.04 kton.day$^{-1}$, whereas the mean 1-$\sigma$ uncertainty is 0.36 kton.day$^{-1}$, i.e. an order of
magnitude larger. Since the criterion of Equation 15 relies on the ratio between these two
quantities, it remains consistently negative throughout non-eruptive intervals.

A further illustration of the adaptability of the method is provided by the fortuitous
overpass by the plume of the Hunga Tonga–Hunga Ha’apai (HTHH) eruption from 18 to
30 January 2022 (Boichu et al., 2023) (see Figure 7c). At the time it reaches la Réunion,
the HTHH plume is diluted, producing a homogeneous non-zero-mean distribution of the
SO$_2$ column amount in the image (Figure 7c1), and a well-marked quadratic component
in the mass-to-distance distribution (Figure 7c2). The inversion interprets this pattern as
resulting from a higher value of the background noise (up to $\hat{\sigma}_{CA} = 1.7$ DU), explaining
the sharp increase of posterior uncertainty, reaching an average 0.96 kton\cdot day^{-1} during
the overpass (time interval highlighted in magenta in Figure 6b).

In the same vein, periodic fluctuations of pixel noise $\hat{\sigma}_{CA}$ (peak-to-peak, 0.1–0.3 DU,
Figure 6b) are caused by regular introduction of noisy swath-edge rows in the area of
interest (noisy stripes in Figure 7a and 7d). Incorporation of these swath-edge rows is a
necessary tradeoff to avoid data gaps at low latitudes. The periodicity of $\hat{\sigma}_{CA}$ results from
the progressive drift of Sentinel-5P ground tracks. Even if their distribution is not
homogeneous across the image, these noisy observations increase the quadratic term, and
the inversion responds by increasing the posterior uncertainty (Figure 7d2). The level of
detection is thus momentarily degraded, but it remains possible to analyze moderate to
strong degassing patterns that are well above the swath-edge noise (such as the plume
displayed in Figure 7a). This strategy avoids repeated interruptions of the time-series,
which is valuable for continuously tracking volcanic emissions at low latitudes.

6 Discussion

6.1 Limitations and recommended usage

The “disk method” introduced in this paper relies on a “slender plume approximation” of
the atmospheric advection-diffusion equation. The approximation requires that advection
(via transport speed $u$) dominates over along-plume diffusion ($D_x$). Recasting the
“slender plume approximation” in terms of the Péclet number (i.e. $P_e = u^2 / D_y k \gg 1$,
Section 3.1) places an upper bound on the product $kD_x$, hence on $kD_y$ (assuming
$D_x \approx D_y$, a common simplification made in numerical models of volcanic plume
dispersion, e.g. Barsotti et al., 2008; Folch et al., 2009). The extent of the $P_e \gg 1$ domain
(or, equivalently, $kD_y \ll u^2$), as a function of $k$ and $D_y$ is represented in Figure S1 for a
range of wind speeds.

In summary, according to the “slender plume approximation”, either $k$ and $D_y$ should
remain “small”, or the plume speed $u$ should be “large”. Recognizing that $k$ and $D_y$ may
be poorly constrained in practice, we here provide general recommendations to adjust the
free parameters of the method ($CA_{min}$ and $r_{max}$) so as to remain within the domain of
validity of the assumptions. The choice of the cutoff threshold $CA_{min}$ and maximum
integration distance $r_{max}$ is here determined as a compromise between (i) detection
threshold for low fluxes and (ii) plausibility of posterior uncertainties.
• **Recommendations for cutoff threshold (**$CA_{\text{min}}$**)**

In a series of sensitivity experiments on synthetic data (Section 4.2), we observed that, for a given diffusivity $D_y$ and gas loss rate $k$, increasing the cutoff threshold $CA_{\text{min}}$ leads to a progressive underestimation of the gas flux (Fig 3b).

Tests on real data at Etna (Fig S10) confirm that increasing the cutoff $CA_{\text{min}}$ systematically leads to a decrease in the estimated SO$_2$ fluxes, primarily for the lower fluxes that prevail during inter-eruptive periods ($< 1$ kton.day$^{-1}$), which essentially drop to zero when $CA_{\text{min}} > 1.0$ DU. Conversely, high fluxes ($> 10$ kton.day$^{-1}$) remain stable up to $CA_{\text{min}} = 1.4$ DU. However, in turn, since the quadratic term becomes negligible, uncertainties become unacceptably small (down to $\hat{\sigma}_{CA} = 0.03$ kton.day$^{-1}$, against a more realistic $\hat{\sigma}_{CA} = 0.9$ kton.day$^{-1}$ for $CA_{\text{min}} = 0.0$ DU).

We do not recommend using an excessively high $CA_{\text{min}}$ in the “disk method”. Instead, it is preferable to keep the cutoff threshold $CA_{\text{min}}$ to a relatively low value, of the order of the noise level $\sigma_{CA}$ or even lower (i.e. $CA_{\text{min}} \lesssim \sigma_{CA}$). A low cutoff $CA_{\text{min}}$ allows for improving the detection level in presence of moderate to strong gas loss or diffusivity. However, $\sigma_{CA}$ is not known a priori, such that currently $CA_{\text{min}}$ needs to be defined by trial-and-error. Future work may focus on identification of representative values for $\sigma_{CA}$ (hence $CA_{\text{min}}$) depending on the setting, latitude or season. Another strategy could be to exploit ancillary information available in the TROPOMI product.

• **Recommendations for maximum integration distance (**$r_{\text{max}}$**)**

In previous synthetic explorations of the effect of noise $\sigma_{CA}$ and wind speed $u$ (Section 4.3), we showed that decreasing the maximum distance of integration $r_{\text{max}}$ makes the inversion moderately less vulnerable to gas loss for a fixed wind speed (Figure 3c), but that this benefit is largely overshadowed by the contribution of wind itself (Figure 3d).

However, tests conducted on real data at Etna show that reducing $r_{\text{max}}$ makes the estimation of the spatially-averaged noise $\hat{\sigma}_{CA}$ less reliable (Fig S11) and likewise of all posterior uncertainties that depend on it. Decreasing the maximum distance $r_{\text{max}}$ leads to a systematic decrease of estimated fluxes for the largest emission peaks (paroxysmal events) and a dramatic increase in the estimated uncertainties (from $\hat{\sigma}_{CA} = 0.9$ kton.day$^{-1}$ for $r_{\text{max}} = 1000$ km, increasing to
\( \dot{\sigma}_{CA} = 2.4 \text{ kton.day}^{-1} \) for \( r_{max} = 200 \text{ km} \), and up to \( \dot{\sigma}_{CA} = 5.6 \text{ kton.day}^{-1} \) for \( r_{max} = 100 \text{ km} \).

In summary, following the criterion defined in Section 4.2, the maximum integration distance \( r_{max} \) should be as large as possible, as long as it satisfies the condition \( r_{max} \lesssim u/k \) (i.e. plume age at \( r_{max} \) should be no older than \( T = 1/k \)).

Unfortunately, \( k \) is generally unknown, but in practice, it can be roughly estimated from the ratio between mean wind speed \( u \) and the length of a typical plume \( L_{plume} \) (i.e. \( k \approx u/L_{plume} \)). This condition is actually equivalent to setting \( r_{max} \) to the length of a typical plume (i.e. \( r_{max} \lesssim L_{plume} \)), which constitutes a simple rule of thumb.

6.2 Incorporation of information on plume altitude

Thanks to its simplicity, the “disk method” can be efficiently and automatically applied to long time-series. In this study, we assumed a constant plume altitude over long time intervals (1 year at Etna, 2 years at Piton de la Fournaise). Here, the “best” altitude was determined by assessing, \textit{a posteriori}, the agreement between the direction of plume of transport observed in the TROPOMI image (estimated crudely by calculating the coordinates of the center-of-mass of the plume with respect to the source) \textit{versus} the direction of wind predicted by ERA-5. (Figure S6). An example of the agreement between observed and predicted wind direction is shown in Figure 7a and 7b. We note that this consistency only represents a necessary condition, but that it is not sufficient to guarantee that the selected altitude, and thus speed, is correct. Indeed, in the presence of an along-plume vertical wind speed gradient (\( \partial u/\partial z \)), wind speed may change in the atmospheric column independently of wind direction.

Alternatively, plume altitude could be deduced directly by incorporating information on SO\(_2\) height estimated by advanced retrieval algorithms. This information is present in the standard TROPOMI L2 product (Hedelt et al., 2019), but is restricted to large column amounts (greater than 20 DU). The more sensitive COBRA product proposed recently by Theys et al. (2022) could also provide estimates for lower concentrations, down to 5 DU. Other algorithms exist for the IASI and CrIS infrared sensors (Clarisse et al., 2014; Carboni et al., 2016; Hyman & Pavolonis, 2020), and could be used in synergy. Ability to easily display co-located data from a variety of satellite products, as in the VolcPlume Platform (Boichu & Mathurin, 2022), eases this task.
We also simplified the analysis by incorporating the TROPOMI SO\(_2\) column amount retrieved with an assumption of a plume center-of-mass at 7 km altitude. This choice is in evident contradiction with the selected ERA-5 altitude at Piton de la Fournaise (3 km) and Etna (4.2 km). Alternatively, it is possible to interpolate between SO\(_2\) column amounts retrieved at two different altitudes (e.g. Carn et al., 2013; Theys et al., 2019). Following this logic, we applied a linear interpolation between the flux time-series estimated from the 7 km and 1 km products, which differ by a factor \(\sim 2-4\) (see Figure S7 for Etna and Figure S8 for Piton de la Fournaise). In more complex situations, plume altitude may substantially vary over time, chiefly as a result of variations in the SO\(_2\) flux (see Section 1). In such situations, it would be straightforward to simultaneously adapt the weights of the interpolation, enforcing an on-the-fly consistency with the altitude used in the plume speed normalization, without necessitating further inversion runs. The implementation of the method in an interactive platform (Boichu & Mathurin, 2022) also facilitates manual exploration of the range of plausible altitudes, wind speeds and fluxes, which is convenient for a near-real time analysis.

Several assumptions however limit the generality of the “disk method”. The main limitation is the assumption of a simple Gaussian plume, steadily spreading from the source at a constant altitude. In reality, temporal variations in emission strength (and thus, of injection height), combined with variability of wind vectors with altitude and time, often lead to more complex plume shapes. In such situations, the plume may be split in distinct parts (e.g. see Figure 5 of Boichu et al., 2015, at Etna), spread or stagnate close to the source (e.g. see Figure 2 of Behera et al., 2023, at Ambrym), or even be entrained back towards the source due to vorticity of atmospheric transport (e.g. see Figure 2 of Boichu et al., 2014, at Eyjafjallajökull). This is often the case for short-lived, intense periods of degassing, such as syn-eruptive, paroxysmal emissions, where rapid variations in mass flux and altitude often take place. In these situations, it remains possible to restrict the analysis to a short range from the source, where complexity is usually less prevalent (as illustrated in the inset of Figure 7a2). Accordingly, this strategy restricts the inversion to the few hours preceding the satellite acquisition. For more complex cases, it is recommended to apply a more advanced inversion method capable of reconstructing of temporal variations of both SO\(_2\) emission rate and altitude, such as back-trajectory analysis (e.g. Esse et al., 2024) or inverse modeling (e.g. Boichu et al., 2015).
6.3 Lessons learned from application to real cases

The “disk method” has been successfully applied for the estimation of the SO$_2$ flux released by two volcanoes exhibiting contrasting styles of volcanic activity. At Piton de la Fournaise, the method measures relatively weak fluxes (often $<2$ kton.day$^{-1}$) during three effusive eruptions lasting between 16 and 38 days. Daily fluctuations as low as 0.5 kton.day$^{-1}$ are captured during the July–August 2023 eruption (Figure 5c).

Accordingly, these low values cannot be directly generalized into a detection threshold, which largely depends on the level of noise. The actual detection level is expected to be higher in more noisy environments, such as at high latitude (e.g. at Bezymianny, see Supporting Text S1). Nevertheless, the capability of the method to quantify a spatially-averaged pixel noise, without any a priori, allows for mapping these uncertainties into realistic error bars on the posterior SO$_2$ flux. Future efforts could be directed towards a validation of these posterior uncertainties against the precision of column amounts reported in the TROPOMI files. Furthermore, we anticipate that applying the “disk method” to the recently released SO$_2$ COBRA TROPOMI products will further improve the quality of the results, both in terms of flux and estimated noise (Theys et al., 2021; Fioletov et al., 2023).

The analysis of a two-year-long time series at Piton de la Fournaise also illustrates how the estimation of pixel noise from the “disk method” may be an asset for robustly and automatically detecting degassing events from a target. The three eruptions of 2021–2023 are detected with no false positives, in spite of being associated with low eruptive fluxes. On the other hand, the overpass by the stratospheric Hunga Tonga plume is translated into a temporary increase of the “apparent” background noise, and does not lead to a false increase of the estimated SO$_2$ flux from Piton de la Fournaise (Section 5.2.3). We also show that the inclusion of noisy swath-edge rows in the data, which is mandatory for providing daily observations without gaps at low latitude, does not substantially impair the results.

Unsurprisingly, by carefully inspecting the cloud fraction and SO$_2$ flux, we observe that a strong cloud cover leads to an apparent decrease in the SO$_2$ flux, since low-altitude SO$_2$ is masked by meteorological clouds. The origin of this bias is traced back to the TROPOMI data, and no simple correction can be applied in post-processing to counter this effect. A pragmatic mitigation strategy may involve discarding estimations for days affected by a
substantial cloud cover (say, > 50%). Fixing a universal threshold for the maximum tolerated cloud cover is not straightforward, as the reliability of the retrieval depends on the signal-to-noise-ratio of the data. Thus, a trial-and-error, case-by-case approach should be preferred. The interactivity offered by the VolcPlume Platform, which provides access to meteorological cloud properties (Boichu & Mathurin, 2022), facilitates this strategy.

More broadly, our analysis outlines a general strategy to leverage the potential of satellite data for the benefit of volcano observatories. Here, we find a reasonable correlation between seismic energy and SO$_2$ flux, both during short-lived eruptions of Piton de la Fournaise and over longer cycles of paroxysmal sequences at Etna. Systematically comparing SO$_2$ fluxes and seismic energy is an efficient approach to detect changes in eruption dynamics, while simultaneously allowing for a diagnosis of caveats that may affect remote sensing products.

7 Conclusion

We introduce the “disk method”, a novel method to calculate daily volcanic SO$_2$ flux from TROPOMI imagery. Based on a Gaussian plume model in the “slender plume approximation”, a SO$_2$ “proto-flux” is estimated by a linear regression (as a function of distance) of SO$_2$ mass integrated in a series of nested circular domains centered on the volcano. Circular integration implies an invariance with respect to the direction of plume transport.

A salient feature of the “disk method” is its ability to jointly quantify the spatially-averaged noise intensity in a satellite image. This allows for deriving robust posterior uncertainties on the SO$_2$ flux and improving the detection level. To do so, we develop a noise model, considering pixel column amounts as random variables that follow a “truncated normal distribution”. We demonstrate that the noise intensity can be estimated from an additional quadratic term in the regression. The noise model is validated experimentally on two datasets affected by noise only. The domain of stability of the inversion with respect to internal atmospheric parameters (gas loss rate, cross-plume diffusivity and wind speed) is constrained from theoretical calculations and sensitivity tests with synthetic and real data.

After completion of the inversion, which is the most computationally-demanding step, information on plume speed can be incorporated by a simple multiplication of wind speed...
with the “proto-flux”, to deduce the SO$_2$ flux. The simplicity of this final step makes it straightforward to explore \textit{a posteriori} a range of plume speed scenarios. This way, uncertainty on plume altitude can be efficiently propagated into a range of possible SO$_2$ fluxes, which represents an additional advantage of the method.

When plume speed is unknown, it can be deduced from global meteorological reanalysis, based on prior knowledge of the plume altitude. Plume altitude can be determined from advanced retrieval algorithms, or by optimizing the agreement between wind direction and the direction of plume transport visible in the satellite images. Here, a fixed ERA-5 pressure level has been used for simplicity, but daily variations of plume altitude, hence speed, could be easily accommodated.

The ratio between estimated SO$_2$ flux and its posterior uncertainty is exploited in a statistical test to automatically flag occurrences of volcanic degassing. This procedure avoids false positives triggered by fluctuations of noise or intrusion of SO$_2$ plumes from an external origin.

Application to three eruptions at Piton de la Fournaise (2021–2023) demonstrates that the method allows for capturing small eruptive fluxes (down to $\sim 0.5$ kton.$\cdot$day$^{-1}$), while remaining immune to the presence of the diluted stratospheric plume from 2022 Hunga Tonga–Hunga Ha’apai eruption overpassing La Réunion. A one-year-long time-series at Etna (2021) shows that the method allows for measuring the SO$_2$ flux for a broad range of degassing intensities, from short-lived episodes of paroxysmal activity (with fluxes $> 10$ kton.$\cdot$day$^{-1}$) to weeks- to months-long intervals of passive degassing (with fluxes $\sim 2$–3 kton.$\cdot$day$^{-1}$). Caveats include the presence of meteorological clouds, which lead to an underestimation of SO$_2$ abundance by the satellite retrieval. Nevertheless, both at Etna and Piton de la Fournaise, we find a reasonable day-to-day correlation between the SO$_2$ mass flux estimated by satellite and seismic energy recorded on the ground.

The “disk method” is adapted to derive daily-averaged emission rates from the standard TROPOMI L2 SO$_2$ product, especially for weakly degassing sources situated in noisy environments. The method has been developed to facilitate automatic processing of large volumes of data, considering (a) that evaluating modest volcanic SO$_2$ emissions close to the measurement noise is a crucial need for certain applications, especially to capture pre-eruptive fluxes, and (b) that information on local wind velocity and plume altitude is not necessarily available at the time of satellite acquisition. The method is generic, and
readily adaptable to other trace gas observations from TROPOMI or from other UV or IR
hyperspectral sensors (IASI, OMI, OMPS).

Implementation of the algorithm as an open-access web app is made available to users at
https://dataviz.icare.univ-lille.fr/so2-flux-calculator, within the framework
of the Volcano Space Observatory Portal (see Supporting Text S5). The app offers
interactive features, such as responsive widgets to ease the adjustment of input
parameters, and interactive visualization tools to assist human inspection and
post-processing. The method is also distributed as an open-source command-line tool in
Python language, available from
https://git.icare.univ-lille.fr/icare-public/so2-flux-calculator. These
implementations enable the computation of multi-year time-series, as well as the
day-by-day, case-by-case analysis of satellite acquisitions in near-real time, including
during the course of an eruption.

Open Research Section

In the framework of the Volcano Space Observatory Portal, the algorithm presented in
this paper is implemented in an on-demand web service, available at
https://dataviz.icare.univ-lille.fr/so2-flux-calculator. The algorithm is also
available as a stand-alone open-source Python package at
(https://git.icare.univ-lille.fr/icare-public/so2-flux-calculator, Grandin et
al., 2024b), distributed under MIT Licence. Datasets presented in this paper were
generated using the Python implementation of the algorithm. Input and output datasets
are available from the Earth System Data Repository (EaSy Data,
https://www.easydata.earth/, Grandin et al., 2024a). Note to reviewers: access to
the dataset is currently restricted. The dataset will be released in the public
domain when the paper is accepted. The dataset is made available for
peer-review as “Data File(s) for Peer Review” (file name: “EaSyData.zip”).

The web-based VOLCPLUME Platform was used for satellite analysis (Boichu &
Mathurin, 2022, https://www.icare.univ-lille.fr/volcplume). VolcPlume is freely
accessible (via https://volcplume.aeris-data.fr) and is hosted by AERIS/ICARE
Data and Services Centre (https://www.icare.univ-lille.fr).

All data used in this study are publicly available:
- Facilities of the European Space Agency (ESA) were used for access to Sentinel-5P TROPOMI Level 2 SO2 products (ESA Copernicus, 2020).
- Facilities of the Copernicus Climate Change Service Climate Data Store (CDS) were used for access to European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-5 global reanalysis (Copernicus Climate Change Service Climate Data Store (CDS), 2023).
- Facilities of the Observatoire Volcanologique du Piton de la Fournaise (OVPF) and Institut de Physique du Globe de Paris (IPGP) were used for access to seismic data acquired at Piton de la Fournaise (Observatoire Volcanologique Du Piton De La Fournaise (OVPF) & Institut De Physique Du Globe De Paris (IPGP), 2008).
- Facilities of the Istituto Nazionale di Geofisica e Vulcanologia (INGV) were used for access to seismic data acquired at Etna (Istituto Nazionale di Geofisica e Vulcanologia (INGV), 2005).

Interactive tools used in the SO$_2$ Flux Calculator web app are based on libraries of the Holoviz ecosystem (Stevens et al., 2015).

The ObsPy library was used for the processing of seismic data (Krischer et al., 2015). The ssxm.py script was used for computing RSAM (https://github.com/ThomasLecocq/ssxm/blob/master/ssxm.py, Lecocq, 2017).

Figure S9, adapted from Chevrel et al. (2023), shows products derived from the MIROVA service (Coppola et al., 2016; Campus et al., 2022) and the HOTVOLC service (Gouhier et al., 2016).

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Figure 7. Left: TROPOMI SO\(_2\) column amount around Piton de la Fournaise (a-b) during the January 2022 eruption, (c) after eruption end and during overpass by the Hunga Tonga Hunga Ha’apai (HTHH) plume, and (d) after HTHH plume overpass. The blue arrows for the eruptive cases (a1) and (b1) show the wind vectors deduced from ERA-5 (700 hPa pressure level), which are consistent with the direction taken by the plume. Right: best-fitting mass-versus-distance regression for the data points derived from integration of SO\(_2\) mass over disks (black dots). Line colors are the same as in Figure 1 (yellow: volcanic; red: noise; green: sum). For each plot, the inset shows a zoom on the four data points within 100 km from the volcano.
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