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TRACKING MAGMATIC PROCESSES THROUGH SO₂ FLUX MEASUREMENTS AT OPEN-VENT VOLCANOES

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ABSTRACT

Volcanic gas monitoring plays a central role in modern volcano surveillance, offering critical insights into magmatic processes and contributing to the forecasting of eruptive events. Among volcanic gases, sulphur dioxide (SO_2) is widely recognized as a key tracer of shallow degassing dynamics and magma transport within the upper portion of a volcano's plumbing system. This thesis investigates the utility of high-resolution SO_2 flux records acquired from permanent Ultraviolet (UV) Camera systems to monitoring the activity of Mount Etna and Stromboli, two persistently active, open-vent volcanoes in Sicily. Particular emphasis is placed on the interpretation of SO_2 degassing trends in relation to magma dynamics and eruptive phases, and on the assessment of the uncertainties affecting SO_2 flux measurements.

The two volcanoes exhibit a wide range of eruptive behaviours and associated hazards, making them ideal sites for testing and refining gas-based monitoring techniques. Stromboli's persistent activity is characterized by mild-to-moderate Strombolian explosions recurring at regular intervals, with occasional more violent major and paroxysmal explosions that can affect inhabited areas and trigger slope instabilities and tsunamis. Etna's activity is dominated by continuous degassing from its summit craters, usually accompanied by mild Strombolian explosions, which is often interrupted by effusive eruptions and powerful lava fountain episodes, which can produce high eruptive columns. These eruptive phenomena pose significant hazards, including destructive lava flows, ash emissions that compromise air traffic and health, and the potential for pyroclastic density currents and lahars with severe impacts on populations and infrastructure.

The decadal SO_2 flux records analysed here allow a variety of topics to be analysed with unprecedented detail and temporal resolution. At Stromboli, I undertake the first long-term (nine years) systematic comparison between near-vent SO_2 fluxes derived from UV Cameras and those obtained from a network of distal scanning spectrometers coupled with the DOAS technique. A large SO_2 flux mismatch between the two time series is observed. This offset is attributed to plume coverage limitations and radiative transfer effects in UV Camera data, and to overestimation in DOAS-derived fluxes using wind speed without accounting for plume dilution from source to the analysis section. A novel integrated SO_2 flux is developed to address these issues, combining DOAS column amounts with UV Camera-derived plume velocities. In parallel, multi-year datasets of SO_2 and CO_2 concentrations are analysed to identify gas-based precursors to explosive events. Statistical results indicate that periods of reduced SO_2 concentrations and fluxes and elevated CO_2/SO_2 ratios consistently precede major explosions. Based on this, a gas-based composite indicator is proposed, which retrospectively forecasts over 70% of events on timescales of weeks. The Stromboli's July 2024 eruption is also examined in detail, with particular focus on the differing behaviour of SO_2 and

CO₂ fluxes as indicators of shallow and deep magmatic degassing, respectively. Results show that the eruption was preceded and accompanied by a large escalation in the CO₂ flux relative to the SO₂ flux, suggesting a deeper magmatic gas source, consistent with the involvement of gas-rich magma ascending from greater depths. These findings highlight the diagnostic value of elevated CO₂ fluxes in detecting deep gas release preceding explosive activity that may not be immediately evident from SO₂ measurements alone.

At Mount Etna, ~10 years of UV Camera data are used to characterise SO₂ flux variations associated with a wide range of eruptive style, from variable effusive eruptions, differing in volume and duration, to paroxysmal lava fountain events. These data have been analysed in combination with geophysical and satellite thermal data, to obtain a more comprehensive understanding of eruptive mechanisms and involved magma volumes. A particular focus was placed on the paroxysmal sequences of 2020-2021, during which the highest SO₂ fluxes and erupted volumes were recorded. The results point to rapid injections of volatile-rich magma into a shallow reservoir located at ~3-4 km depth as the main driver of these intense eruptive phases, causing a sharp increase in magma supply rate above the sulphur exsolution threshold (~150 MPa).

Taken together, these findings advance our understanding of degassing dynamics at open-vent systems and contribute to a deeper insight into eruptive mechanisms. By addressing key methodological uncertainties and integrating multiple datasets, these studies contribute to refining monitoring strategies and strengthening interpretations of gas-based eruption precursors at active volcanoes.

CHAPTER 1

INTRODUCTION

Active and potentially dangerous volcanoes are increasingly monitored by advanced geophysical and geochemical networks (Acocella et al., 2023; Sparks et al., 2012), with the aim of detecting changes in seismic activity (Matoza and Roman, 2022), ground deformation (Biggs and Pritchard, 2017), thermal emissions (Reath et al., 2019a) and volcanic gas composition (Edmonds, 2021; Kern et al., 2022), which can herald the transition from quiescent to eruptive phases (Poland and Anderson, 2020; Sparks and Cashman, 2017). Among these techniques, monitoring volcanic gas composition stands out as a particularly sensitive indicator, having long been recognized as a valuable tool for detecting changes in magmatic activity preceding eruptions (e.g., Aiuppa et al., 2010, 2025a, 2025b or Chapter 6, 2025c; Caltabiano et al., 2004; Menyailov, 1975).

Volcanic plumes are primarily composed of water vapor (H_2O), carbon dioxide (CO_2), and sulphur dioxide (SO_2), which represent the most abundant gas species released during volcanic degassing. In addition to these major components, plumes also contain variable range of minor and trace gases, including hydrogen sulphide (H_2S), carbon monoxide (CO), hydrogen chloride (HCl) and hydrogen fluoride (HF). Magmatic volatiles exhibit limited solubility in silicate melts, causing their exsolution and the formation of a separate gas phase under crustal to upper mantle conditions (Papale et al., 2022; Sparks, 2003). Due to their high mobility, this gas phase can decouple from deeply stored magma and migrate toward the surface independently (Edmonds et al., 2022). This process of magmatic degassing significantly influences magma evolution during storage and ascent, playing a central role on both magmatic differentiation and eruptive behaviour (Anderson, 1975; Edmonds, 2021; Edmonds et al., 2022; Fischer and Chiodini, 2015). Moreover, gas bubble nucleation, driven by decompression and cooling during magma ascent, provides the driving force for explosive volcanism (Gonnermann and Manga, 2003), the most dangerous expressions of volcanic activity.

Among the various gas-related parameters currently monitored at volcanoes, the SO_2 flux stands out as one of the most informative indicator of magma supply rates and degassing dynamics within the upper portions of volcano's plumbing systems (Oppenheimer et al., 2011, 2014; Wallace and Edmonds, 2011). In mafic systems, the onset of SO_2 exsolution typically occurs at pressures below approximately 150 MPa, corresponding to depths of less than ~ 3 km (Métrich et al., 2010; Moretti and Papale, 2004), which makes it particularly sensitive to shallow magmatic processes. As a result, SO_2 plays a pivotal role in tracking volcanic unrest and has been recognized as a key parameter in the assessment of volcanic hazards. Despite being the least abundant among the three

major volcanic gas species, SO₂ exhibits very low natural atmospheric abundances and strong absorption features in the ultraviolet (UV) region of the electromagnetic spectrum, making it easily measurable by spectroscopic remote sensing methods (e.g., Kantzas and McGonigle, 2008; Oppenheimer et al., 2011). Over the past two decades, significant progress has been made in the instrumental monitoring of volcanic SO₂ fluxes (Oppenheimer et al., 2014), both with satellite (e.g., Carn et al., 2016, 2017; Coppola et al., 2019) and ground-based techniques (e.g., Aiuppa et al., 2021; Salerno et al., 2009a), achieving increasingly high-resolution and accurate measurements.

Building on these developments, this PhD thesis explores the role of SO₂ emissions for monitoring the degassing behaviour and pre- to sin-eruptive activity of Mount Etna and Stromboli, two persistently active open-vent volcanoes located in Sicily, Italy (Figure 1.1). Mount Etna, situated on the eastern coast of Sicily, is the highest and most active volcano of Europe, known for its frequent effusive and paroxysmal lava fountain events. Stromboli, the northernmost of the Aeolian Islands archipelago, is famous for its continuous mild explosive Strombolian activity, often referred to as the “Lighthouse of the Mediterranean.”

In particular, results here reported come from the analysis of SO₂ flux time series acquired using UV Camera technology, a ground-based remote spectroscopic technique renowned for its high spatial and temporal resolution. In particular, I exploit the use of long-term, semi-continuous SO₂ flux records delivered by permanent, stand-alone UV Camera systems (Delle Donne et al., 2017, 2019). To gain a more comprehensive understanding of volcanic processes, SO₂ flux data are integrated with satellite thermal observations and geophysical parameters. Satellite-based thermal observations allow for the measurement of volcanic radiant heat fluxes emitted by active lava flows. These fluxes can be empirically converted into lava discharge rates through calibration with ground-based data or using physical models that link surface temperature and emissivity to effusion rates. By integrating the discharge rates over time, the total volumes of lava erupted during the activity can be estimated. Additionally, among the geophysical parameters, volcanic tremor exhibits key complementary with SO₂ flux analysis. This continuous seismic signal is almost ubiquitously observed at active volcanoes and is generated by the movement of gas bubbles or bubbly magma within the conduits (Ripepe and Gordeev, 1999; Zuccarello et al., 2013). Because tremor amplitude is modulated by the same genetic processes that control SO₂ exsolution (Nadeau et al., 2011), its integration with SO₂ flux data enables a robust interpretation of magmatic dynamics and gas transport processes.

This thesis also investigates the main sources of uncertainty affecting SO₂ flux measurements using UV Cameras, as a contribute to the development of increasingly accurate and reliable monitoring strategies. An attempt to reduce uncertainties in SO₂ flux measurements is performed at Stromboli volcano by combining measurements obtained from UV Camera and scanning

spectrometer coupled with the DOAS (Differential Optical Absorption Spectroscopy) technique, which represents the most widely adopted ground-based tools for SO₂ flux monitoring in recent years. Despite these techniques are well established, achieving continuous and reliable SO₂ flux measurements remains challenging due to methodological and operational constraints (e.g., Johansson et al., 2009; Kern et al., 2010a, 2010b; Klein et al., 2017; McGonigle et al., 2005a, 2005b), and a systematic, long-term comparison between the two techniques is crucially required.

During the course of my PhD, I also investigated the degassing dynamics and the role of SO₂ flux during eruptive events at Stromboli and Etna volcanoes, focusing on improving eruption forecasting and enhancing the understanding of eruptive dynamics and the source mechanisms driving explosive processes. The results obtained from this research are presented and discussed in Chapters 4, 5, 6, 7, 8 and 9. While some of these findings have already been published in peer-reviewed scientific journals, others are original contributions that are presented here for the first time.

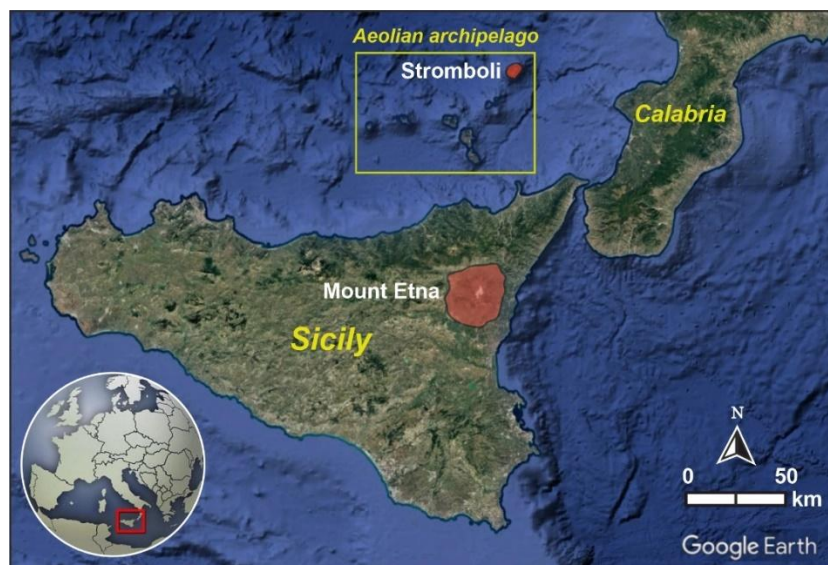


Figure 1.1 – Geographic location of Mount Etna and Stromboli volcanoes.

CHAPTER 2

STATE OF THE ART: GROUND-BASED SO₂ FLUX MEASUREMENTS

2.1 Introduction

The history of volcanic SO₂ monitoring began in the 1970s with the use of first Correlation Spectrometers (COSPEC; Millan et al., 1976; Moffat and Millan, 1971; Newcomb and Millan, 1970). These instruments were originally designed for the measurement of point emissions of SO₂ and NO₂ from industrial and chemical plants, but their potential application in volcano monitoring (SO₂) was quickly recognised (Stoiber and Jepsen, 1973), enabling the first estimates of SO₂ flux by ground-based remote sensing. The introduction of more compact and cost-effective ultraviolet spectrometers in the early 2000s marked a turning point in volcanic gas monitoring. In contrast to COSPEC, which operates over a narrow ultraviolet spectral range using an internal mechanical correlation mechanism (Millán, 1980), modern spectrometers are capable to capture the full UV spectrum that can be subsequently processed using the Differential Optical Absorption Spectroscopy (DOAS) technique to retrieve SO₂ column densities. Thanks to their portability, improved spectral resolution and ability to detect multiple gas species simultaneously, these instruments have become the gold standard for SO₂ measurements (Galle et al., 2003; Platt and Stutz, 2008) and are now widely adopted for routine SO₂ monitoring by volcano observatories. When deployed in fixed, multi-angle configurations, automated networks of scanning spectrometers enable remote sensing of volcanic gas emissions from several viewpoints, allowing for a continuous integrated assessment of a “bulk” gas output (Arellano et al., 2021; Edmonds et al., 2003; Galle et al., 2010; Salerno et al., 2009a, 2009b). However, their accuracy is limited by assumptions regarding plume geometry, height, and especially transport speed, typically inferred from wind data (e.g., using meteorological models or local anemometers), which can introduce substantial uncertainties (McGonigle et al., 2005a, 2005b).

The advent of UV Camera systems (McElhoe and Conner, 1986; Mori and Burton, 2006, 2009; Bluth et al., 2007) has provided an alternative technique for SO₂ flux measurements, enabling higher spatial and temporal resolution compared to traditional spectrometric methods. Near-vent deployments enable vent-specific observations and allow the detection of rapid degassing phenomena such as puffing, passive emissions and small explosive bursts (Tamburello et al., 2012). These high-frequency measurements allow for the detection of short-term variations in volcanic gas output, improving the integration of gas data with seismic and deformation signals (Dalton et al., 2010; Delle

Donne et al., 2017). This integration enhances the interpretation of shallow conduit dynamics and provides critical insights into magmatic processes, thereby contributing to more effective short-term eruption forecasting (Dalton et al., 2010; Delle Donne et al., 2017; Holland et al., 2011; Nadeau et al., 2011; Aiuppa et al., 2025b). Furthermore, the recent implementation of optical flow algorithms on acquired images allows for direct estimation of plume velocity near the source, significantly improving flux accuracy (Delle Donne et al., 2017, 2019, 2022; Gliß et al., 2017; Peters and Oppenheimer, 2018). However, the UV Camera method, as other spectroscopic techniques used in volcanology (Oppenheimer et al., 2011), is constrained by visibility issues related to local topography and meteorological conditions, as well as by radiative transfer effects in optically thick or distal plumes (Kern et al., 2010a, 2010b, 2012, 2013).

2.2 Basic theory

Remote volcanic gas sensing spectroscopic techniques mainly rely on the attenuation of light by a volcanic gas plume. These methods are broadly categorized into active and passive, depending on the source of the detected light. Active remote sensing techniques use artificial light sources to illuminate the target, providing their own energy source for measurements. In these systems the radiation is emitted by the instrument directed toward the surface of interest, and the spectral response, resulting from reflection, absorption or scattering processes, is analysed. This controlled illumination enables measurements to be carried out independently of natural light conditions, allowing for continuous data acquisition, including at night. In contrast, passive remote sensing uses natural light sources, primarily solar radiation passing through, and being scattered by, the volcanic plume. Passive techniques are advantageous because they are relatively simple to operate and present relatively low operational costs. However, their applicability is limited to daylight hours and depends on favourable weather and illumination conditions.

The attenuation of light by volcanic plumes includes both a continuous component, due to scattering and broadband absorption by air molecules and aerosols, and discrete absorption features associated with specific volcanic gas species (Figure 2.1), as described by the Lambert-Beer Law:

$$I(\lambda) = I_0(\lambda)e^{-\sigma(\lambda)NL} \quad (\text{Eq. 2.1})$$

where $I(\lambda)$ and $I_0(\lambda)$ denote the spectral irradiance (power per unit area per unit wavelength) after and before interaction with the target gas (typically measured in $\text{W}\cdot\text{m}^{-2}\cdot\text{nm}^{-1}$), $\sigma(\lambda)$ is the absorption cross-section of the absorbing gas species, N is the number of molecules per volume unit, and L represents the effective optical path length through the plume. However, the application of the Lambert-Beer law (Eq. 2.1) relies on the assumptions that the optical path through the volcanic plume is straight, the gases are uniformly distributed and that multi-scattering effects are negligible. It should

be noted that for derived quantities, such as total gas mass or flux, deviations from a straight path may introduce errors, and more advanced radiative transfer models may be required.

In volcanological applications, SO₂ has been the primary target of remote sensing studies due to its strong absorption features in the Ultraviolet (UV) region. Its typically low background atmospheric concentrations (ppb) and relatively high abundance in volcanic plumes (ppm), makes SO₂ more suitable for remote detection than other major volcanic gas species such as water vapor (H₂O) and carbon dioxide (CO₂), whose elevated and variable atmospheric concentrations significantly limit their identification. However, most remote sensing techniques do not directly measure the concentration of a gas, but instead determine a column density, which describes the integrated absorber concentration along the average photon path, from the light source to the detector. Integrated Column Amounts (ICA) are derived by spatially integrating these column density values across a plume cross-section. The SO₂ flux is then calculated by multiplying the SO₂ column amounts with the plume transport velocity, assumed from local wind speed or directly estimated from the plume motion using optical flow or cross-correlation techniques.

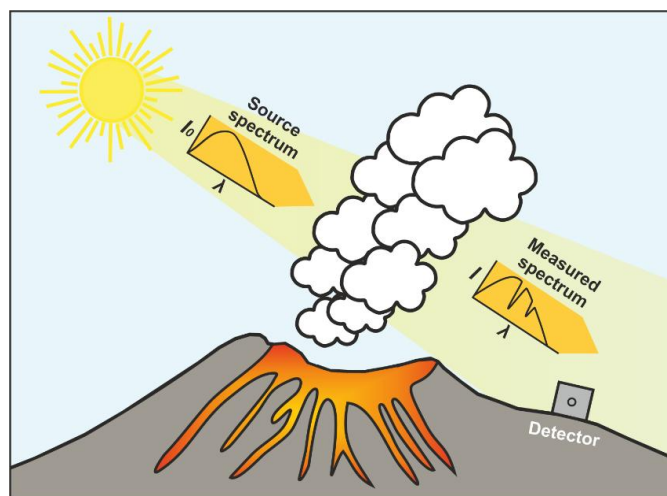


Figure 2.1 - Schematic illustration of solar radiation transmission through a volcanic plume prior its detection. The source spectrum and the measured spectrum differ as a function of the plume's composition, due to absorption in specific wavelength (λ) by distinct gaseous species.

2.3 Sources of uncertainty

2.3.1 Radiative transfer

Remote spectroscopic measurements of volcanic SO₂ flux are fundamentally based on the application of the Lambert-Beer law, which relates the measured attenuation of solar radiation to the amount of gas along the optical path. However, this approach rests on the simplifying assumption that radiation travels in a straight line through the plume, hence neglecting possible multiple scattering effects. In the context of remote sensing of volcanic plumes, radiative transfer refers to the

physical processes governing how sunlight or artificial radiation interacts with volcanic gases and aerosols within the plume and the surrounding atmosphere (Kern et al., 2010a, 2013). When light travels through a volcanic plume, it can be attenuated by the presence of absorption gases, or scattered by ash particles and condensed water droplets (Figure 2.2). Moreover, sunlight is also scattered throughout the atmosphere by air molecules, aerosol particles and water droplets, such as those found in clouds. These interactions alter the intensity and spectral composition of the detected signal and can significantly affect the accuracy of gas column retrievals if not properly accounted for. In light of the above, using the solar light for measurements, the optical path from the sun to the instrument varies for each individual photon and cannot be precisely defined. Furthermore, while some photons are scattered in the atmosphere behind the plume, others are scattered toward the instrument from the region between the instrument and the plume. These photons do not pass through the plume and therefore their contribution does not contain spectral absorption structures originating from plume constituents. This effect, called light dilution, dilutes the apparent absorption signal, resulting in an underestimation of the retrieved column densities.

Radiative transfer effects become particularly significant under conditions of high plume optical thickness (Kern et al., 2013), which may occur in case of high aerosol and ash load plumes. Volcanic plumes typically contain significant quantities of water vapor initially emitted as gas at high temperatures, which may condense in aerosol particles during atmospheric cooling. In contrast to SO_2 , which produces narrow-band absorption features in the UV spectral range, volcanic ash and aerosols exhibit broadband absorption that extends across a wider range of wavelengths. As a result, volcanic plumes are often optically opaque, especially in near-vent conditions, where gases remain relatively undiluted by atmospheric processes. These conditions enhance particle scattering, increasing the effective path length through the plume compared to the idealized straight-line path, potentially leading to overestimations of column amounts. However, in cases of strong absorption into the UV wavelengths, either due to high in-plume concentrations of SO_2 , or in presence of absorbing aerosols, photons traveling along longer paths are more likely to be absorbed before reaching the detector. This is related to a wavelength-dependent shortening of the average photon path within the plume (Kern et al., 2010a). Furthermore, in presence of scattering aerosols inside the plume, photons are increasingly likely to be redirected out of the plume toward its surface, without penetrating deep into the plume. However, while long photon paths still tend to dominate under conditions of weak absorption, strong SO_2 absorption effectively suppresses the long trajectories. As a result, the measured signal becomes dominated by short paths, leading to an overall underestimation of column densities. While these effects have been known for long time (Millán, 1980), accurately accounting for radiative transfer effects is still a challenge. However, significant progresses have been

made in recent years to mitigate these effects, through advances in radiative transfer modelling and the development of correction algorithms for volcanic plume measurements (e.g. Kern et al., 2010a, 2012).

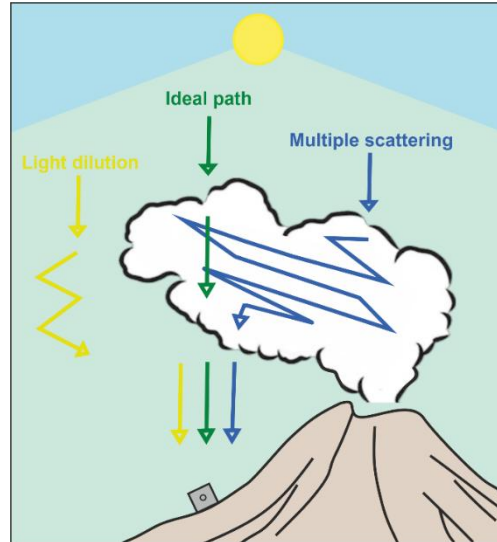


Figure 2.2 – Schematic representation of the radiative transfer effects. Arrows show the different possible photon paths from the sun to a remote sensing instrument. The green arrow shows the idealized photon path, where light passes through the plume in a straight line. The yellow arrow represents the light dilution effect. Light is scattered between the plume and the instrument, without passing through the plume. The blue path depicts the case of multiple scattering occurring inside the plume causing an extension of the light path (Kern et al., 2010a).

2.3.2 Plume speed evaluation

An accurate estimation of the plume transport speed is essential for reliable volcanic gas flux measurements. However, this is a complex task because the actual motion of the plume is affected by both internal and external factors. Near the vent, gas ascent is essentially driven by convection and still retains much of its initial buoyancy-driven velocity, resulting from its elevated temperature and low density relative to the surrounding air. As the plume rises and moves downwind, its transport becomes increasingly controlled by wind conditions, which can vary significantly with altitude, time, and local topography. For this reason, operational networks often rely on external meteorological data or in situ anemometric measurements to estimate wind speed at plume altitude, using it as a proxy for the actual transport velocity of the volcanic plume. However, the wind field at plume level may not accurately represent the motion of the plume itself, as wind measurements taken at the summit often do not accurately represent wind conditions at the location where gas flux measurements are performed, typically a few kilometres downwind. This assumption can introduce significant uncertainties in the calculated fluxes using wind speed, with average errors ranging from 10% to 40%, and in some cases exceeding 100% in distal measurements (Arellano et al., 2021; McGonigle

et al., 2005a, 2005b; Salerno et al., 2009a; Williams-Jones et al., 2006). Despite these limitations, wind speed remains the most practical solution for routine flux estimation in many spectroscopic techniques, such as using scanning spectrometers.

Several approaches to directly estimate gas velocity from image-based techniques have been developed in recent years, finding broad applications in UV Camera systems at high resolution. Among these, cross-correlation analysis and optical flow algorithms represent two of the most commonly applied methods in volcanic plume studies. The former provides velocity estimates by tracking the time delay between plume structures across spatially offset profiles, while the latter enables the reconstruction of velocity fields by analysing the motion of gas between consecutive frames. Modern optical flow algorithms leverage on sophisticated mathematical models, enabling more accurate and robust tracking of plume motion (e.g., Delle Donne et al., 2019). When applied in near-vent conditions, these techniques can capture plume motion at its source, before wind advection and mixing at larger scales modify its structure. Near-vent applications allow the plume's observation before it is significantly advected by wind, improving the reliability of velocity retrievals and reducing uncertainties in SO₂ flux estimates (Delle Donne et al., 2017, 2019, 2022; Gliß et al., 2017; Peters and Oppenheimer, 2018).

2.4 The DOAS technique

2.4.1 Measurement principle

In contrast to COSPEC, which samples only a few discrete spectral elements, modern spectrometers capture the full UV spectrum that can be subsequently processed using the Differential Optical Absorption Spectroscopy (DOAS) technique to retrieve SO₂ column densities. Owing to their portability and affordability, UV spectrometers have been extensively deployed on active volcanoes and are now widely adopted for routine SO₂ monitoring by volcano observatories.

SO₂ flux measurements can be performed using two general approaches: mobile traversing beneath the plume (e.g., using vehicles, boats, or aircrafts), or scanning from fixed positions downwind of the emission source. While mobile methods are often used for campaign-based measurements, the stationary configuration has emerged as one of the most widely adopted methods for continuous ground-based SO₂ flux monitoring, particularly due to its easy automatization (Galle et al., 2010; Salerno et al., 2006). By rotating a telescope along a vertical plane intersecting the gas plume (Edmonds et al., 2003; Galle et al., 2007), typically using rotating heads driven by a stepper motor, this technique enables the automatic retrieval of multiple integrated SO₂ column amounts across the plume's cross-section during the day. When combined with plume velocity estimates, this allows flux quantification at high temporal resolution (generally few minutes). Traditional scanning

DOAS networks deploy multiple fixed spectrometers at strategic locations around the volcano, typically a few kilometres from the vent, enabling the scanning planes of each instrument to intercept all potential plume directions under prevailing wind conditions. The instruments perform scans in azimuthal planes, either flat or conical, by rotating over an angular range of approximately 180° , with regular steps of few degrees. Complete scans of the plume cross-section are obtained when the wind disperses the volcanic plume over at least one of the spectrometers of the network. Networks of scanning spectrometers (Figure 2.3), such as the FLAME (FLux Automatic MEasurement) at Stromboli (Burton et al., 2009, 2015; Salerno et al., 2009a), have proven to be effective tools for providing real-time, fully automated measurements of SO_2 flux, offering the advantage of remotely capturing volcanic plumes from multiple viewing angles. A global network of scanning spectrometers is today operated by the NOVAC initiative (Arellano et al., 2021; Galle et al., 2010).

The principle of DOAS is based on the concept of differential optical depth, which separates the broadband spectral features, mainly due to light scattering, from the narrowband absorption structures caused by trace gases. To perform a DOAS analysis, two spectra are required: a first spectrum recorded intercepting the volcanic plume and containing the absorption features of volcanic gases and the so called “reference spectrum”, ideally representing the original spectrum without the presence of volcanic gases (Solomon et al., 1987). The differential optical depth is then derived from the ratio of these two spectra, enabling quantification of trace gas column densities. As it inherently measures the full spectrum of the incident radiation, the DOAS technique offers a distinct advantage over other methods. This allows for a more detailed analysis and the potential to correct or mitigate radiative transfer-related uncertainties during retrieval processes.



Figure 2.3 – UV scanning spectrometer used for remote sensing of volcanic gas emissions. The depicted instrument is part of a network of fixed scanning spectrometers designed to automatically retrieve SO_2 fluxes from volcanic plumes (photo from [INGV newsletter](#)).

2.4.2 Sources of uncertainty

In volcanological applications, the acquisition of reference spectra presents significant challenges, especially during real-time DOAS measurements with automated UV scanning instruments. This difficulty arises from the presence of wide plumes above the stations, preventing the acquisition of clear-sky reference spectra (free from volcanic gas absorption). Various strategies, such as employing synthetic or modelled reference spectra, have been developed to mitigate this issue (Salerno et al., 2009b), though each approach introduces its own limitations and potential sources of uncertainty. Furthermore, determining gas fluxes from a network of ultraviolet scanning spectrometers relies on determination of several interdependent factors. These include the geometry and height of the volcanic plume, an estimation of wind speed, and the proper quantification of multiple scattering effects. Many of these parameters cannot be independently measured with high accuracy, adding uncertainty to the flux estimation. Plume height and geometry are typically estimated through geometric triangulation. In some cases, visual observations, webcam footage, or empirical relations between wind speed and plume altitude are used to constrain plume elevation (Queißer et al., 2019; Salerno et al., 2009a). However, among all factors involved in flux measurements, the use of wind speed data as proxies for effective plume speed represents the largest source of uncertainty in scanning measurements using DOAS (Barrington et al., 2022; McGonigle et al., 2005a, 2005b; Salerno et al., 2009a; Stoiber et al., 1983).

2.5 The UV Camera method

2.5.1 Measurement principle

Since their development (Bluth et al., 2007; Mori and Burton, 2006), Ultraviolet (UV) Cameras have significantly advanced the observation of volcanic SO₂ fluxes, extending the applicability of SO₂ flux measurements within volcano monitoring frameworks. These instruments (Figure 2.4) provide high spatial and temporal resolution imaging of SO₂ distributions in volcanic plumes (e.g., Delle Donne et al., 2017; Ilanko et al., 2020; Wilkes et al., 2023), allowing to detect fast degassing phenomena (Dalton et al., 2010; Pering et al., 2015; Tamburello et al., 2012) and a more effective integration with geophysical datasets (Dalton et al., 2010; Delle Donne et al., 2017; Holland et al., 2011; Nadeau et al., 2011).

UV Cameras operate on the principle of contrasting the brightness of the pixels in the presence/absence of SO₂ absorption, by exploiting the strong absorption of solar UV radiation by SO₂, which is particularly significant in the 300-320 nm range (Figure 2.5; Vandaele et al., 1994). This can be done using a narrow bandpass filter centred at approximately 310 nm, where the absorption of solar radiation by SO₂ is maximum (Figure 2.5). To correct for broadband effects such

as scattering by ash and aerosols, a second wavelength around 330 nm, where SO₂ absorption is negligible, is also used. This dual-band imaging can be implemented either through two co-aligned cameras each equipped with a different filter, or a single camera equipped with a rotating filter wheel. However, the two-camera setup is more commonly adopted, as it allows for the simultaneous acquisition of the two wavelengths, minimizing motion-related errors and enabling effective discrimination between gas absorption and aerosol scattering. Furthermore, compared to single-camera systems with a rotating filter wheel, the dual-camera setup avoids mechanical complexity, and improves both sampling frequency and robustness in field deployments.

A qualitative measure of an un-calibrated absorbance A can be obtained for each camera pixel according to the Lambert-Beer Law, which relates the absorption of light to the concentration of the absorbing gas:

$$A = -\log_{10} \frac{I_{310}}{I_{330}} - A_0 \quad (\text{Eq. 2.2})$$

where A is the absorbance, I_{310} and I_{330} are the pixel intensities associated with cameras mounting the 310 and 330 nm filters, while A_0 is the absorbance level associated with a clear background sky sub-area of the image.

Absorbance obtained using Eq. 2.2 can be converted into SO₂ column densities typically using calibration gas cells of known SO₂ concentration or employing a co-aligned UV spectrometer to provide real-time spectroscopic reference measurements within a portion of the camera's field of view (Lübcke et al., 2013). This procedure yields the proportionality ratio between absorbance and SO₂ column densities. Calibration using UV spectrometers offers a major advantage over traditional methods based on gas cells, as it allows for continuous and in situ calibration that reflects actual atmospheric and instrumental conditions. In contrast, gas cells provide a static reference that may degrade over time and fail to capture the variability of field measurements. By integrating SO₂ column density along a line of pixels intercepting the plume cross-section, an Integrated Column Amount (ICA) can be obtained. Finally, the SO₂ column amounts are multiplied by the plume transport speed to retrieve the gas flux.

One of the key advantages of UV Cameras is their capacity to retrieve plume transport speed with high precision (e.g., Bluth et al., 2007; Delle Donne et al., 2019; Peters and Oppenheimer, 2018). Plume speed is commonly obtained either through cross-correlation of successive ICA profiles along the direction of transport or by applying optical flow algorithms that estimate the velocity profile by comparing motion through successive frames. Recent integration of optical flow algorithms into permanent UV Camera systems (e.g., Delle Donne et al., 2017, 2019, 2022; Gliß et al., 2017; Peters and Oppenheimer, 2018) have allowed to directly estimate the plume's vertical velocity near the emission source, where atmospheric turbulence and wind influence are minimal. This capability

significantly reduces uncertainties related to plume speed and, by extension, to SO₂ flux estimation (Delle Donne et al., 2019).



Figure 2.4 – Typical dual-band UV Camera configuration, coupled with an UV Spectrometer for calibration of images and a visible camera. The visible camera is useful to check the quality of the images and the environmental conditions.

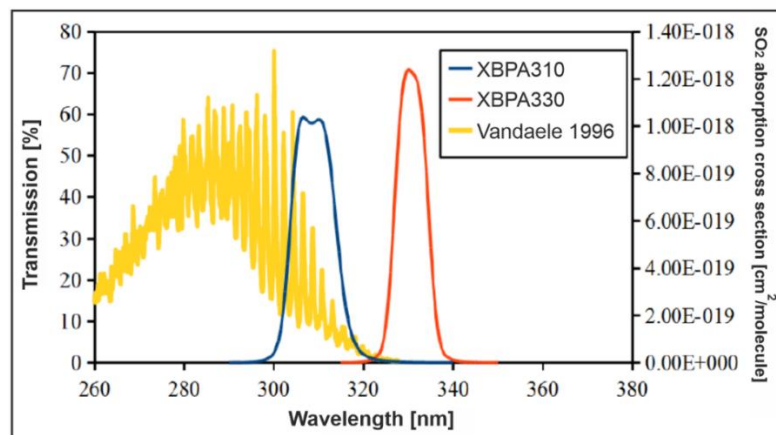


Figure 2.5 - Sulphur dioxide absorption cross section (Vandaele et al., 1994), with the transmission curves of the two band-pass filters used in the UV Camera system: XBPA310 centred at 310 nm (on-band, high SO₂ absorption) and XBPA330 centred at 330 nm (off-band, minimal SO₂ absorption).

2.5.2 Sources of uncertainty

Despite their high temporal resolution, UV Cameras rely on narrow pass-band filters rather than capturing the full UV spectrum, limiting their ability to correct for radiative transfer effects. This spectral limitation prevents the application of advanced retrieval techniques typically used to account

for atmospheric scattering effects both inside and outside the plume. The use of dual-band imaging in UV Cameras is one strategy to mitigate aerosol scattering, but this correction can only compensate for a small portion of the radiative effects. Although UV Cameras are generally more susceptible to radiative transfer effects, coupling them with a co-located UV spectrometer pointing to a known sub-area within the camera field of view, allows for a more accurate calibration of SO₂ column densities. The use of the UV spectrometer allows to quantitatively measure the full UV spectrum discriminating the different bandwidth contributions. However, acquiring data along a single line of sight in a defined region of the UV Camera image (e.g., Delle Donne et al., 2019; Lübcke et al., 2013) does not capture the full spatial variability of radiative transfer conditions across the entire plume imaged by the camera. As a result, image calibration may not be fully representative of all pixels in the camera's field of view, particularly under heterogeneous plume or background conditions.

When using UV Cameras for SO₂ flux measurements, the morphological complexity of the crater area can limit optimal instrument placement and affect the line-of-sight geometry, introducing additional inaccuracies, especially for fixed stations. Optimal instrument placement is therefore crucial to ensure a representative view of the entire plume. For an accurate estimation of gas flux, instruments must ideally capture the full cross-section of the plume. While scanning spectrometer networks are typically located at several kilometres from the source and are capable to capture the plume in its entirety using different viewing angles, UV Cameras require a more careful placement. Their effectiveness depends on an appropriate balance between instrument siting, prevailing wind direction, and summit morphology. Due to the complex topography of the summit area, especially in multi-vent volcanoes, the instrument's field of view may be limited and incapable of fully capturing the spatial extent of the gas plume. As a result, parts of the plume may remain hidden behind crater rims or other topographic features. This effect can be severely enhanced during days of strong wind, when the plume is forced downward or channelled along the ground behind topographic obstacles. Under such conditions, the geometric constraints imposed by both the instrument's viewing direction and the summit terrain may lead to an incomplete view of the plume, which can significantly underestimate the evaluated column densities and consequently gas fluxes.

Meteorological conditions also play a critical role in measurement quality. Ideally, the sun should be positioned behind the camera to ensure homogeneous sky illumination across the field of view. While the presence of meteorological clouds behind the camera field of view generally does not pose a major issue, clouds located between the observer and the gas plume can severely obstacle the measurements, due to scattering and absorption effects.

CHAPTER 3

THE UV CAMERA TECHNIQUE: HARDWARE, SOFTWARE AND DATA PROCESSING

3.1 Permanent UV Camera systems

The SO₂ flux data used in this PhD thesis were acquired using permanent, fully autonomous UV Cameras managed by the University of Palermo, installed at Mount Etna and Stromboli volcanoes. At Stromboli, the permanent UV Camera system has been operational since May 2014 at the Rocchette site, located on the northeastern upper flank of Stromboli at approximately 750 m above sea level. Positioned just 500 m northeast of the active vents, this site provides a clear view of the northeastern sector of the crater terrace (Figure 3.1a). The UV Camera system employed at Mount Etna has been operational since 2014 and is installed on the La Montagnola ridge at an elevation of 2600 m. This vantage point offers an excellent view of both the Southeast and Central crater areas, located approximately 3 km from the camera site (Figure 3.1b). These systems allow for continuous monitoring of SO₂ fluxes with high temporal resolution (0.5 Hz), using algorithms capable of autonomously managing image acquisition, such as the automatic tuning of the exposure associated with UV Cameras (Delle Donne et al., 2019). Images collected from the two aligned UV Cameras (filtered at 310 and 330 nm) are processed using ad hoc algorithms into single absorbance images, calibrated using gas cells of known SO₂ concentration and with data obtained from a co-aligned UV spectrometer. Calibration with the UV spectrometer allows a quantitative measurement of the full UV spectrum, discriminating the contributions of different wavelength bands and reducing errors related to radiative transfer effects. Spectra acquired along the central line of sight of the image are used to determine the SO₂ column quantitatively, converting the absorbance of all pixels in the image into calibrated SO₂ column densities (McGonigle, 2007). Technical specifications of the UV Camera systems employed in this thesis are detailed in the Methods of Chapters 5, 6, 7, 8 and 9.

The employed systems apply the Lucas and Kanade (Lucas and Kanade, 1981) optical flow algorithm to estimate the plume velocity profile directly from the image sequence. However, the image processing workflow is non-trivial and includes image pairing and synchronization, computation of absorbance from dual-band filtered images and spatial integration of SO₂ column densities across the plume cross-section. To enhance the signal-to-noise ratio of volcanic SO₂ emissions and reduce the influence of atmospheric effects, images processing is conducted on a restricted image region, typically focused as close as possible to the gas source above the crater area.

At Stromboli, this task is facilitated by the relatively simple morphology and more favourable weather conditions. Additionally, the proximity of the station to the active vents (~ 500 m) and its position on the crater terrace allow for a consistent and clear view of the northern craters. This configuration allows the automatization of image processing through an algorithm capable to identify and extract a well-defined sub-region directly above the crater area, thereby optimizing the quality and reliability of the measurements. Here, SO_2 column amounts and velocity profiles are calculated, obtaining a SO_2 flux time series with a temporal resolution of 0.5 Hz, providing detailed insight into gas dynamics at sub-minute timescales. Daily SO_2 fluxes are then automatically retrieved by averaging fluxes associated with best-viewing conditions during the day, applying a visibility index, calculated as the unsigned ratio between the mean pixel intensity associated with the camera FOV's portions capturing sky and ground, respectively (Delle Donne et al., 2019). SO_2 measurements are then selected by setting a threshold on the visibility index and discarding measurements below the threshold. The amount of data discarded depends strongly on the meteorological conditions, but on days with minimal cloud cover the full set of measurements is generally usable.

In contrast, Mount Etna presents more challenging operational conditions, due to its greater elevation and complex summit morphology. The summit area is frequently affected by adverse weather conditions, particularly during winter months. The UV Camera is located at a greater distance from the craters (~ 3 km) and the complex topography of the southern crater area makes it difficult to consistently analyse the gas plume directly above its emission source, especially under high wind conditions, which can rapidly displace the imaged plume. While an automated image processing algorithm was initially implemented for the Etna's UV Camera system (Delle Donne et al., 2019), manual processing was later preferred, as it yielded more consistent and reliable results under the variable observational conditions at the site. Manual inspection allows for frame-by-frame validation by an expert operator, ensuring greater accuracy and consistency in flux measurements.

Manual image processing at Etna was performed using a MATLAB custom-developed algorithm, which replicates the logics of the automated procedure, while allowing greater flexibility and control by the operator, hence optimizing results based on specific observational conditions. This tool enables the visualization of the synchronized frames acquired by both UV Cameras (filtered at 310 and 330 nm), alongside the corresponding absorbance images and frames from a co-aligned visible camera. The algorithm allows manual placement of background clear-sky regions within the 310 and 330 nm images (Figure 3.2a,b), which are used to compute absorbance values (Figure 3.2c), as well as the selection of the analysis area where SO_2 column densities are integrated and the plume velocity profile is derived (Figure 3.2c). The analysis area must be positioned to intersect the full cross-section of the plume, perpendicular to its main transport direction. Under calm wind conditions,

when the plume rises vertically, this region is typically placed directly above the crater area. However, strong wind conditions are common at Etna, frequently causing the plume to be deflected eastward, or more rarely westward. In such cases, due to the complex crater morphology, the analysis area is manually adjusted to intersect the plume as close to the source as possible, ensuring reliable measurements despite the plume deflection. Along this area, the SO₂ column densities are integrated, obtaining the Integrated Column Amounts (ICA, in kg/m). ICAs are then multiplied by plume speed (m/s) obtained by applying an optical flow algorithm capable of tracking gas fronts in consecutive frames (Delle Donne et al., 2019; Lucas and Kanade, 1981), resulting in a 0.5 Hz SO₂ flux time series (kg/s).

Despite their numerous advantages, maintaining permanent UV Camera stations in remote or challenging volcanic environments poses considerable logistical and technical difficulties. Continuous operation requires robust infrastructure for data transmission and storage, which can be limited by unstable power supply, adverse weather conditions or insufficient network coverage. Routine maintenance is also essential, as the optical windows must be regularly cleaned to avoid volcanic ash deposition or condensation, which can severely degrade image quality. In addition, camera lenses and filters may need periodic replacement due to degradation, particularly in acidic environments. Nonetheless, when properly maintained, permanent UV Camera networks represent one of the most powerful tools for high-frequency gas monitoring at open-vent volcanoes, offering valuable insights into degassing processes and their relationship to eruptive behaviour.

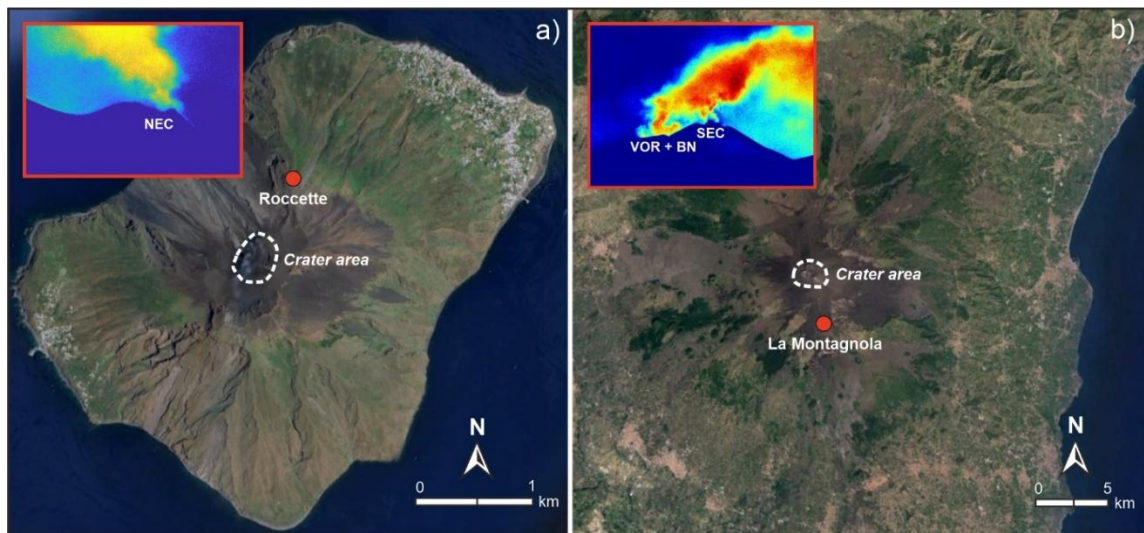


Figure 3.1 – Location and absorbance examples of UV Camera stations. a) Geographical position of the UV Camera installed at Roccette (Stromboli) and b) at La Montagnola ridge (Mount Etna), both selected to provide optimal viewing geometry of the volcanic plume.

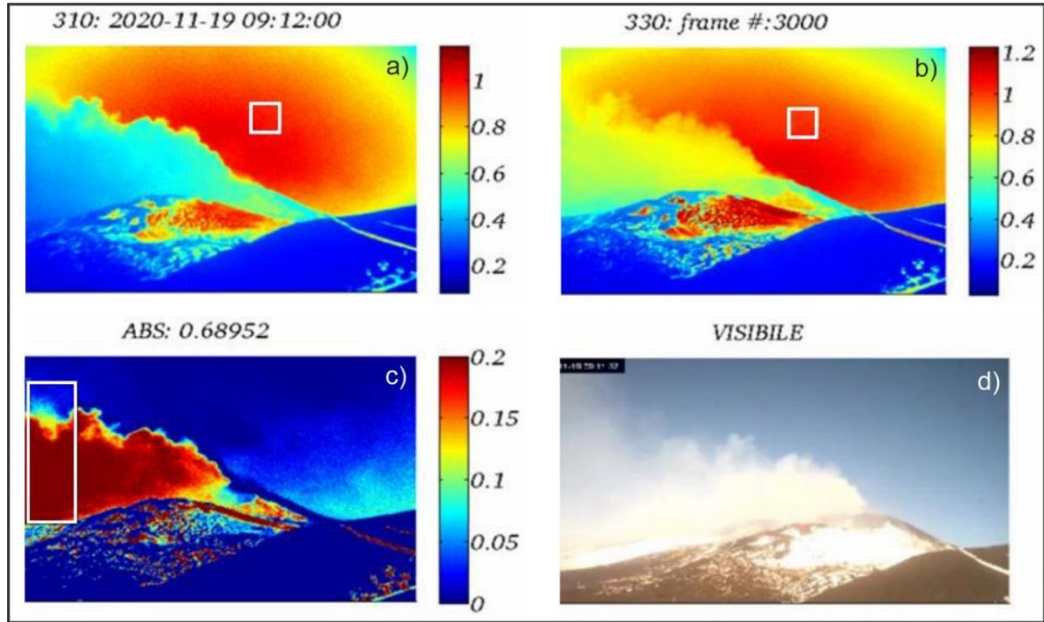


Figure 3.2 - Interface of the MATLAB-based application developed for the manual processing of UV Camera data recorded at La Montagnola (Etna). a,b) Example of images acquired by the 310 and 330 nm cameras. The white square shows the operator-selected atmospheric background region. c) Absorbance image obtained by combining the 310 and 330 nm frames, showing the selected plume cross-section (white area) used for the SO_2 flux calculation. d) Co-aligned visible camera.

3.2 Target volcanoes

3.2.1 Stromboli Volcano

Stromboli is the northernmost of the seven islands forming the Aeolian Archipelago, located in the southern Tyrrhenian Sea. It is world-recognized for its persistent open-vent volcanic activity, characterized by mild-to-moderate Strombolian explosions at regular intervals of approximately 10-20 minutes (Ripepe et al., 2008), originating from vents clustered within the crater terrace. This terrace is located approximately 750 m above sea level in the upper part of the Sciara del Fuoco, a prominent horseshoe-shaped depression formed by multiple flank collapses (Rosi et al., 2013). By convention, the crater area is divided into three main sectors: the southwestern (SW), central (C), and northeastern (NE) craters.

In addition to its "ordinary" explosive regime, Stromboli also experiences more energetic explosive events, classified as "major explosions" and "paroxysmal explosions." Major explosions typically occur a few times per year and are capable of ejecting large volcanic bombs and lithic blocks that can fall across the upper flanks of the volcano (Aiuppa et al., 2025b; Bevilacqua et al., 2020; Corradino et al., 2021). Paroxysmal explosions, though less frequent (occurring every few years), are substantially more powerful and can disperse ballistic material over several kilometres, potentially affecting low-altitude areas and inhabited zones (Andronico and Pistolesi, 2010; Métrich et al., 2021; Pioli et al., 2014). Explosive activity at Stromboli is occasionally accompanied by effusive eruptions,

with lava flows descending the Sciara del Fuoco and reaching the sea (Rosi et al., 2013). Both lava flows and large explosive events can destabilize the volcanic slope, inducing landslides that affect both subaerial and submarine portions of the edifice, potentially generating tsunamis. The presence of permanent settlements, Stromboli village to the northeast and Ginostra to the southwest, introduces additional exposure to volcanic hazards. Although these inhabited centres are located at lower elevations and at some distance from the summit vents, powerful paroxysmal explosions and associated phenomena (e.g., ballistic fallout, pyroclastic density currents, or tsunami) represent a serious risk, necessitating continuous monitoring of the volcano-related parameters.

3.2.2 Mount Etna

Mount Etna, located on the eastern coast of Sicily (Italy), is the largest active volcano in Europe, covering an area of approximately 1250 km² and reaching a current elevation of around 3330 m above sea level (Bonaccorso et al., 2004). It is one of the most active basaltic volcanoes on Earth and a major point source of volcanic SO₂ to the atmosphere (Allard, 1997; Caltabiano et al., 2004; Carn et al., 2017). Its persistent activity and accessibility have made it one of the most studied volcanoes worldwide.

Mount Etna is structurally complex and currently hosts four active summit craters: Voragine (VOR), Bocca Nuova (BN), the Northeast Crater (NEC), and the Southeast Crater (SEC). Its activity is primarily characterized by continuous degassing from its summit craters, usually accompanied by mild Strombolian eruptions. However, this ongoing activity is often interrupted by effusive eruptions (Andronico et al., 2005; Branca and Del Carlo, 2004, 2005), which typically occur at both the summit and along the rift zones on the volcano's flanks, and by intense explosive paroxysmal lava fountaining episodes (Andronico et al., 2021b) that can last from a few hours to over 24 hours. Paroxysmal lava fountain episodes are among the most energetic manifestations of Etna's activity, whose occurrence become more frequent during the past four decades (Behncke et al., 2014; Calvari et al., 2022a; Clocchiatti et al., 2004; Corsaro et al., 2017; Corsaro and Miraglia, 2014; Corsaro and Pompilio, 2004; Edwards et al., 2018). These explosive events are capable to generate high eruptive columns up to 14 km (Corsaro et al., 2017) and are often characterized by sequences of events, with the most energetic sequence occurring recently during the 2020-2021 period (Aiuppa et al., 2023 or Chapter 9; Calvari et al., 2022a). These eruptive phenomena pose significant hazards, including lava flow damage to settlements, ashfall impacting air traffic and health and the potential for pyroclastic density currents and lahars, which threaten both human life and infrastructures.

CHAPTER 4

OVERVIEW OF THE OBTAINED RESULTS

4.1 Overview of SO₂ flux time series at Stromboli

Since June 2014, the flux of SO₂ at Stromboli has been continuously monitored by the UV Camera station located at Roccette, providing a daily measurement for 88% of the days until December 2024. Since the beginning of its installation, numerous eruptive events have occurred, many of which are recorded as clear signatures within the degassing time series (Figure 4.1). In Chapters 5, 6, and 7, I analysed the degassing dynamics derived from the UV Camera measurements in relation to these eruptive episodes, with the aim of gaining a deeper understanding of degassing and eruptive mechanisms at Stromboli, and of exploring the utility of the SO₂ flux as a potential eruption forecasting tool. Particular attention is also given to identifying and quantifying potential sources of uncertainty, in order to improve the reliability and robustness of SO₂ flux measurements.

Chapter 5 focuses on a comparative analysis of SO₂ flux measurements at Stromboli volcano over a nine-year period (2014-2022), obtained using the near-vent UV Cameras (~500 m from the vents) and distal (~2 km) scanning DOAS spectrometers (FLAME Network). The analysis reveals a systematic discrepancy between the two datasets, with the scanning DOAS fluxes being, on average, ~200% higher than those derived from the UV Camera. This offset is interpreted as the result of both UV Camera column amounts underestimation, due to limited plume coverage and radiative transfer effects in optically thick plumes, and DOAS-derived flux overestimation, likely caused by using wind speed without accounting for the incomplete atmospheric dilution of gas puffs during transport. To address these limitations, a novel integrated SO₂ flux dataset is proposed, combining UV Camera-derived plume velocities using optical flow with DOAS-derived column amounts, back-calculated at source using an experimentally derived plume dilution function. This approach aims to provide a more robust and representative SO₂ flux time series, less influenced by environmental and observational constraints.

Chapter 6 investigates the relationship between volcanic gas composition and the occurrence of major explosive events at Stromboli volcano. In this chapter, both SO₂ fluxes and nearly continuous measurements of CO₂ and SO₂ concentrations are analysed, collected between 2020 and 2023. Statistical analyses reveal a significant association between major explosions and periods characterized by low SO₂ concentrations and fluxes and high CO₂/SO₂ ratios. Based on these results, a volcanic gas-based composite indicator was developed, which, in retrospective analysis, successfully forecasts the majority of the events on timescales of weeks.

Chapter 7 examines SO₂ and CO₂ flux time series acquired at Stromboli prior, during and after its last eruption in July 2024. The analysis focuses on the interpretation and analysis of volcanic gas flux variations in the run-up to the eruptive event. Results show that the eruption was preceded and accompanied by a large escalation in CO₂ flux, with a comparatively smaller increase in the SO₂ flux. This behaviour suggests the eruption was preceded by a burst in deeply sourced magmatic gas release, consistent with the involvement of gas-rich magma ascending from greater depths and later erupted in the July 11 paroxysm. The findings highlight the diagnostic value of CO₂ flux monitoring for detecting deep degassing processes that may not be immediately evident from SO₂ measurements alone.

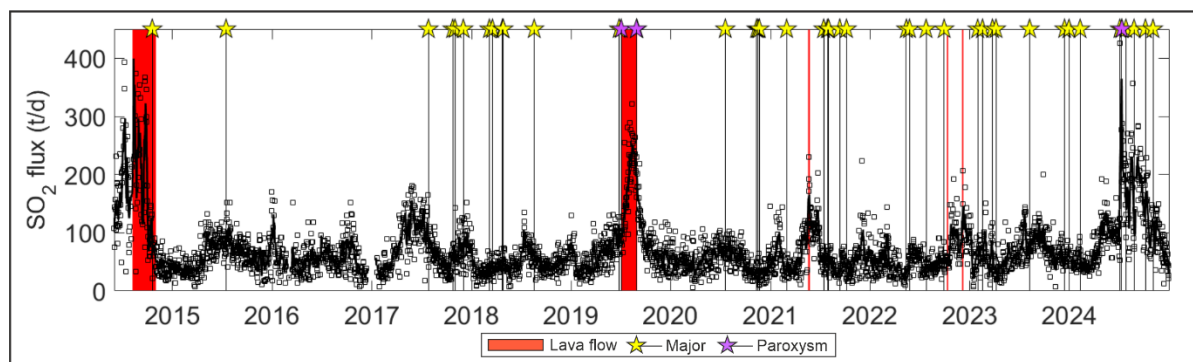


Figure 4.1 – Time series of daily average SO₂ flux measured by the UV Camera installed at Roccette (Stromboli). The black line represents a 7-day moving average. Red bars indicate lava flows, while yellow and purple stars mark the occurrence of major and paroxysmal explosions respectively.

4.2 Overview of SO₂ flux time series at Mount Etna

The UV Camera system at La Montagnola has been operational since June 2015, continuously recording data with a focus on the contribution from the Southeast Crater, which has been the main source of eruptive activity in recent years (Figure 4.2). Images processing provided a daily reliable measurement for 61% of the days until December 2024. During the observation period, Etna displayed a wide range of eruptive behaviours, alternating between quiescent phases, characterized by passive degassing and/or Strombolian activity, and more intense episodes, including effusive eruptions of varying magnitude and paroxysmal lava fountain events. Chapters 8 and 9 present an analysis of SO₂ flux variations associated with the main eruptive phases of the volcano, with a focus on the particularly intense paroxysmal sequences of 2020-2021.

Chapter 8 presents a nearly decade-long (November 2015 to 2024) record of SO₂ flux emissions at Mount Etna, derived from a permanent UV Camera system and combined with satellite-based thermal flux data. The analysis explores the relationship between SO₂ degassing dynamics and eruptive activity, resolving temporal trends across pre-, syn- and post-eruptive periods of major

eruptive phases. Despite variations in source location, eruption intensity, and instrumental limitations, the UV Camera system successfully detects clear SO₂ flux increases above background levels, quantified using a “success rate”, which shown detection efficiencies of 61%, 72% and 72% for the pre-, syn- and post-eruptive periods respectively, and improved performance during larger eruptive events. Coupling SO₂ flux-derived magma supply with thermal-derived magma output rates highlights a systematic volumetric imbalance between degassed and erupted magma, with only ~27% of the degassing magma discharged at the surface and the remainder (~73%) retained within the shallow plumbing system. This integrated gas-thermal approach emphasizes the strong variability of Etna’s eruptive behaviour and provides new insights into magma storage and transport dynamics.

Chapter 9 focuses on the analysis of SO₂ fluxes recorded at Mount Etna’s during the two major paroxysmal sequences that occurred between December 2020 and October 2021. Data from the permanent UV Camera system reveal a distinct increase in SO₂ emissions, rising from baseline levels (≤ 3250 t/day) during ordinary activity, reaching values ≥ 4200 t/day prior and during lava fountaining episodes. These degassing patterns are interpreted in the context of seismic and thermal data, supported by modelling of sulphur exsolution based on melt inclusion analyses. Results indicate that the onset of paroxysmal activity corresponds to a marked increase in magma supply rate exceeding 4 m³/s above the sulphur exsolution threshold (~150 MPa). These observations support the hypothesis that such eruptive sequences are triggered by the injection of volatile-rich magma into a shallow reservoir located approximately 3-4 km beneath the summit. The results underscore the dynamic nature of magma transport and degassing processes at Etna and highlight the importance of continuous gas flux monitoring to enhance the understanding of eruption precursors.

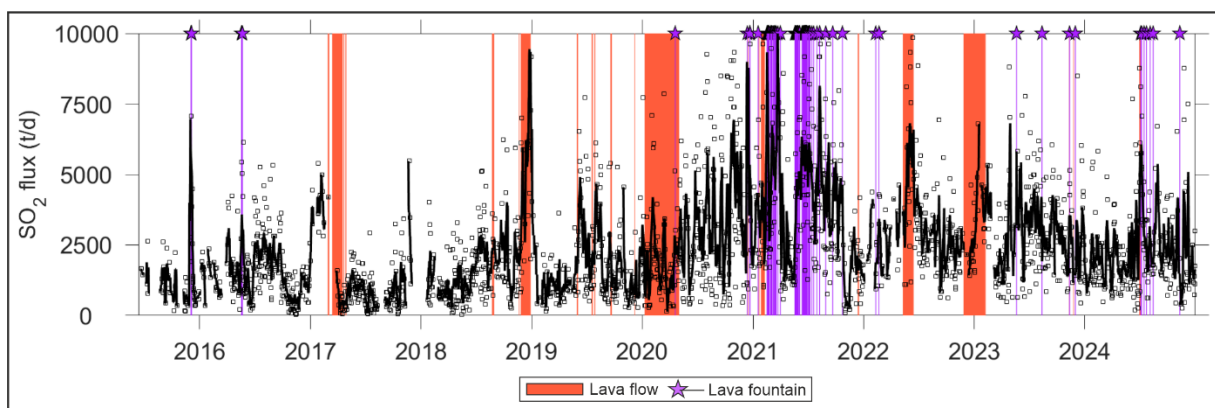


Figure 4.2 – Time series of daily average SO₂ flux measured by the UV Camera installed at La Montagnola (Mount Etna). The black line represents a 7-day moving average. Red shaded areas indicate lava effusions, while purple stars mark the occurrence of paroxysmal lava fountain episodes.

CHAPTER 5

IMPROVED VOLCANIC SO₂ FLUX RECORDS FROM INTEGRATED SCANNING-DOAS AND UV CAMERA OBSERVATIONS

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Abstract

Volcanic SO₂ flux is a key indicator of magma influx into shallower portions of magmatic plumbing systems, and as such is central to volcano monitoring. However, observations have traditionally been challenged by a variety of technical and methodological caveats and limitations, to overcome which it is required an intercomparison of different observational techniques and, where possible, their integration. Here, we compare ~9 years (2014 to 2022) of SO₂ flux records at Stromboli obtained through (i) a near-vent (~500 m) UV Camera system and (ii) a network of DOAS spectrometers scanning the distal (~2 km) bulk plume. We find a large (133 t/d on average) systematic offset between the SO₂ flux time series streamed by the two observational techniques, with the flux from the scanning spectrometers being ~200% higher on average than UV Camera flux. We propose this mismatch to derive from a combination of (i) SO₂ flux underestimation by the UV Camera, as caused by incomplete coverage of the plume (due to topography of the crater area) and radiative transfer issues in the optically dense, near-vent plume, and (ii) SO₂ flux overestimation by the distal scanning spectrometers', caused by non-ideal (incomplete) atmospheric dilution of source-released gas puffs during atmospheric transport. Our analysis suggests this latter process to be dominant, imparting a positive wind speed dependence and a marked seasonality to the distal scanning spectrometers' fluxes, and causing them to significantly overestimate the source SO₂ fluxes. Finally, we propose a novel integrated SO₂ flux record, based on the combination of UV Camera-derived gas velocities and DOAS-derived SO₂ integrated column amounts (back-calculated at source using an experimentally derived plume dilution function). We expect this SO₂ flux time series to be less affected by external factors (e.g., meteorological, illumination and volcano topography conditions) than using any of the two techniques alone and hence a better proxy of volcano behaviour. We

recommend testing of the integrated UV Camera-scanning-DOAS method at other volcanoes to explore its utility for improved volcanic degassing characterization.

5.1 Introduction

Measurements of volcanic gases provide critical information to constrain magma degassing processes, and their control on volcanic activity style (Edmonds, 2021; Edmonds et al., 2022; Fischer and Chiodini, 2015). Of the several gas-related parameters that are today monitored at volcanoes, the sulphur dioxide (SO₂) flux has long been known as an especially useful tracer of the rate of magma supply into (and degassing within) the shallower portions of volcano plumbing systems (Oppenheimer et al., 2011, 2014; Wallace and Edmonds, 2011).

Despite being less abundant in volcanic gases than the major species H₂O and CO₂, SO₂ exhibits very low natural atmospheric abundances and strong absorption features in the ultraviolet region of the electromagnetic spectrum, making it easily measurable by spectroscopic remote sensing methods (e.g., Kantzas and McGonigle, 2008; Oppenheimer et al., 2011). The SO₂ flux has thus been increasingly implemented in volcano monitoring programs since the development of the correlation spectrometer (COSPEC) in the 1970s (e.g., Millan et al., 1976; Moffat and Millan, 1971; Newcomb and Millan, 1970). In the early 2000s, the COSPEC was progressively abandoned after the advent of more compact and cost-effective CCD-based ultraviolet spectrometers (e.g., Galle et al., 2003), using the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt and Stutz, 2008). These new spectrometers coupled with spectroscopy method, as the DOAS technique, represented a major breakthrough in volcanology, as they paved the way to the implementation of the first fully automated networks of scanning spectrometers, today widely used at volcano observatories worldwide (e.g., Arellano et al., 2021; Edmonds et al., 2003; Galle et al., 2010; Salerno et al., 2009a, 2009b). Moreover, DOAS offers an intrinsically more accurate spectroscopic analysis of the acquired spectra (Platt and Stutz, 2008), it allows various chemical species to be quantified in a single spectrum, and - thanks to the availability of spectral information - helps reducing radiative transfer issues such as multiple scattering inside the volcanic plume, or light dilution effects (e.g., Kern et al., 2010a, 2012; McElhoe and Conner, 1986). Eventually, the advent of UV Cameras (Bluth et al., 2007; Burton et al., 2015; Mori and Burton, 2006) has provided the chance to measure volcanic SO₂ fluxes at increased temporal and spatial resolution. An especially useful advantage of UV Cameras is that they can output plume speed in near real-time (e.g., Bluth et al., 2007; Delle Donne et al., 2019; Peters and Oppenheimer, 2018), and that their much improved temporal resolution can allow imaging fast degassing phenomena (Dalton et al., 2010; Pering et al., 2015; Tamburello et al., 2012), and hence a

more effective integration between volcanic gas and geophysical datasets (Dalton et al., 2010; Delle Donne et al., 2017; Holland et al., 2011; Nadeau et al., 2011).

Use of scanning spectrometers and UV Cameras comes with both advantages and disadvantages (e.g., Johansson et al., 2009; Kern et al., 2010a, 2010b; Klein et al., 2017). Spatial and temporal resolutions of scanning-DOAS spectrometers are lower than with UV Cameras, but the ability to scan the plume from distance, and from different viewing directions, allows to fully capture (and characterise) the “bulk” gas plume. Disadvantages exist, however, especially arising from assumptions that must be made on (i) geometry and height of the plume (Johansson et al., 2009; Salerno et al., 2009a) and, most importantly, (ii) plume speed (e.g., McGonigle et al., 2005a, 2005b). Although techniques in principle exist to derive plume transport speed directly from spectrometers (McGonigle et al., 2005a, 2005b; Williams-Jones et al., 2006), applying these methods in real time to data streamed by permanent scanning networks has revealed impractical or even impossible (Arellano et al., 2021), forcing the adoption of alternative solutions, especially the use of wind speed (from meteorological models or anemometers) as a proxy for the real plume velocity. Use of wind speed as a proxy for plume speed may, however, be unjustified or even erroneous (e.g., Arellano et al., 2021; Barrington et al., 2022; Salerno et al., 2009a) and can lead to errors in the average of 10-40% but up to >100% in flux measurements (e.g., McGonigle et al., 2005a, 2005b; Williams-Jones et al., 2006).

UV Cameras, in contrast to scanning spectrometers, are typically deployed near the active vents (e.g., Delle Donne et al., 2017; Ilanko et al., 2020; Wilkes et al., 2023), and measure the two-dimensional SO₂ distribution in the plume with a greater spatial and temporal resolution, although with reduced spectral information which might lead to large uncertainty (Kern et al., 2010a, 2010b). The development of permanent UV Camera systems that directly apply optical flow algorithms to the acquired images (e.g., Delle Donne et al., 2017, 2019, 2022; Gliß et al., 2017; Peters and Oppenheimer, 2018), allows estimating the plume vertical velocity profile at source, where turbulence (and other atmospheric effects) have little (or no) effect. This significantly reduces the errors associated with plume speed (and hence flux) measurements (Delle Donne et al., 2019). However, the proximity to the active vents often causes the plume to be only partially imaged by UV Cameras, due to morphological complexity of volcano summits or unfavourable wind directions (Delle Donne et al., 2022). Furthermore, targeting the proximal and (often) most optical opaque portion of volcanic plumes cause UV Cameras measurements to be more prone to errors related to radiative transfer issues (Kern et al., 2010a, 2012, 2013).

The several caveats and limitations of both scanning spectrometers and UV Cameras, discussed above, suggest that a systematic intercomparison study between the two techniques is now crucially

needed. Here, we tackle this task by presenting a systematic comparative analysis of two long-term (9 years) SO_2 flux time series, taken at Stromboli volcano (in the Aeolian archipelago, Sicily; Figure 5.1) from (a) a proximal (~ 500 m from source) permanent UV Camera system and (b) a distal (~ 2 km from source) network of scanning-DOAS spectrometers. Our aim is to show that the integrated use of UV Camera and scanning spectrometers can contribute to improved volcanic SO_2 flux records than possible with any of the two techniques alone.

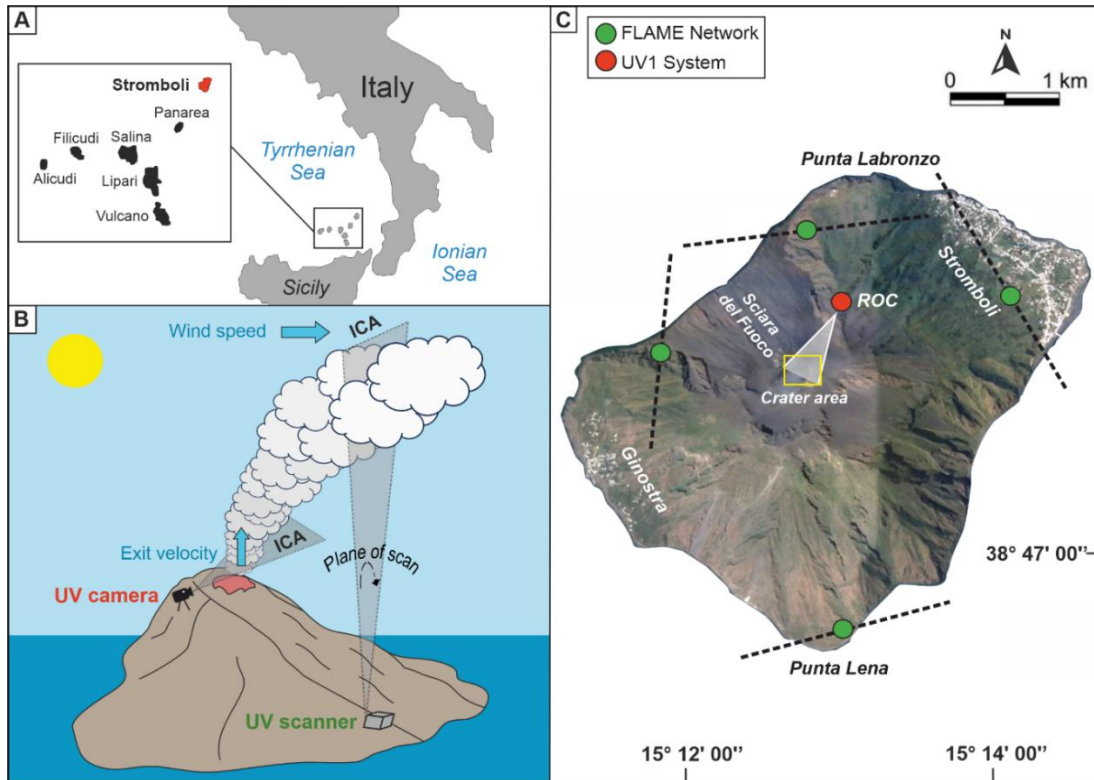


Figure 5.1 – A) Location of Stromboli Island; B) Sketch of the main parameters that take part in the calculation of the SO_2 flux with scanning spectrometers and UV Cameras. Not in scale. Gray areas represent examples of plane of scan of UV scanner spectrometers and field of view (FOV) of UV Camera. C) Location of the scanning spectrometers and position of the UV Camera system at Rocette (ROC). Black dashed line indicates the UV scanner's plane of scan; white solid lines enclose the UV Camera's FOV.

5.2. Methods

Our analysis here is based on the inter-comparison of two independently derived SO_2 flux time series, obtained at Stromboli from data streamed by two distinct monitoring networks (see Figure 5.1): i) a network of 4 UV scanning spectrometers (here referred to as FLAME network, managed by INGV-OE; Burton et al., 2009; Salerno et al., 2009b) and ii) a permanent UV Camera system (here referred to as UV1, managed by UniPa; Delle Donne et al., 2017, 2022). Both instrument types derive the SO_2 flux as the product of the SO_2 integrated column amount (ICA, integrated over a plume cross-section perpendicular to the plume transport direction) multiplied by plume speed:

$$SO_2 \text{ Flux} = SO_2 \text{ Integrated Column Amount (ICA)} \times \text{Plume speed} \quad (\text{Eq. 5.1})$$

The two instrument types have distinct acquisition, processing, and ICA/plume speed calculation routines, as detailed below.

5.2.1 The scanning spectrometer DOAS - FLAME network

The Stromboli's FLAME (FLux Automatic MEasurement) network consists of four ultraviolet scanning spectrometers located near to the shoreline of the island, at ~2 km distance from the active vents (Figure 5.1-C). Each spectrometer is optically connected, via an optical fiber, to a telescope that views a 45° mirror mounted on a rotating head driven by a stepper motor. This allows the acquisition of a series of spectra along a planar arc above the automatic station, which is conveniently placed on the ground to intercept the degassing plume emitted from the volcano's summit. Bulk daily SO₂ flux measurements are performed when the wind disperses the volcanic plume above at least one of the instruments of the network. To perform a scan from a fixed position, it is necessary to characterize the geometry and height of the plume (e.g., Johansson et al., 2009). This is obtained by using a radiant geometry to reduce plume cross-sections and applying a correction when the plume is not directly above the scanner (Burton et al., 2009; Salerno et al., 2009a). In detail, for every scanning angle, the width of the arc of the scanning i^{th} is inverted by the product of the angle α in radiant subtending the scanning i^{th} and the distance between the scanner and the centre of gravity of the total measured scanned-profile. The constant radius for the calculation allows to avoid obtaining long steps between measurement points near the horizon, but implicitly forces the plume into a curved shape. This approach was adopted as the alternative linear geometry might introduce much larger errors for broad flat plumes and/or when the plume is close to the horizon, as the calculated lengths between each point in the cross-section become unfeasibly large. Once the scan cross-section is inverted, a correction for the orthogonality respect of the normal component of the wind-plume transport speed is carried out by considering the cosine of the angle between the plume direction and the scanner-crater direction (see Salerno et al., 2009a for further details). The importance of constraining the height of the plume in stationary scanning applications has been well explained by Edmonds et al. (2003). In Montserrat, this issue was solved by geometry using the intersection of two SO₂ maximum concentration vectors measured by two scanners. However, on Stromboli this is more challenging due to the highly variable wind direction; hence, the height of the plume was obtained by experimentally establishing the (linear) dependence of plume height on wind speed. Direct observations of the volcanic plume showed that for wind speed up to 5 m/s the plume can rise 100-200 m above the crater terrace and for speeds up to 20 m/s the plume descend and could be grounding. Therefore, we used a combination of empirical comparison with traverse measurements and

observations of pairs of scans from different stations that observed the same plume at various wind speeds, to deduce a relationship between plume height and wind speed, with height decreasing as wind speed increased. The initial estimates for the relationship between plume height and wind speed were refined empirically through comparison with traverse data collected over 3 months in 2007 (see Burton et al., 2009 on the approach and validation of the plume height inversion at Stromboli).

SO₂ column amounts are retrieved in real time processing the recorded open-path UV spectra using the DOAS technique applying a modelled reference spectrum rather than a measured one, to avoid complications that arise when the plume encompasses a wide part of the scan arc and no clear sky spectra are available (Salerno et al., 2009b). Integrated column amounts recorded across a scan plume are then multiplied by the estimated plume-transport speed to determine the flux. Finally, individual fluxes are summed over the whole plume to obtain the total flux for a given scan in metric tonnes per day. Daily bulk SO₂ flux are obtained by averaging all the measurements recorded over the day. Wind speed is retrieved from the LAMI meteorological model (Limited Area Model Italy) from the European Center for Medium-Range Weather Forecasts (ECMWF) to obtain SO₂ flux. This is a mesoscale forecast model nested in the European Centre for Medium-Range Weather Forecasts (ECMWF, <http://www.ecmwf.int/>). It has a 7 km grid spacing and 35 vertical terrain-following levels. LAMI gives an output every 3 h and produces 48 h forecast once a day (e.g, Doms and Schättler, 1999).

Uncertainties in SO₂ flux by stationary automatic scanning array arise from several sources, such as DOAS retrieval using a modelled reference spectrum, multiple scattering effects, plume speed and height of the plume. The sum of the individual errors yields a total uncertainty between -22 and +36% (Salerno et al., 2009a).

5.2.2 The permanent UV Camera – UV1 system

The permanent and fully autonomous UV Camera system dedicated to the detection of SO₂ has been installed in May 2014 in the Roccette Site (ROC in Figure 5.1-C), on the northeastern upper flank of Stromboli, approximately 750 m above sea level and facing the northernmost craters. This site, located at only 500 m NE off the active vents, offers an excellent view of the northeastern sector of the crater terrace, while potentially missing a substantial (~30%; Delle Donne et al., 2022) fraction of the gas emitted by the partially hidden South-West and Central craters (see also Aiuppa et al., 2021).

The UV Camera system is designed to output high-rate (0.5 Hz) long-term SO₂ flux records in continuous mode for ~6 hours per day. It is equipped with two cameras sensitive to UV-radiation fitting two distinct bandpass optical filters. The absorption of sunlight by SO₂ is significantly reduced

at ~ 320 nm (Vandaele et al., 1994) and the two filters operate respectively in the range 300-320 nm (where there is maximum absorption) and 320-340 nm (beyond the absorption region). The first filter measures the optical density of SO₂, while the second one is used to compensate for aerosol scattering (Dalton et al., 2009; Kantzas et al., 2010; Kern et al., 2010b; Mori and Burton, 2006). A detailed list of components and their specifications is provided in Delle Donne et al., (2017, 2019, 2022). Couples of simultaneously acquired images are post-processed by merging them into a single absorbance image, applying the Lambert-Beer law. This process enables the quantification of the relative absorption of scattered UV sunlight by any volcanic SO₂ present within the cameras' field of view. Data from a collocated UV spectrometer (Ocean-Optic USB 2000+), along with periodic field calibration campaigns using gas cells of known SO₂ concentration, allow calibration the UV Camera images (Delle Donne et al., 2017). Column densities over the entire images are then obtained by applying a proportionality ratio between absorbance and SO₂ column density (McGonigle, 2007). For calibration of UV Camera images, see also Lübcke et al. (2013). Measurement principles of the UV Camera station are detailed in Kantzas et al. (2010), Kern et al. (2010b), Mori and Burton (2006).

Plume speed is calculated by applying the Lucas and Kanade optical flow algorithm (Lucas and Kanade, 1981) to sets of consecutive images. This involves an automatic selection of high intensity pixels within the plume showing high spatial coherence in consecutive frames. Plume speed profiles, in both horizontal and vertical directions of movement are derived by averaging the calculated two-dimensional velocity field and filtering out velocity points with low coherence (Delle Donne et al., 2017). Tests conducted by applying the algorithm to artificial images with known particles velocities indicate errors in the inferred velocities to be <5% (Delle Donne et al., 2019). It is important to consider that our plume speed measurements are conducted on a restricted image portion, capturing rectangular subregions located just above the crater rim (see Supplementary Figure S5.1 and S5.2). In this near-vent region, the SO₂ absorption signal is maximized while atmospheric effects, such as air entrainment in the plume and complexities in the local wind field, are minimized. This is proven by careful analysis of plume transport vectors inferred by the algorithm, showing that nearly vertical ascent of hot, buoyant gas puffs predominates over transport caused by the local wind field.

A SO₂ flux density profile is then obtained from combination of plume speed and SO₂ column density profiles. This SO₂ flux density profile is calculated along an ideal section linking the centers of the rectangular subregions and by multiplying SO₂ column density profile by the normal component of the velocity profile relative to this ideal section. Finally, a 0.5 Hz frequency step of SO₂ column amount and total flux are determined by integrating respectively the column densities and the density flux over the total length of the profile.

SO₂ daily fluxes are obtained averaging fluxes associated with best-viewing conditions during the day, applying a visibility index, calculated as the unsigned ratio between the mean pixel intensity associated with the camera FOV's portions capturing sky and ground, respectively (Delle Donne et al., 2019). SO₂ measurements are then selected by setting a threshold on the visibility index and discarding measurements below the threshold. The entire process described above is performed using a MATLAB[®] code, that provides time series of SO₂ flux in near real-time (Delle Donne et al., 2017, 2019). The main advantage of the UV Camera system is that the plume is directly imaged at its emission point, and the real motion of the vertically rising gas thrust identified and quantified. As driven by momentum and buoyancy, movement of the gas fronts is influenced by gas characteristics (e.g., temperature, density) while remaining less affected by the local wind speed field.

There are many potential sources of errors that can contribute to biases in UV Camera derived SO₂ fluxes. Broadly speaking, a main source of bias is the presence of ash and aerosols in the plume and in the atmosphere that can mask the SO₂ signal. While attempts are made at minimizing the impact of ash/aerosols by use of our automatic algorithm that discards images associated with cloudy conditions and/or periods with extremely low SO₂ signal to noise ratio (see Delle Donne et al., 2019), intrinsic errors caused by radiative transfer issues do still persist, especially when high SO₂ column densities and aerosol optical thickness are encountered (Kern et al., 2012, 2013).

The ideal condition for the remote sensing of volcanic plumes using scattered light would one in which photons detected by the instrument pass the plume in an idealized straight line (Kern et al., 2012, 2013). However, in real field conditions, volcanic plumes are often not transparent, but rather partly to visibly opaque because of condensation water droplets and aerosols. These, along with volcanic ash, cause complex scattering effects of photons, substantially altering the plume optical path length. In particular, for high aerosol loads and in opaque plumes (as in the near vent field), multiple scattering becomes wavelength dependent (Kern et al., 2010a, 2012, 2013) and, at wavelengths of 300-310 nm, the average photon paths in the plume are typically much shorter, e.g., short paths that only penetrate the plume outer boundaries become major contributors to the measured signal. This effect can cause a general underestimation of the UV-camera sensed SO₂ fluxes. One additional point to consider is that some photons may also be scattered in the direction of the instrument without having passed through the plume at all. Since these photos do not contain spectral absorption structures, they generate a dilution of the measured column density. This so called 'light dilution' effect is mainly controlled by the distance of the observation point from the target plume (Campion et al., 2015; Kern et al., 2010a).

In light of the above, and following Kern et al. (2010b), we identify the following main intrinsic sources of error in our UV Camera-based SO₂ sensors: (i) the imperfect correction of

scattering/absorption effects, (ii) changing in background spectral intensity, (iii) non-perpendicular illumination of filters and (iv) uncertainty related to radiative transfer effect (see Table 1 in Kern et al., 2010b). This latter point is especially critical, as UV Camera data alone are inadequate for fully evaluating the role of the many possible radiative transfer effects (see above). From the results of Kern et al. (2010b), we estimate the sum of these individual errors, as square root of the sum of the squares of the individual errors, to contribute to a total minimum uncertainty of $\sim \pm 32\%$.

5.2.3 Volcanic activity and seismic tremor

To explore the relationship between our derived SO₂ flux time series and volcanic activity state, we compare our gas results with daily volcanic tremor records taken by the Laboratorio di Geofisica Sperimentale (LGS) of the University of Firenze, and a chronology of non-ordinary eruptive events (e.g., those deviating from the regular, persistent strombolian activity; Calvari and Nunnari, 2023). These include 2 two long-lived effusions (August 6- November 13, 2014 and July 3-August 31, 2019; Giudicepietro et al., 2020; Mattia et al., 2021; Valade et al., 2016; Zakšek et al., 2015), 32 brief (minutes/hours) lava overflows from the summit craters, and 31 larger than normal (Bevilacqua et al., 2020; Métrich et al., 2021; Rosi et al., 2013) explosions (2 paroxysms and 29 major explosions: data taken from INGV and LGS monitoring reports, publicly available at (<https://cme.ingv.it/bollettini-e-comunicati/bollettini-ingv-stromboli>; <https://cme.ingv.it/bollettini-e-comunicati/categoria-1>)).

Persistent degassing at active volcanoes is typically associated with volcanic tremor (e.g., Konstantinou and Schlindwein, 2003; McNutt, 2002; Williams-Jones et al., 2001). The correlation between SO₂ and tremor has been explored in different volcanic systems (Delle Donne et al., 2022; Edmonds et al., 2003; Patanè et al., 2013; Zuccarello et al., 2013), and increases in tremor amplitude are typically observed prior to and during effusive eruptions, and synchronously with explosive activity (e.g., Aiuppa et al., 2023; Bonaccorso et al., 2011; Brandsdóttir and Einarsson, 1992; Cannata et al., 2008; Fischer et al., 1994; Nadeau et al., 2015; Ripepe et al., 2009; Valade et al., 2016). The physical processes generating volcanic tremor are thought to be linked to the unsteady mass transport of magma dynamically interacting with the surrounding rocks (e.g., Steinberg and Steinberg, 1975). Nevertheless, various other mechanisms (e.g., Benoit and McNutt, 1997; Gordeev, 1993) and models have been proposed as potential sources of tremors (e.g., Aki et al., 1977; Chouet et al., 1987), depending on the specific characteristics of individual volcanoes and eruption styles. Continuous volcanic tremor at Stromboli has been attributed to sustained degassing and bubble coalescence within the shallow conduit (Ripepe et al., 2002; Harris & Ripepe, 2007; Ripepe and Gordeev, 1999) due to the viscoelastic response of the magma, which reacts to the pressure decrease that occurs as

gas bubbles expand during depressurization (Chouet et al., 1987; Ripepe and Gordeev, 1999). Ultimately, volcanic tremor is closely linked to degassing processes within the magmatic column, and its temporal increases are typically associated with phases of escalating magmatic activity (e.g., Aiuppa et al., 2023; Delle Donne et al., 2022).

The volcanic tremor here reported is the daily average, at ROC station, of RSAM estimated in a time window of 60 sec and in the frequency band 0.9 – 10 Hz which includes the main spectral content of seismic tremor at Stromboli (Chouet et al., 1997; Ripepe and Gordeev, 1999). This frequency band enhances the seismic signals induced by the volcanic activity avoiding to affect the tremor amplitude from remote tectonic earthquake, sea storm which have a typical frequency smaller than 0.9 Hz and from rockfall events characterized by a frequency content larger than 10 Hz (Giudicepietro et al., 2023). We therefore use volcanic tremor as an independent proxy for volcanic activity level, also considering it is poorly influenced by atmospheric disturbances that impact volcanic gas observations.

5.3. Results

The SO₂ daily averaged fluxes, obtained by processing of the UV Camera and scanning-DOAS records during 2014-2022, are compared to each other in Figure 5.2-A, and with tremor amplitude in Figures 5.2-A and 5.2-B. We find a large mismatch between the two SO₂ time series, both in terms of absolute values and (often) in temporal fluctuations. The daily SO₂ fluxes derived from UV1 have a mean of 67 ± 49 t/d (range: 6-572 t/d) and are therefore systematically lower than those obtained from FLAME (mean: 200 ± 92 t/d; range: 22-720 t/d) (here and thereafter, mean values are reported with their standard deviations). The two fluxes hence diverge by 133 ± 95 t/d on average. This corresponds to a circa +200% mismatch (calculated as $100 \cdot 133 / 67$).

The daily SO₂ fluxes obtained by the two methods show no evident correlation ($R=0.20$; Figure 5.2-C). The correlation improves only slightly ($R=0.34$) if long term-trends (30-day mobile averages) are compared instead (see also Supplementary Figure S5.3). The Pearson Correlation Coefficient (R) between the two flux time series increases with increasing UV1 fluxes, while no correlation is found with the FLAME-derived fluxes (Figure 5.2-D). From this evidence we infer that the two measurements show coherent fluctuations mainly when UV1 fluxes increase.

Correlation with volcanic tremor (Figure 5.2-C) is best for UV1-derived SO₂ fluxes and is much improved if monthly averaged values are used instead of daily averages (R values of 0.61 and 0.44, respectively). The FLAME-derived flux is less correlated with tremor, both on a daily ($R=0.25$) and monthly ($R=0.39$) basis. Both SO₂ fluxes coherently increase with volcanic tremor in correspondence with the two major effusive eruptions (2014 and 2019) and, although less apparently, during some of

the (minor) lava overflown events (purple stars), suggesting both parameters respond to (and are suitable tracers of) escalating transport of bubbly magma in the shallow conduit (Ripepe et al., 2005).

Since, for both methods, the fluxes are calculated as a product between ICAs and transport velocities, we analyse these two components separately to explore the factors that lead to differences between fluxes.

The SO₂ ICAs time series (Figure 5.3, middle panel) independently returned by the two methods are broadly consistent, with the FLAME ICAs (mean: 0.58 ± 0.46 kg/m; range: 0.04-5.19 kg/m) being ~32% higher on average than the UV1 ICAs (mean: 0.44 ± 0.22 kg/m; range: 0.07-2.15 kg/m).

The plume velocities used by the two methods are radically different (see Figure 5.3, bottom panel). The optical flow algorithm used by UV1 returns a mean plume speed of 1.73 ± 0.66 m/s (range: 0.39-7.59 m/s), while the mean velocity used by FLAME is 5.90 ± 4.17 m/s (range: 0.48-25.18 m/s). This mismatch is not surprising considering that the velocities used by the two methods correspond to different properties, as the UV Camera directly estimates the vertical motion of the volcanic plume at exit (e.g., right above the vents) with the FLAME adopts an independently estimated (from meteorological models) horizontal wind speed at plume altitude. Note the plume is intercepted by the scanning spectrometers at ~2 km downwind from that imaged by the UV Camera (Figure 5.1).

The exercise illustrated in Figure 5.3 indicates the SO₂ flux discrepancy is dominantly determined by a plume vs. wind speed inconsistency between the two methods (see histograms in Figure 5.3-B). This is additionally supported by Figure 5.4-A, showing that the flux ratio (expressed as FLAME/UV1) exhibits periodic fluctuations that parallel the seasonal fluctuations in wind speed from FLAME (wind speed is typically stronger during the Sicilian winter season). The FLAME/UV1 flux ratio is indeed correlated with wind speed ($R=0.58$), implying the difference between the two methods is governed by external (non-volcanogenic) factors. Importantly, the SO₂ ICA ratio is not affected by a seasonal periodicity (Figure 5.4-B) and is poorly correlated with wind speed ($R=-0.35$). Hence, isolating the ICAs measured with scanning-DOAS (e.g., removing the wind component from the flux measurements), we find a good correlation between the FLAME's ICAs and the UV1 flux ($R=0.60$; Figure 5.4-C).

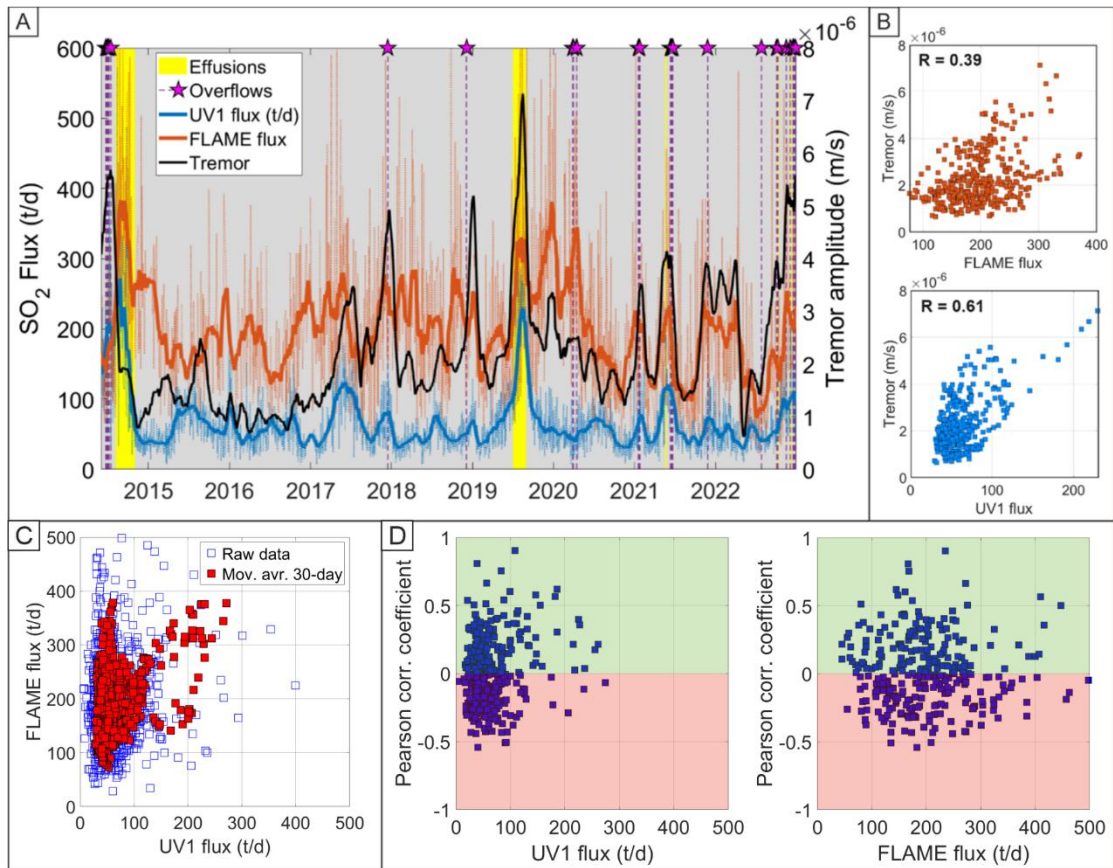


Figure 5.2 – A) Daily-averaged SO₂ fluxes measured from UV Camera (UV1 system) and scanning spectrometers (FLAME network) with volcanic tremor estimated in the frequency band 0.9 – 10 Hz. Solid lines represent a 30-day moving averages. Effusions and overflows are also reported. B) Scatter-plot showing the correlation between fluxes and volcanic tremor. C) Scatter-plot between the two fluxes, comparing the R values for both raw data and 30-days averaged data. D) Rolling correlation of Pearson Correlation Coefficient (in a 90-day centred window) with UV1 and FLAME SO₂ fluxes. The red zone marks the anti-correlation ($R < 0$), while green zone marks the positive correlation ($R > 0$). The two measurements exhibit coherent fluctuation mainly when UV1 fluxes tend to higher values.

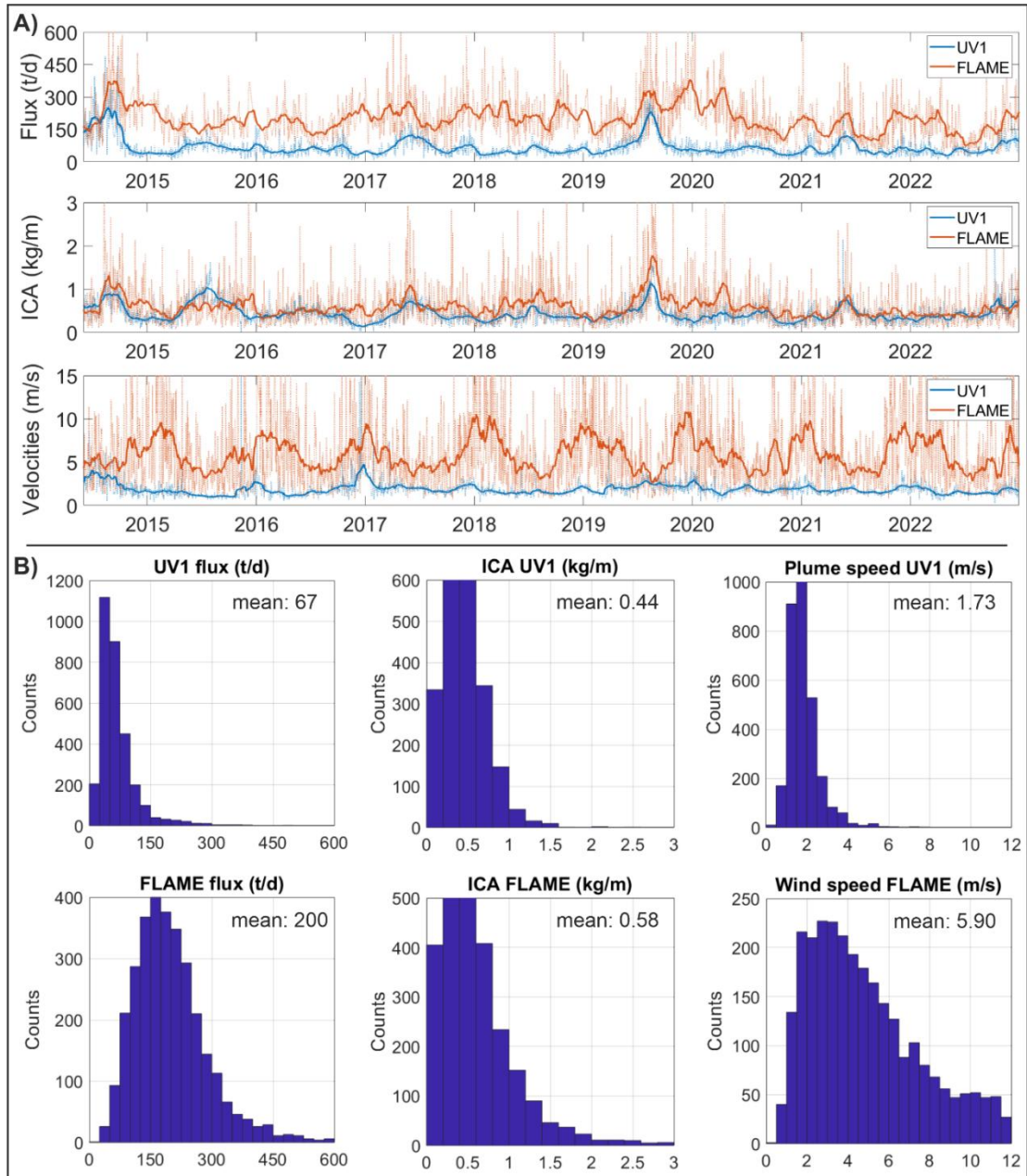


Figure 5.3 – A) Time series of measured SO_2 fluxes and their components, with their 30-day moving averages. Panel on top shows the SO_2 fluxes (t/d), panel in the middle shows the Integrated Column Amounts of SO_2 (ICA, in kg/m) and the bottom panel shows the velocities (m/s). B) Histograms of time series reported in panel A, with their mean values. The SO_2 flux discrepancy is mainly determined by differences in utilized velocities between the two methods.

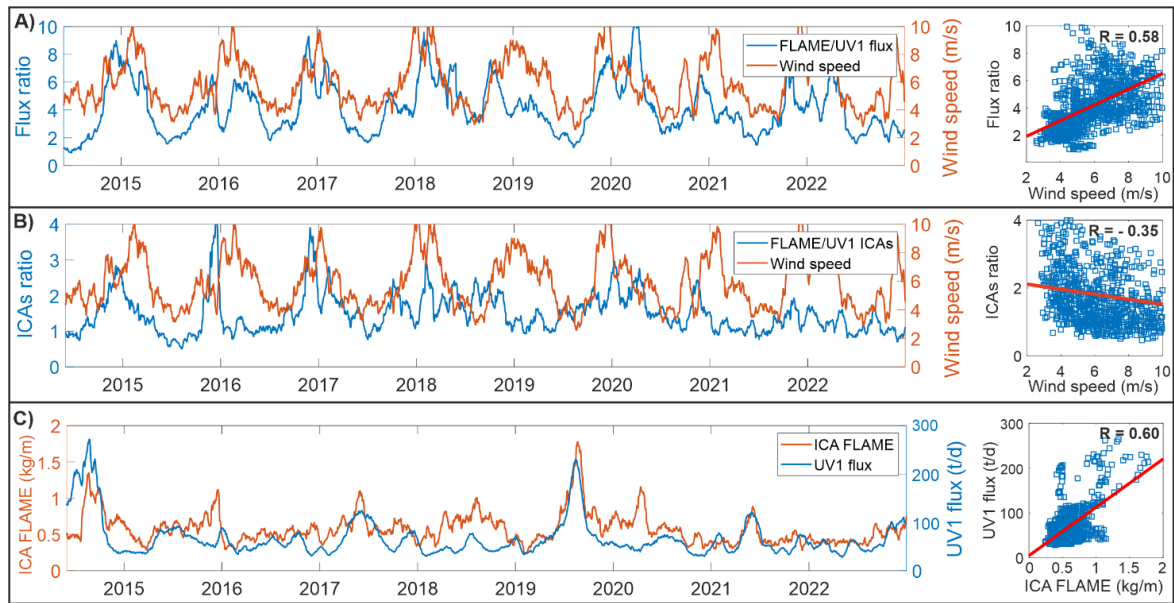


Figure 5.4 – A) Ratio between daily-averaged SO_2 fluxes calculated as FLAME over UV1 and wind speed from LAMI meteorological model used by FLAME to derive the SO_2 fluxes (30-day averaged time series). The individual daily-averaged fluxes can differ from each other by up to 10 times (see flux ratio, blue line), with a mean of 4 during all the analysed period. Flux ratio displays cyclic fluctuations that match the seasonal fluctuations in wind speed. B) SO_2 Integrated Column Amounts (ICA) ratios estimated for both methods with wind speed (30-day averaged time series). ICA ratio is not affected by seasonal periodicity and is poorly correlated with wind speed. C) FLAME's ICAs with flux from UV1. Isolating the FLAME SO_2 column amounts (thus removing the wind component), these show a good correlation with UV1 flux.

5.4. Discussion

5.4.1 Causes of the SO_2 flux discrepancy

The Stromboli's UV Camera and the FLAME scanning-DOAS network observe during daytime the same atmospheric plume (Figure 5.1) but from different distances. Within the range of their relative uncertainties, they should - at least in principle - output consistent SO_2 flux results. Our 9-years intercomparison study, instead, finds the two fluxes to be remarkably different (Figures 5.2, 5.3), with the FLAME flux being systematically higher (by ~ 133 t/d on average, and up to 10 times as a maximum) than the UV1 flux (Figure 5.4-A). The temporal fluctuations of the two flux time series are also poorly correlated on a daily/monthly basis (Figure 5.2-C). However, if we split the entire observational periods into subperiods characterised by specific volcanic activity styles (Figure 5.5), and calculate flux averages for these sub-intervals, we find the FLAME vs. UV1 fluxes to align along an almost linear trend of 0.92 slope (correlation coefficient $R=0.91$). The significant slope of this line (~ 1) suggests that the difference between the two measurements is attributable to a systematic error resulting in a fixed offset. The two observational networks, therefore, coherently respond to changes in volcanic activity (Figure 5.5), e.g., their derived fluxes consistently increase from their mean values (white filled square) to pre- and post-effusive phases (yellow and orange squares), and

then peak during effusive periods (red square). Yet, the 119 t/d intercept of the best-fit regression line confirms the large discrepancy between the absolute FLAME vs UV1 fluxes (Figures 5.2, 5.3, 5.5).

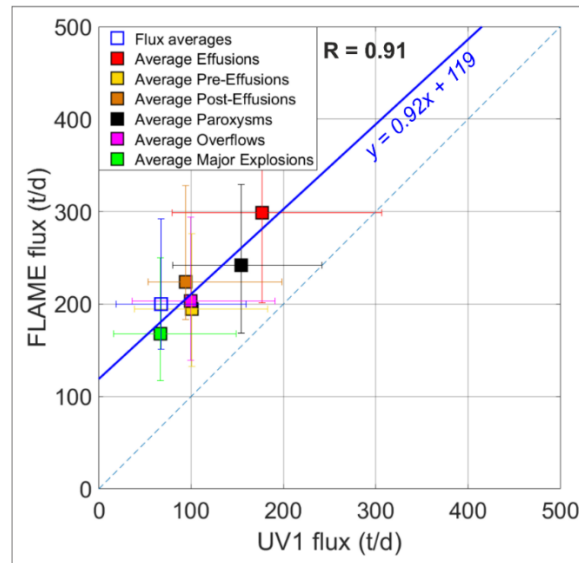


Figure 5.5 – Averaged SO_2 fluxes associated with specific volcanic activity styles. These subset fluxes are calculated by averaging the SO_2 flux results obtained: (i) in the 30 days prior (pre-effusions) and after (post-effusions) the effusive events, (ii) during days related to effusions (red square), and (iii) in the 14 days windows centred on all catalogued overflows, major explosions and paroxysms. We used a larger window for effusions (30 days) due to their longer duration, as phenomena involving larger volumes of magma, whose increasing/decreasing trends in SO_2 fluxes are generally observed over longer time intervals. Empty blue square identifies the average of all FLAME and UV1 daily fluxes. These activity style specific fluxes for FLAME and UV1 values are linearly correlated.

There are several factors that can potentially explain the flux discrepancy observed, especially considering that the two techniques operate differently, and are applied to distinct portions of the volcanic plume. These are discussed below.

5.4.1.1 Topographic effects and radiative transfer issues

One first possible cause of mismatch is related to the interplay between the position of the UV1 camera system and the morphology of the crater area (Figure 5.1). In contrast to the FLAME scanners that can fully scan the entire bulk plume from ~2 km distance, our UV Camera observations are taken from a proximal (~500 m from source) site that offers an incomplete view of the plume because of geometrical constraints (UV Camera viewing direction and field of view) and topography of the summit crater region. These cause the plume issuing from the central and southwestern craters to be partially hidden by the northeastern crater ridge (Delle Donne et al., 2017). In a dedicated study in which two UV Cameras were used, viewing the plume from different (roughly opposite) viewing directions (Delle Donne et al., 2022), the SO_2 flux from UV1 was found to underestimate the total

flux by $\sim 30\%$ on average (weekly averaged fluxes obtained by the two camera systems differed one each other by $28 \pm 20\%$, up to a maximum of 79%). It is well possible that such topography related underestimation of the UV1 SO_2 flux increases during days of especially strong wind, when (as seen elsewhere; Bonadonna et al., 2015) the plume is forced at ground level and/or channelled behind the crater(s). However, during 1 year of observations, taken in a variety of wind fields (including a number of strong wind days), Delle Donne et al. (2022) never found the topographic effect on the UV1 flux to exceed a factor 79% . Moreover, visual inspection of the UV Camera images indicates the FLAME/UV1 offset (Figure 5.2) to persist independently of plume geometry, e.g., in both days of vertically rising plume during calm wind and days of bended plume under strong wind. Also, there is no evidence of strong plume transport speeds in our UV1-based record (Figure 5.3-A), nor is the ratio of ICAs correlated with wind speed (Figure 5.4-B), indicating that the observed mismatch cannot be attributed to an enhanced topographic effect during strong wind days. All this taken together, it is reasonable to conclude that the topographic effect can account for only a small fraction ($\sim 30\%$) of the far-larger observed FLAME/UV1 offset ($\sim 200\%$; Figures 5.2-A, 5.3).

In addition to topography, there is a radiative transfer effect to consider that would result from the denser, and hence more optically opaque plume imaged by UV1, if compared with the distal (and more transparent) distal plume detected by the FLAME network. The effects of radiative transfer are a complex function of distance between source and observation point, plume SO_2 column densities, and aerosol optical thickness (Kern et al., 2012, 2013). In particular, the light dilution effect scales with distance of the observation point from the target plume, and can lead to SO_2 flux underestimations of several tens of %. Campion et al. (2015) estimated for Stromboli a 25% SO_2 underestimation due to light dilution for a target plume at 2.2 km distance. Applying the same equation of Campion et al. (2015) to the more proximal plume sensed by the UV1 camera system (distance from target plume, ~ 500 m), we find the light dilution effect to potentially account for only $\sim 10\%$ of the measured camera pixel intensities (Delle Donne et al., 2022). Given this, we exclude therefore that light dilution can explain the FLAME flux excess over the UV1 flux. In plume scattering can, at least in principle, cause a severe SO_2 flux underestimation in the dense, near-vent UV1 plume. However, in-plume scattering becomes significant in ash-laden plumes (a condition automatically excluded from analysis in our case from use of the visibility index; cfr. 5.2.2) or for SO_2 column densities exceeding $2000 \text{ ppm}\cdot\text{m}$ (Kern et al., 2013). These conditions (SO_2 column density $>2000 \text{ ppm}\cdot\text{m}$) are reached in the UV1 Stromboli plume only during the short-lived strombolian explosions that contribute little to the overall gas budget (Pering et al., 2020; Tamburello et al., 2012). In contrast, during passive degassing and/or puffing (the dominant modes of gas release at Stromboli; Tamburello et al., 2012), the SO_2 column densities range from $<<1000 \text{ ppm}\cdot\text{m}$ up to

circa 2000 ppm·m (see Supplementary Figure S5.1 for a snapshot of typical range of column densities by UV1). In these conditions we considered the typical error budget related to radiative transfer effects on UV Camera measurements to be ~30% (see Table 1 in Kern et al., 2010b), well below the ~200% factor that would be required to account for the FLAME/UV1 flux mismatch.

Finally, we conclude that network geometry and radiative transfer issues, although certainly playing a role, cannot justify alone the large SO₂ flux mismatch observed between the two methods.

5.4.1.2 Plume transport and dilution behaviour as the key factors

Although topography and radiative transfer effects (5.4.1.1) do certainly contribute some systematic offset between the two time series, they can hardly explain the strong seasonality of the FLAME/UV1 SO₂ flux mismatch, and especially its strong dependency on wind speed (Figure 5.4-A). As observed in Figure 5.3, the discrepancy between the two flux time series mostly reflects a difference in the used velocities, with the mean wind speed used by FLAME (5.90 m/s) being ~240% higher than the averaged plume speed detected by UV1 (1.73 m/s). Our observations then imply that the causes for the observed flux mismatch need to be searched in the processes that govern the transport behaviour of the plume, from the UV Camera measurement point (~500 m) to FLAME scanners (~2 km).

We initially explore the possibility that the mismatch is due to local modifications of the wind field during transport, as caused by interaction with the (complex) volcano topography (e.g., Favalli et al., 2004; Graziani et al., 1997), as documented by scanning spectrometer observations at other volcanoes (Nadeau and Williams-Jones, 2009; Williams-Jones et al., 2008). For example, Nadeau and Williams-Jones (2009) compared SO₂ emission results at Masaya volcano obtained through DOAS traverse taken at two distinct measurement sites located at different distances along the plume axis. They found SO₂ emissions from the two sections to differ significantly one each other. They noted the more distal measurement site to be located on a ridge at higher elevation than the volcano summit, over which the local trade winds are forced to accelerate. They then argued that the consequence of this wind speed acceleration was a plume dilution along its axis of propagation, resulting in smaller SO₂ integrated column amounts and hence lower SO₂ fluxes if wind intensity changes were not taken into account. They then concluded that use of a single plume speeds for different analysis sections, without considering the local topography, can lead to large flux discrepancies.

We argue that these topographic effects are minimised at Stromboli because of the relatively regular, cone-shaped morphology of the island. It is in principle possible that, during very strong winds, the plume is forced down along the Sciara del Fuoco depression, to hence stagnate and move

at slower rate than the wind speed at the volcano top. This may cause an over-estimation of the FLAME SO₂ flux if plume speed at the wrong altitude is used in the calculations. However, systematic observations of the video footages recorded by the several monitoring webcams operative on the island indicate that these extreme conditions are very rare at Stromboli, while the plume altitude normally oscillates within only a few hundred meters around the volcano top elevation. Since we focus here on long-term variations (monthly averages) over approximately 9 years of observations, the effects of rare vortical and anomalous events are significantly minimised.

We next test the hypothesis that the FLAME/UV1 SO₂ flux mismatch is caused by the transport behaviour of the volcanic plume upon its downwind transport, from source (UV1) to the FLAME scanners' measurement distance (~2 km). The transport behaviour of a volcanic gas plume can be approximated by two end-member processes (Figure 5.6). A Gaussian continuous model (Turner, 1970; Figure 5.6-A) describes the transport and dilution behaviour of a plumes continuously emitted (at constant flux) from source, and in which gas concentrations are linearly diluted upon increasing plume transport distances. In such conditions, where laminar and steady flow prevail (and no turbulence), the plume is passively transported by the wind (advection), and in-plume concentrations gradually decline with distance from source (dilution) proportionally to plume widening. Inherent in the concept of a continuous-type plume is that the SO₂ flux is conservative (invariant with distance) irrespective of prevailing wind speed ($F_0=F_1$; Figure 5.6-A). We have analytically simulated the atmospheric behaviour of a Gaussian continuous plume (see Supplementary Figure S5.4) to predict that, in this plume transport model, the flux remains constant with distance (Supplementary Figure S5.4-A,B) independently on plume transport speed (Supplementary Figure S5.4-D), while the plume ICA gradually decreases with increasing plume speed (Supplementary Figure S5.4-C). This occurs because, in accordance with the principle of mass conservation, to maintain a constant gas flux in each section of the dispersing plume, any increase in velocity must be counterbalanced by a proportional dilution of gas concentrations.

The alternative end-member plume transport model is that of a puffing plume (Figure 5.6-B). In this case the volcanic plume is not emitted continuously and with a variable in time emission rate. This non-continuous behaviour is especially suitable to simulate the dispersion of pollutants emitted as discrete puffs or parcels, when the release time and sampling times are very short compared to the travel time from the source to the receptor. In the puffing plume model, each puff is treated as a concentrated mass of pollutants that moves and spreads with the wind, at a speed influenced (among other factors) by atmospheric stability and turbulence. As individual puffs are rapidly transported by wind, atmospheric dilution is minimised (Turner, 1970), so that plume ICA do not scale inversely to plume speed, and the source gas flux is not conserved upon transport ($F_0 \neq F_1$; Figure 5.6-B). The

extent to which this model is appropriate for volcanic plume remains untested. Volcanic plumes at open-vent volcanoes are indeed typically emitted in puffs (rhythmic pulsations) of over-pressured gas outbursting at the magma-atmosphere interface (Barrington et al., 2022; Harris and Ripepe, 2007). However, volcanic gas puffs are concentrated regions of gas at the boundary of which sharp concentration gradients exist. Subject to turbulence, air entrainment will be favored at each puff boundary, causing some extent of mixing with air and diffusion. Hence, a pure puffing model is an unrealistic, end-member representation of volcanic plume behavior. If however a volcanic plume is at least partially of “puffing” mode, then the gas fluxes measured in distal locations may not be representative of the source flux.

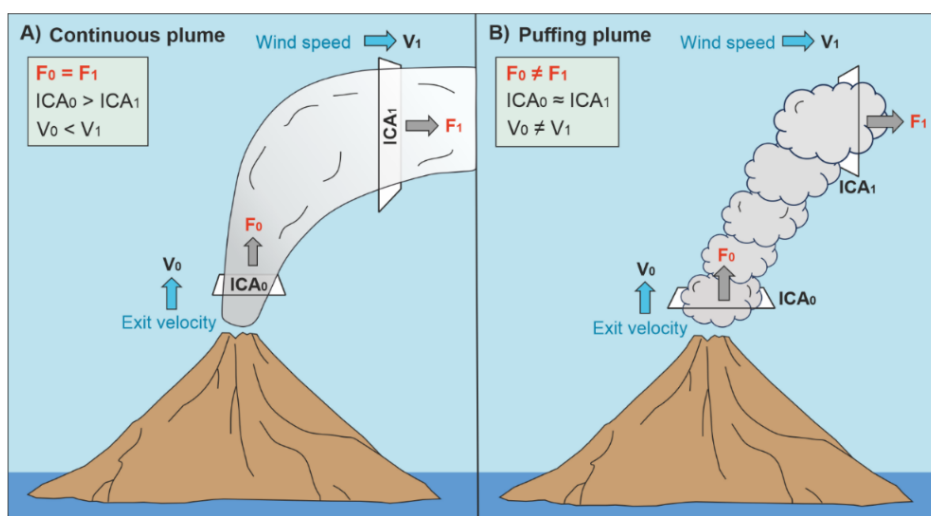


Figure 5.6 – A) Sketch of a continuous plume model, not in scale. Column amounts are gradually diluted over distance ($ICA_0 > ICA_1$) under the effect of increasing size of the plume due to dispersion and proportionally with wind speed ($V_0 < V_1$), in order to maintain a constant flux. B) Sketch of an extremized puffing/impulsive plume model, not in scale. Column amounts are not diluted over distance ($ICA_0 \approx ICA_1$) and wind velocities are different than those near-vent ($V_0 \neq V_1$). The resulting flux at distance changes ($F_0 \neq F_1$) due to differences in estimated velocities. Note that this is only an extremization and do not represent the real behaviour of a puffing plume. The dilution of individual puffs is intentionally omitted to present an extreme model for comparison as an alternative endmember.

Our combined UV Camera-scanning-DOAS observations offer an opportunity to characterise the Stromboli’s plume dispersion behaviour during atmospheric transport from source. We examine the continuous vs. puffing behaviour of Stromboli’s plume in the bi-logarithmic plot of Figure 5.7. Solution of our Gaussian continuous model is obtained by solving Equation S5.1 (see Supplementary materials) at a fixed distance of 2 km (average distance of the FLAME spectrometers to the summit craters). As previously stated, the Gaussian continuous model predicts (blue line in Figure 5.7) gas column amounts to decrease linearly with increasing wind speed (in a Log-Log space). A pure puffing plume behaviour, in which emitted puffs are advected by wind with no dilution occurring, is instead expressed by the black line in Figure 5.7. Our Stromboli’s experimental data, illustrated in Figure 5.7

in the form of FLAME/UV1 ICA ratio vs. wind speed (from FLAME), plot along a linear alignment (orange line in Figure 5.7) intermediate between the continuous (Gaussian model) and the puffing (with absence of dilution) model lines. This experimentally derived atmospheric behaviour of Stromboli's plume implies that in-plume SO_2 column amounts are indeed diluted upon atmospheric transport and advection, but not at the rate that would be required by the Gaussian continuous plume hypothesis. If the Stromboli's plume is hence intermediate in nature between the two end-member processes of Figure 5.7, then combining distally observed ICAs with wind speed into an SO_2 flux (Eq. 5.1) causes an overestimations of the real (source) flux. We consider this the dominant control factor on the large FLAME vs. UV1 flux mismatch.

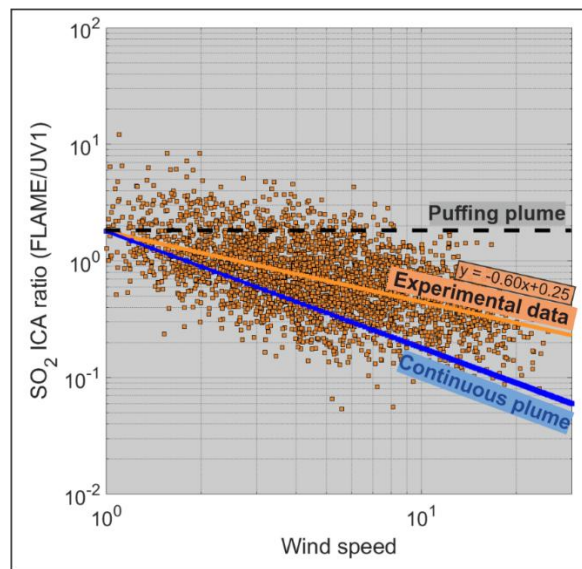


Figure 5.7 – Distribution of our experimental data in a bi-logarithmic plot, in the form of FLAME/UV1 ICA ratio vs. wind speed. Note that the measured UV1 ICAs are corrected (multiplied) by factor 1.60 to compensate against the 60% underestimation caused by topographic/network geometry issues and radiative transfer effects (see text). These data plot along a linear alignment (orange line) intermediate between the continuous (Gaussian) and the extremized puffing (with absence of dilution) model lines. The blue line represents the output of our Gaussian continuous plume modelling (considering a constant emission rate equal to the intercept value of the experimental line data), while the black one represents an extremized puffing plume behaviour (idealized and not modelled) with absence of dilution. Our experimental line suggests that SO_2 column amounts within the plume could be non-ideally diluted upon atmospheric transport and deviates from the ideal Gaussian continuous plume model.

5.4.2 Toward a combined SO_2 flux record

To summarise, our results show that both observation methods used at Stromboli come in with advantages and disadvantages. The near-vent UV Camera measurements likely underestimate the real SO_2 ICAs (and hence fluxes) due to the optically opaque nature of the plume, and because of the geometry of the observation network that offers an incomplete view of the plume. These may concur to cause a total ~60% negative offset (under-estimation) of the SO_2 flux. In contrast, use of wind

speed data to convert ICAs into SO₂ fluxes (as done by FLAME) may represent the main source of error in distal measurements with scanning spectrometers, and introduces a marked seasonal periodicity (Figure 5.4-A) that is obviously not volcano-related.

If we are to obtain a more accurate record of volcanic SO₂ fluxes, then we must take advantage of pros of each of the two techniques, while mitigating the impact of their intrinsic errors and caveats. We therefore suggest a novel, integrated SO₂ flux record obtained by combining (i) the near-vent plume velocities obtained by processing (using an optical flow algorithm; see Supplementary Figure S5.2) of UV1 images and (ii) the SO₂ ICAs derived from the FLAME network. Notably, for the obtained flux to be meaningful, the FLAME ICAs must be first back-calculated at source (where plume speed is measured by UV Camera), trying to remove the effects of winds. This is obtained by quantifying the extent of plume dilution upon plume transport from UV1 to FLAME, using the coefficients of the experimentally derived (for Stromboli) dilution function (Figure 5.7):

$$Dilution = \frac{ICA_{FLAME}}{ICA_{UV1}} = 10^b * V_{wind}^a \quad (Eq. 5.2)$$

where a (-0.60) and b (0.25) are respectively the slope and the intercept of the Log-Log experimental linear function (orange line in Figure 5.7).

Equation 5.2 can then be rearranged to calculate ICA at source (ICA_0) as:

$$ICA_0 \approx ICA_{UV1} * C = \frac{ICA_{FLAME}}{10^{b*V_{wind}^a}} \quad (Eq. 5.3)$$

where $C = 1.60$ is an amplification factor that takes into account the ICA_{UV1} underestimation (caused by topographic/network geometry and radiative transfer constraints).

The so obtained source ICAs (ICA_0) average at 0.70 ± 0.34 kg/m (range: 0.11-2.66 kg/m). The integrated flux (in t/d) is then:

$$Combined\ SO_2\ flux = ICA_0 \cdot V_{UV1} \quad (Eq. 5.4)$$

where V_{UV1} is the optical flow derived velocity used from UV1.

The combined SO₂ flux is illustrated as a red solid line in Figure 5.8-A, where it plots in an intermediate position between the UV1 and FLAME fluxes (mean of 108 ± 78 t/d; range: 12-785 t/d). This combined flux allows exploiting the robustness of DOAS concentration measurements, while reducing the noise generated by wind interference: note the increasing correlation with tremor, from $R=0.39$ (for FLAME flux) to $R=0.54$ (Figure 5.8-B).

We quantify the uncertainty on the derived combined flux by combining uncertainties arising from (i) plume velocities inferred from the optical flow algorithm, (ii) SO₂ integrated column amounts (ICAs) streamed by FLAME and (iii) in the experimentally derived dilution function (Figure 5.7). As previously discussed, the application of the optical flow algorithm on the UV1-acquired images leads to errors <5% (we consider 5% for this purpose). The total error associated with measurements of

SO₂ flux from the FLAME network amounts to maximum -22 +36% (Salerno et al., 2009a). Excluding from this the error associated with wind speed and plume height, the mean residual error of the FLAME network is $\leq 15\%$. To determine the error contribution deriving from the use of our experimental dilution line, we use a root mean square error (RMSE) to estimate the average difference between the predicted values (regression line) and the actual values, providing an indication of how well the model's predictions match the observed data. The RMSE value for data in Figure 5.7 is 0.71, leading to a total combined uncertainty for the combined SO₂ flux of $\sim \pm 72\%$.

We stress that this “combined SO₂ flux” methodology can potentially be applied at any volcano where a network of scanning spectrometers is operational, and where near-vent gas exit velocity profile can be retrieved by any visible or UV Camera operating in the near field.

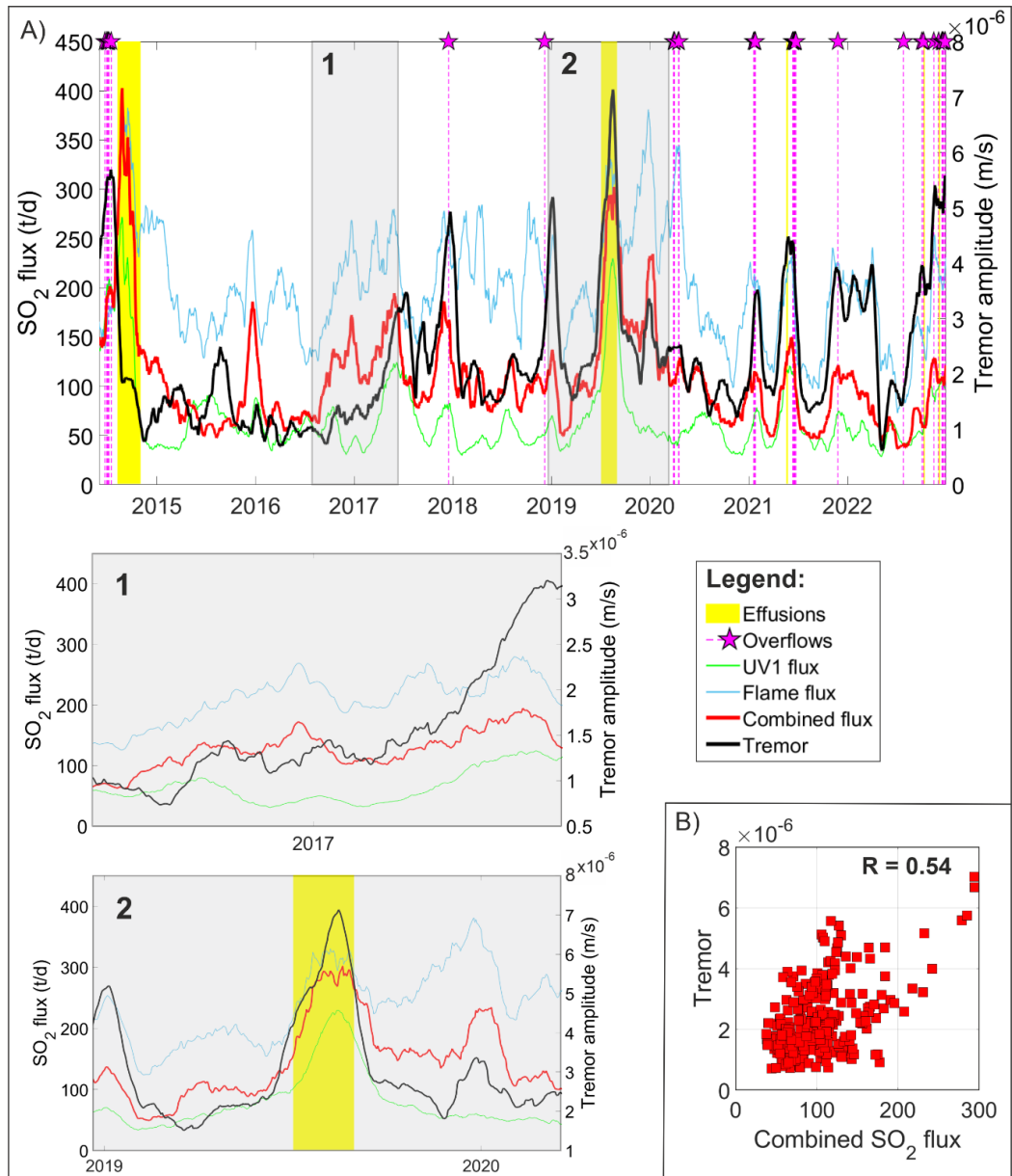


Figure 5.8 – A) SO_2 fluxes from FLAME network (azure line) and UV1 system (green line) compared with the new combined flux (red line) and volcanic tremor (black line). Effusions and overflows are also shown (see Legend). The grey panels (1 and 2) show a focus on two sub-intervals, to better highlight the synchronous fluctuations of the combined flux with the volcanic tremor. B) Scatter-plot between the new combined flux and volcanic tremor. Removing the wind component from scanning spectrometers SO_2 flux measurements improve the general correlation with volcanic tremor.

5.5. Conclusions

Our Stromboli's results here document some of the challenges encountered while attempting to derive robust SO_2 flux records from data streamed by automated instrumental networks of scanning spectrometers and UV Cameras. Our 9 years of observations demonstrate a large mismatch between SO_2 flux time series derived from scanning spectrometers and UV Cameras, with the former being a factor $\sim 200\%$ higher on average. We attribute this mismatch to a combination of causes, including (i) observational limits caused by morphology/topography of the volcano's summit (ii) radiative

transfer issues, and (iii) complex plume transport behaviour. The first two processes mostly cause a negative offset (under-estimation) in the UV Camera SO₂ flux time series; while the third process, caused by incomplete (non-ideal) atmospheric dilution of source-released gas puffs upon atmospheric transport, imparts a positive wind speed dependence and a marked seasonality to the distal scanning spectrometers' results. Our analysis suggests this latter effect to be the dominant one, causing the distal fluxes to significantly exceed those estimated by the UV Camera in the near-vent field. The positive correlation found at several volcanoes between SO₂ flux and wind speed (e.g., Barrington et al., 2022; Dinger et al., 2021) suggests that the non-ideal plume transport/dilution behaviour we find evidence for at Stromboli may be more common at volcanoes than so far realised.

Based on this consideration, we propose a combined approach, integrating (a) gas velocities derived from optical flow algorithms implemented into UV Cameras system with (b) SO₂ column amounts obtained by DOAS processing of spectra delivered by scanning spectrometers (one properly back-calculated at near-vent conditions). We expect this combined SO₂ flux to be less affected by external factors (e.g., meteorological, illumination and volcano topography conditions), and hence a better proxy of volcano behaviour, with obvious benefits for volcano monitoring and volcanic hazard assessment.

This work represents an attempt to eliminate the effect of wind from DOAS measurements, which, despite their strong spectroscopic accuracy, rely on wind data to derive fluxes, making it the primary source of error. Our work suggests that integrating observations from near-vent UV Cameras and distal scanning spectrometers into a combined SO₂ flux may lead to more accurate measurements, mitigating against the effects of caveats/limitations associated with each individual technique. The large pre- (2014 eruption) and sin-eruptive (2014 and 2019 effusive eruptions) SO₂ flux increases seen at Stromboli (Figure 5.8-A), being well above the typical baseline SO₂ levels observed during regular (persistent) strombolian activity, are good hints in this direction.

SUPPLEMENTARY MATERIAL

Testing the velocity field measured by optical flow

Optical flow algorithms have demonstrated good performance in determining the velocity profile of volcanic gas plumes (e.g. Delle Donne et al., 2019). Moreover, application of these algorithms often represents the sole opportunity to directly measure the velocity of volcanic plumes without relying on indirect proxies, such as wind speed.

The UV1 system calculates plume SO₂ column densities and velocities within rectangular regions across (and right above) the crater terrace (Supplementary Figure S5.1), hence maximizing

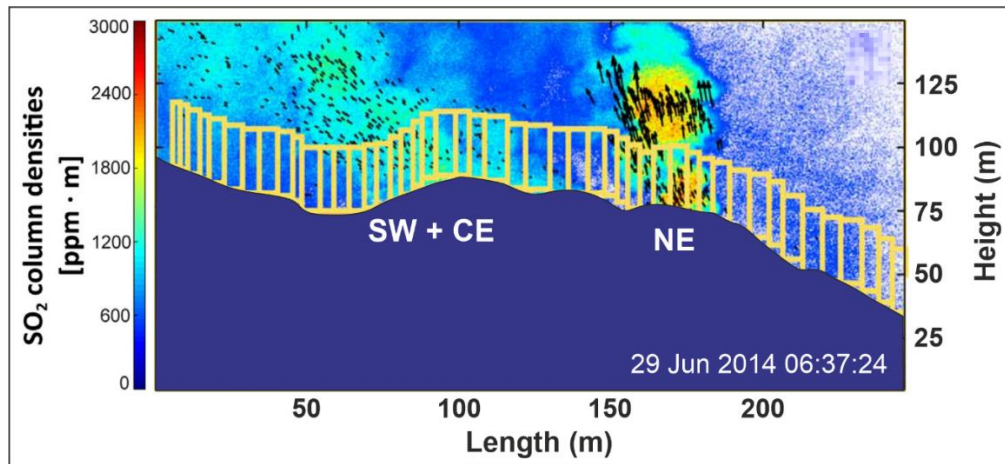
the possibility of gas detection and minimizing atmospheric effects such as air entrainment in the plume and complexities in the local wind field. This approach, however, returns averaged velocities lower than the effective maximum plume's speed if considered on its own. This is because the rectangular cross-section also includes non-degassing portions of the crater terrace, whose ~ 0 velocities contribute to the section-averaged speed. To verify the algorithm's efficacy in accurately determining the real plume's speed, it is essential to isolate the degassing portions within the crater terrace, and calculate the gas exit speed inside these specific regions only (Supplementary Figure S5.2).

We test this by recalculating (in 1 year data test subset) the SO_2 fluxes while setting a Column density threshold, and hence considering only velocities for regions of the rectangular cross-section above that threshold. This allows isolating only degassing portions as close as possible above the vent and to evaluate the gas velocity only where the SO_2 signal is maximized. Since velocities from UV1 are calculated as averages over the whole cross-section (sum of all rectangular regions in Supplementary Figure S5.1), we obtain progressively higher averaged speeds while increasing the Column density threshold used. For example, our estimated plume velocities increase from 2.51 ± 1.16 m/s (range: 0.97-11.34 m/s) at 1000 ppm·m threshold, to 3.26 ± 1.65 m/s (range: 0.93-10.95 m/s) at 1500 ppm·m threshold, and finally 4.27 ± 2.74 m/s (range: 0.65-29.40 m/s) at 2000 ppm·m threshold (Supplementary Figure S5.2). However, the fluxes are independent on the threshold used (Supplementary Figure S5.2). This is because, if for example the threshold is decreased, then the consequent average speed decrease is counterbalanced by an increased actively degassing plume cross-sectional area.

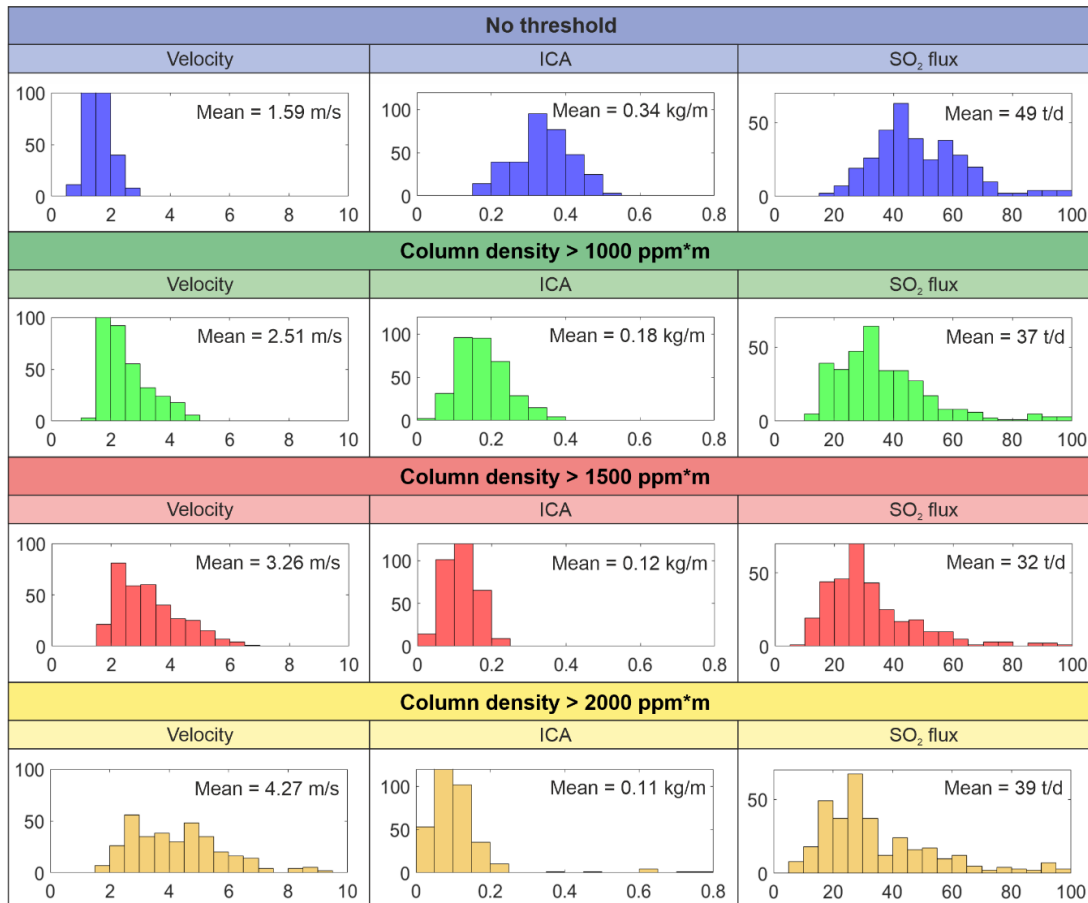
Tests show that use of the 2000 ppm·m threshold would only allow very concentrated gas pulses (typically observed during strombolian explosions) to be taken into account in the calculations, hence neglecting the (dominant; Tamburello et al., 2012) contribution of persistent, passive degassing (typically associated with less concentrated plumes with lower, gas buoyancy velocities). In contrast, use of the 1000 ppm·m threshold may leads to more noisy results, as impacted by random dispersion of gas patches.

We emphasise that the velocities we obtained within these “active” rectangular cells of the plume cross-section used are representative of buoyant plume rise, and hence depend on plume density ρ (Morton et al., 1956; Yamamoto et al., 2008). Considering an ambient air density of 1.08 kg/m^3 (at 750m and 25°C), a plume speed of 3.26 m/s (mean value associated to the 1500 ppm·m threshold) and considering a height of the plume front above the vent of 5 m (median of rectangular subregions), we obtain a gas plume density of $\sim 0.37 \text{ kg/m}^3$, as previously found for typical Strombolian plumes (e.g., Delle Donne et al., 2022: $\sim 0.35 \text{ kg/m}^3$). Our gas density value is also

consistent with that calculated using the ideal gases law ($\rho = PM/RT = \sim 0.43 \text{ kg/m}^3$), using the average chemical composition of the Stromboli's magmatic gases (Aiuppa et al., 2010b), a gas exit temperature of 625 K obtained from thermal camera measurements (Harris and Ripepe, 2007) and an atmospheric pressure (at 750 m elevation) of 92.91 kPa.



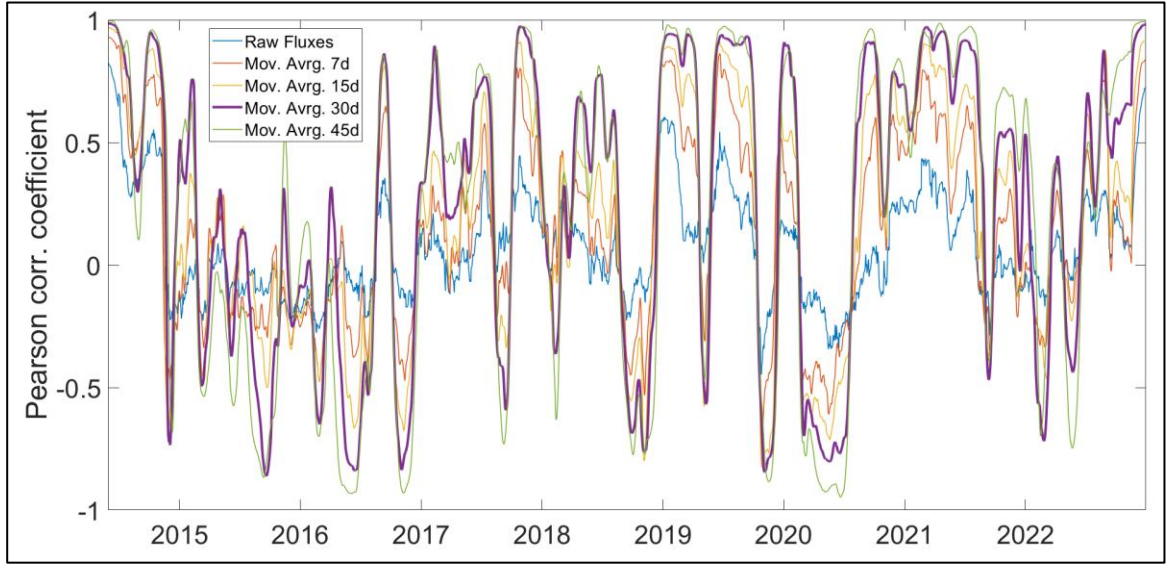
Supplementary Figure S5.1 - Rectangular subregions (yellow rectangles) on crater terrace where column densities and plume velocity profile are retrieved automatically from the UV1 system, modified from Delle Donne et al., 2022.



Supplementary Figure S5.2– Histograms of daily mean fluxes and their components from UV1, calculated into restricted degassing portions of the summit crater area, applying increasing threshold on Column Density's values and extracting corresponding SO₂ ICAs and velocity values. For increasing threshold the daily mean velocities improve and the ICAs decrease, while the resulting fluxes decrease slightly and proportionally to the reduction of the degassing area.

Selection of the most suitable correlation window to display long-term correlations between measurements

The correlation between UV1 and FLAME SO₂ flux time series (Figure 5.2-C) has been examined in a range of timescales. Since we are interested in reducing the noise and short-term variability between the two degassing time series, we have chosen a 30-day moving average window to emphasise the long-term coherency of the two trends (Figure 5.2-C). This window leads to a better correlation between the two fluxes, as observed in Supplementary Figure S5.3, where the Pearson Correlation Coefficient (R) is represented over a 90-day centred window for increasing moving average intervals. The 30-day moving average window yields a better gain in terms of correlation rather than anticorrelation. Over this threshold the Pearson coefficient tends to acquire an opposing trend, mainly shifting towards the anticorrelation.



Supplementary Figure S5.3 – Rolling correlation in a 90-day centred window between the two SO₂ flux daily measurements (UV1 and FLAME) for increasing moving average windows. The 30-day moving average window (purple line) results in a stronger positive correlation compared to others. Beyond this interval, the Pearson coefficient tends to exhibit a contrasting pattern, primarily leaning towards negative correlation.

Gaussian continuous plume dispersion model

The Gaussian plume model is a mathematical model commonly used to predict the dispersion of air pollutants released into the atmosphere (e.g., Woodward et al., 2021; Zannetti, 1990) from continuous sources. It is particularly useful for assessing the atmospheric concentrations of gas molecules downwind their source, such as an industrial stack or a point source of emissions such as a volcanic vent (e.g., Copelli et al., 2019; Lotrecchiano et al., 2020; Middleton et al., 1979).

We use the Gaussian plume function implemented in Turner (1970) to simulate the atmospheric concentrations that would result from the downwind dispersion of gas emitted from a continuous and punctual volcanic SO₂ source. To start, an orthogonal Cartesian reference system is set, with its origin centred at source and the x axis parallel to the wind direction. The y axis is horizontal and perpendicular to the x axis, while the z axis in the vertical direction corresponds to the elevation from the ground. The gas concentration $C(x,y,z)$ is retrieved using the distribution of mass in both vertical and cross-wind dimensions at a given downwind distance, following the equation:

$$C(x, y, z) = \frac{Q}{2\pi\mu\sigma_y\sigma_z} \left\{ \exp\left(\frac{-(z-h)^2}{2\sigma_z^2}\right) + \exp\left(\frac{-(z+h)^2}{2\sigma_z^2}\right) \right\} \left\{ \exp\left(\frac{-(y)^2}{2\sigma_y^2}\right) \right\} \quad (\text{Eq. S5.1})$$

where C is the pollutant concentration as a function of downwind position (x,y,z) , Q is the mass emission rate, μ is the mean wind speed evaluated at the effective release height, σ_y and σ_z represent the standard deviations of plume concentration distribution in the horizontal and vertical planes, and h is the effective stack height (Turner, 1970; Woodward et al., 2021). The assumptions made in deriving this formula include steady state conditions, no spatial or temporal variations in the wind or

atmospheric stability, no upper-level inversion, no deposition of the contaminant or chemical reactions between it and other substances, and continuous point sources emitting at constant rates (Q).

For any downwind section of the plume x , the ICA (Integrated Column Amount) can be defined as:

$$ICA(x) = \int_y \int_z C(x, y, z) dy dz \quad (\text{Eq. S5.2})$$

If we combine Equations S5.1 and S5.2, we obtain the formula relating the flux as we obtain by multiplication of ICA for the wind velocity μ to the emission flux at the source Q:

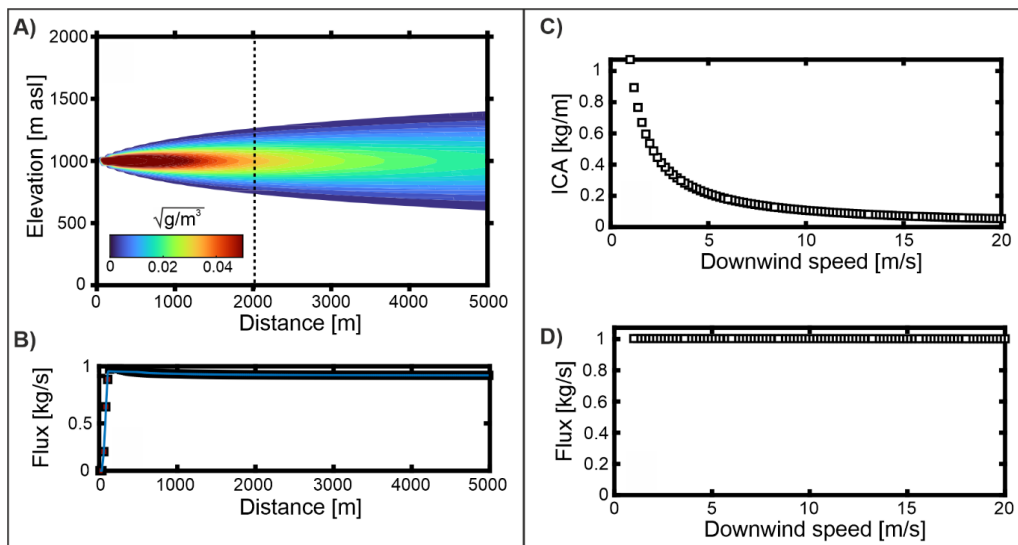
$$\int_y \int_z C(x, y, z) dy dz * \mu = \frac{Q}{2\pi\sigma_y\sigma_z} \int_{-\infty}^{\infty} e^{\frac{-z^2}{2\sigma_z^2}} dz \int_{-\infty}^{\infty} e^{\frac{-y^2}{2\sigma_y^2}} dy \quad (\text{Eq. S5.3})$$

Since the term $\int_{-\infty}^{\infty} e^{\frac{-z^2}{2\sigma_z^2}} dz \int_{-\infty}^{\infty} e^{\frac{-y^2}{2\sigma_y^2}} dy = 2\pi\sigma_z\sigma_y$ then the multiplication between ICA taken at any given distance x from the source and the wind velocity is equal to the source flux Q . This validates the approach to use the wind velocity with the measured ICA at any distance to derive fluxes at the source, under the assumption of a constant mass emission rate. To verify the model derived from analytical formulation, we modelled the gas dispersion from a theoretical stack emitting gas at 1 kg/s under fixed (Supplementary Figure S5.4-A,B) and variable wind conditions (Supplementary Figure S5.4-C,D).

Supplementary Figure S5.4-A illustrates the predicted downwind gas concentrations in the plume, obtained by assuming a fixed emission rate (1 kg/s) and wind speed (of 10 m/s). The model predicts a significant decrease in gas concentrations at increasing distance from source, as the plume expands and dilutes. However, the calculated flux for each vertical plume section remains constant, and coincident with that at source (Supplementary Figure S5.4-B). Supplementary Figure S5.4-C illustrates the predicted ICA for a fixed downwind distance of observation (2000 m, see dashed line in Supplementary Figure S5.4-A) and using the same input flux of the source, but under variable wind conditions (1 to 20 m/s). In this modelling, any increase in the plume transport speed produces a dilution of the plume, but the resulting flux, obtained multiplying the ICA by the downwind speed, remains constant and equal to same input flux of 1 kg/s (Supplementary Figure S5.4-D). Therefore, the flux calculated with this Gaussian model does not vary neither with increasing distance from the emission centre (Supplementary Figure S5.4-B), nor with wind intensity (Supplementary Figure S5.4-D).

These model simulations imply that, in real field (volcano) conditions, proximal (UV Camera) and distal (scanning spectrometer) fluxes should match one each other if gas is emitted continuously and a Gaussian behaviour prevails. In such end-member condition, characterised by laminar and steady flow (and no turbulence), the plume would be expected to be passively transported by the

wind, and in-plume concentrations to gradually decline with distance from source, proportionally to plume widening (so as to preserve the gas flux at source). It is important to remind that ICAs, either measured in a distal section from scanning spectrometers or under near-vent conditions by UV Cameras, are multiplied by the gas transport speed to obtain a flux measurement. However, while ICAs contain information about the magmatic degassing process (and dilution patterns), transport velocities are not inherently linked to volcanic processes, but are rather controlled by atmospheric phenomena.



Supplementary Figure S5.4 – A) Flux Density along down-wind direction for a constant emission of 1 kg/s and assuming a Gaussian plume dispersion model for a wind transport speed of 10 m/s. A progressive dilution and expansion occurs as the plume spreads in the downwind direction, but however the resulting ICAs (Integrated Column Amounts) remain constant for every distances, so that if they are multiplied by the wind transport speed (assumed to be constant at every point of the grid), we obtain a flux estimation (in B) equal to the input flux of 1 kg/s. In C and D the downwind transport speed dependence on ICAs and flux are shown. For the same downwind distance of 2000 m of observation (see dashed line in A) and using the same input flux of the source, any increase in the plume transport speed will dilute the plume (decrease of ICA in C), but however the calculated flux (D), obtained multiplying the ICA for the downwind speed, remains constant and equal to same input flux of 1 kg/s that we used for the plume modelling work.

CHAPTER 6

VOLCANIC GAS CHANGES PRIOR TO STROMBOLI'S MAJOR EXPLOSIONS ARE STATISTICALLY SIGNIFICANT

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Abstract

The generally mild activity of mafic, open-vent volcanoes is punctuated by the periodic occurrence of sudden, larger-than-normal explosive eruptions. Examples of these sudden mafic explosive events are the so-called “major explosions” that occur (2 to 4 times a year on average) at Stromboli volcano in Italy. These relatively small explosions (Volcanic Explosivity Index, $VEI < 1$) occur without no obvious precursory change in surface activity, and therefore pose a threat to volcanologists/monitoring staff, and to the population in the most severe cases. Past work has found a link between periods of high CO_2/SO_2 ratios in the volcanic gas plume and the occurrence of such explosions, but this association has never been statistically verified. Here, we report on nearly continuous observations of volcanic gas plume CO_2 and SO_2 concentrations and ratios in a diluted plume, as well as SO_2 flux measurements, recorded in a four years (2020 to 2023) activity period of Stromboli volcano, during which 22 such major explosions took place. Using the Conditional Logit model and the Receiver Operating Characteristic (ROC) analysis, we establish a statistical association between the occurrence of major explosions and periods of reduced SO_2 concentrations (very significant) and fluxes, and high CO_2/SO_2 ratios (significant), in the volcanic plume. These findings are interpreted in light of a simplified conceptual model that explains major explosions as caused by gas bubble accumulation at a rheological discontinuity, resulting from deceleration of the shallow convecting magma that supplies the “regular” Strombolian activity. Using results of statistical analysis, we develop a volcanic gas-based Composite indicator that successfully forecasts (by a-posteriori analysis) 71% of the events on timescales of week(s). However, we find that this Composite indicator is associated with a large (32%) False Positive rate and hence low precision (20%). The

significant role of SO₂ concentrations in the model indicates that other factors, such as plume direction and inter-crater variations in gas composition, may need to be taken into account for improving the forecasting performance of the method. Thus, while our results emphasize the importance of gas plume observations in volcano monitoring, they also highlight their current limitations as eruption forecasting tools.

6.1 Introduction

Active, potentially harmful volcanoes worldwide are monitored by increasingly sophisticated monitoring networks (Acocella et al., 2023; Sparks et al., 2012), with the goal of detecting changes in patterns and trends of monitored parameters in the run-up to eruption (Poland and Anderson, 2020; Sparks and Cashman, 2017). In addition to seismicity (Matoza and Roman, 2022), ground deformation (Biggs and Pritchard, 2017) and thermal emissions (Reath et al., 2019a), volcanic gas geochemistry has long been recognised (e.g., Menyailov, 1975) as a potentially very useful volcano monitoring tool (Edmonds, 2021; Kern et al., 2022). Magmatic volatiles have finite solubilities in magmas, causing them to partition into a separate volatile phase at upper mantle to crustal conditions (Papale et al., 2022). This magmatic degassing process (Oppenheimer et al., 2014) is a critical driver of magma evolution during storage and pre-eruptive ascent, and plays a key control on if, and with which modalities and rates, magma will ultimately erupt (Edmonds and Wallace, 2017). From a volcano monitoring perspective, the intrinsically mobile nature of gas bubbles implies that they can separate and escape from deeply stored magma (Edmonds et al., 2022) to be eventually emitted at the surface well before magma.

There is vast, documented evidence in the geological literature on the use of volcanic gas chemistry as eruption precursor (Edmonds et al., 2021). Reported observations are the most diverse in terms of methodology used, targeted emission type (fumaroles, plumes, soil diffuse emissions, groundwaters), and measurement temporal resolution; these range from multi-species, long-term monitoring of fumarole compositions, through periodic (monthly to yearly) direct sampling surveys (e.g., Vaselli et al., 2010) to single species (e.g., SO₂ flux) near-real time (temporal resolution of seconds to minutes) observations with instrumental networks (e.g., Delle Donne et al., 2017; Arellano et al., 2021). An especially useful advance has come with the advent of compact, multi-sensor gas sensing units (e.g., Multi-GAS) that can be used for near real-time temporal monitoring of the CO₂/SO₂ ratio in volcanic gas plumes (e.g., Aiuppa et al., 2007, 2010a, 2021; De Moor et al., 2016a, 2016b, 2019).

Notwithstanding their significant potential, the use of volcanic gas observations as truly exploitable eruption forecasting tools is challenged by a lack of quantitative data analysis testing the

statistical robustness of any identified gas “precursor” (although notable exceptions exist; Kunrat et al., 2022). This partly reflects the relatively novel and immature technology in volcanic gas science, which requires frequent field maintenance from expert operators and has hitherto hampered wider (global) deployment of gas sensing networks. As such, complete (temporally continuous) long-term volcanic gas datasets are limited to only a few, well-instrumented volcanoes. This, in conjunction with the relatively infrequent volcanic events occurring at any given volcano, has led to monitoring gas data being insufficient (in number, continuity, and quality) to systematically test patterns indicative of a certain volcanic behaviour. In many cases, the reported “precursory” gas variations refer to only one specific eruptive event (e.g., Aiuppa et al., 2017, 2018), leaving the question on their systematicity and repeatability unanswered.

Stromboli volcano in Sicily (Southern Italy) hosts a well-instrumented volcanic gas monitoring network (Allard et al., 2008; Aiuppa et al., 2009, 2010a, 2021; Burton et al., 2009; Inguaggiato et al., 2013, 2017; Delle Donne et al., 2017, 2022; Federico et al., 2023; Lo Bue Trisciuzzi et al., 2024) and, owing to its relatively steady and well-understood eruptive behaviour (Rosi et al., 2013; Pioli et al., 2014), is an ideal test site to quantitatively explore the link between volcanic gas variations and volcano behaviour. Here, we focus on an intermediate class of explosive events referred as “Major Explosions” that, in view of their eruption dynamics (Bertagnini et al., 1999; Andronico and Pistolesi, 2010) and relatively high occurrence rate (Bevilacqua et al., 2020), represent one of the most hazardous volcanic phenomena at this volcano (Rosi et al., 2013). It has been previously shown (Aiuppa et al., 2011) that major explosions are often anticipated – by days to weeks – by remarkable increases in the volcanic gas plume CO₂/SO₂ ratio; these have been explained as being due to precursory leakage of CO₂-rich gas bubbles from an accumulating foam on top of a deeply stored (> 3 km depth) mafic magma (whose later, rapid ascent then triggers the explosion itself) (Aiuppa et al., 2010a, 2011, 2021).

Here, we analyse the statistical association between the temporal variations in plume chemistry/flux at Stromboli and the occurrence of major explosions from January 2020 to December 2023. Our aim is to quantitatively test the potential utility of volcanic gas plume geochemistry as an exploitable eruption forecasting tool.

6.2 Stromboli’s major explosions

Stromboli’s major explosions are transient, impulsive (total duration of tens of seconds) explosions that produce discrete (unsustained) eruptive columns ~200 meters in height on average (Figure 6.1). These violent blasts typically erupt moderate volumes (10^2 to $\leq 10^4$ m³) of mostly coarse, vesicular pyroclastic fragments and ballistics that cover the crater terrace and the volcano’s slope

without leaving a continuous blanket of tephra deposits (Bertagnini et al., 1999; Andronico and Pistolesi, 2010; Rosi et al., 2013; Pioli et al., 2014). Major explosions thus share similar eruption dynamics with, but have larger dispersal areas and higher erupting volumes than, the regular Strombolian explosions ($\sim 10 \text{ m}^3$) that exemplify Stromboli's ordinary activity (Ripepe et al., 2008). Major explosions occur at an average rate of $\sim 1\text{-}4$ events per year (~ 140 events catalogued in the 1879-2020 period; Bevilacqua et al., 2020), with no visible precursory change in surface activity. In the very short-term ($< 300 \text{ s}$ before), each major explosion is typically anticipated by mild ground uplift ($< 1 \text{ } \mu\text{rad}$), interpreted as caused by pressure build-up in the upper conduit system due to rapid expansion of gas and/or volatile-rich magma into the shallow stored conduit magma (Ripepe et al., 2021). Whereas magma fragmentation is unquestionably a shallow process, there is debate on whether overpressure develops in the shallow conduit itself (i.e., in response to formation of a viscous plug, and hence permeability drop; Falsaperla and Spampinato, 2003; Calvari et al., 2012), or if instead it is triggered by the arrival of more deeply sourced gas/magma. The latter hypothesis is corroborated by the frequent eruption of highly vesicular (pumiceous) Low Porphiricity (LP) magma fragments during most major explosions (Andronico and Pistolesi, 2010; La Felice and Landi 2011; Pioli et al., 2014; Voloschina et al., 2023; Insinga et al., 2024). This LP magma component is instead always ejected during paroxysmal eruptions (the most intense manifestations of Stromboli volcano) and, according to petrological (Métrich et al., 2001, 2010, 2021; Voloschina et al., 2023) and experimental (Pichavant et al., 2009, 2022) evidence, cannot derive from previously shallow stored magma, but rather requires rapid ascent from a $> 3 \text{ km}$ deep magma storage zone. The emission of CO_2 -rich gas prior to major explosions (Aiuppa et al., 2011, 2021; Voloschina et al., 2023) is also an indication of a deeply operating process in the run-up to the explosion. These precursory gas changes are the object of this study.

In the four years temporal window covered by this study (2020 to 2023), there were a total of 22 major explosions, with an occurrence rate (estimated on a 3-month basis) of 0 to 4 events (Figure 6.2a). As previously found (Bevilacqua et al., 2020), these events occur in clusters, in which several events repeat in relatively narrow intervals (of days to a few months). In our study interval, we identify three main clusters (labelled as *I*, *II* and *III* in Figure 6.2a) in July-December 2020, July-October 2021, and May 2022-April 2023, interspersed within periods with no (or less frequent) events. We include two impulsive explosive events (orange stars; on October 9 and December 4 2022) in our statistics, during which no vertically rising eruptive clouds were observed (as normally occurs during major explosions), but rather the rapid formation of turbulent, hot pyroclastic density currents (PDC) from the active vents (Figure 6.1, bottom panels). These PDCs, rapidly emplaced along the Sciara del Fuoco scar, also led to a sequence of small tsunamis entering the sea, as indicated by

monitoring reports of the Laboratorio Geofisica Sperimentale (LGS, Università di Firenze) (see also Global Volcanism Program, 2023). Our analysis is hence based on 24 non-regular explosive events in total.

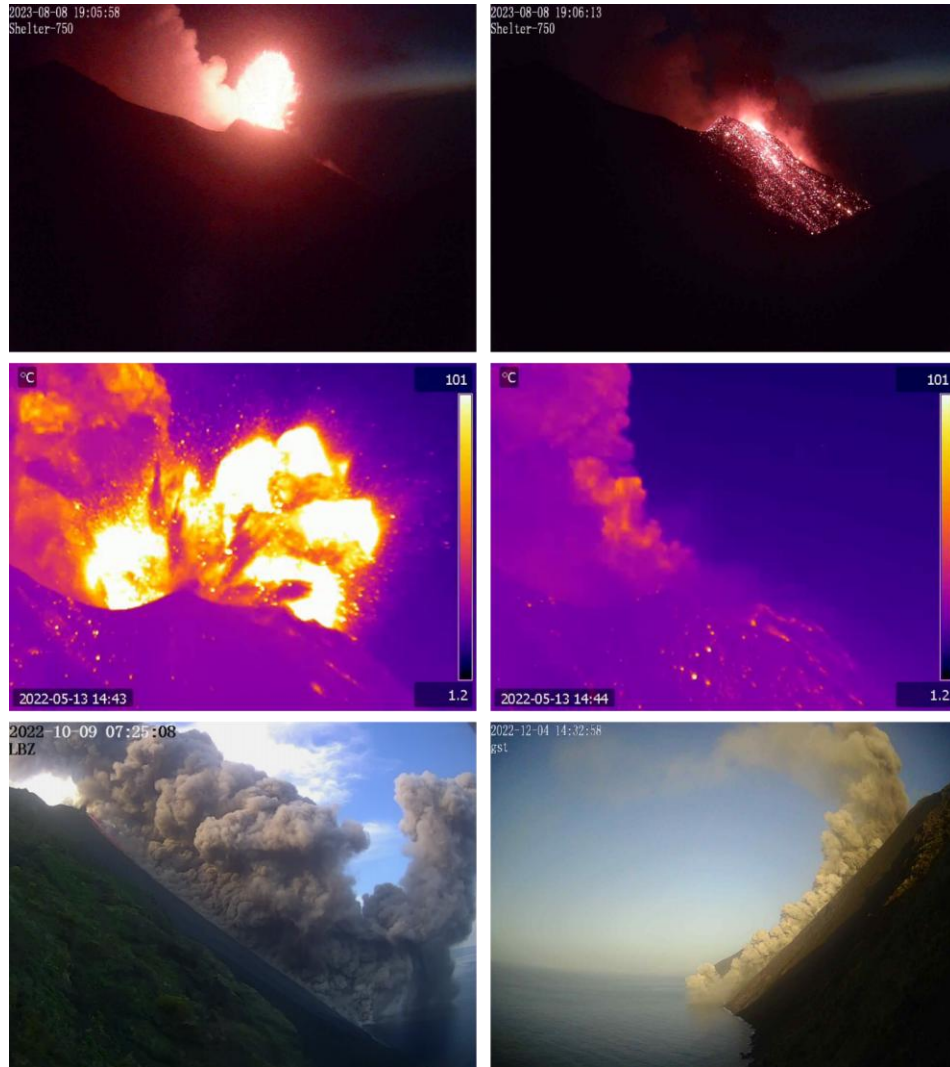


Figure 6.1 - Images of selected major explosions, as recorded by the LGS monitoring cameras (<http://lgs.geo.unifi.it/>). Top images, the 8 August 2023 major explosion, showing eruption onset (left) and the fallout dispersal area (right). Central images, the 13 May 2022 major explosion, showing the eruption onset (left) and the fallout dispersal area (right); Bottom images, pyroclastic density currents in the Sciara del Fuoco, formed during the October 2022 and 4 December 2022 collapse events.

6.3 Methods

6.3.1 Volcanic Gas Dataset

Statistical analysis is applied to a volcanic gas dataset (Figures 6.2b-e, 6.3) acquired by the geochemical monitoring network run by DiSTeM-Università di Palermo, between January 2020 and December 2023.

The SO₂ flux time series (Figure 6.2b) is obtained by processing sets of images acquired by a permanent, fully automated UV Camera system. Functionalities of the UV Camera, including data acquisition and processing routines, have been described elsewhere (Delle Donne et al., 2017, 2022; Lo Bue Trisciuzzi et al., 2024). Results, listed in Supplementary Table S6.1 and illustrated in Figure 6.2b, are expressed in the form of daily averaged SO₂ fluxes (metric tonnes per day, t/d), obtained by averaging all successful flux quantifications obtained (at 0.5 Hz rate) during 6h-long daily temporal windows (acquisition limited to best sunlight illumination conditions).

Figures 6.2c-e are time series of plume SO₂ and CO₂ concentrations (in ppmv), and of their relative proportions in the plume (the molar CO₂/SO₂ ratio), obtained by processing data streamed by two fully automated Multi-GAS units, located in the summit crater area (~500 m distance from the crater terrace). The Multi-GAS units (see Aiuppa et al., 2021 for detailed descriptions) are configured to capture (at 1 Hz rate) the output signals from a set of pre-calibrated, gas-specific sensors, mounted in series, and exposed to the target gas plume. Acquisitions are automatically activated (by a datalogger) at regular intervals (typically every 4 hours), and their duration is modulable (from ~300 to 1800 s), depending on the SO₂ concentration levels encountered to save power (the unit is programmed to halt acquisitions below a pre-set concentration threshold). The sensors' output signals (counts, or mV) are automatically converted into gas concentrations (ppmv) using lab-determined calibration functions (stored in the datalogger). These concentration time series are then processed in near-real time by a R-based script that sequentially scans the dataset using a 60 s-long moving window, searching for (and identifying) intervals with (i) high temporal coherence (Pearson correlation coefficient ≥ 0.9) between CO₂ and SO₂ concentrations time series, and (ii) SO₂ concentrations at/above a 1 ppm threshold. We note that more conservative (higher) SO₂ concentration thresholds have been used in previous work (e.g., Aiuppa et al 2009). For each 60 s temporal window in which both conditions above are met, the algorithm saves the maximum SO₂ concentration (shown in Figure 6.2c), the range (maximum – minimum) Δ SO₂ and Δ CO₂ concentration levels (the latter shown in Figure 6.2d), and calculates the corresponding CO₂/SO₂ molar ratio (Figure 6.2e) (Aiuppa et al., 2021). Use of the Δ CO₂ concentration helps to resolve the volcanic concentration signal (as expressed by the maximum) above the atmospheric background (as approximated by the minimum; note the atmospheric background is negligible for SO₂). The ratio is obtained from the slope of the best-fit linear regression function calculated from the Δ SO₂ and Δ CO₂ concentration time series within the processing window (Aiuppa et al., 2021). Circa 155,000 triplets (maximum SO₂ and Δ CO₂ concentrations, and CO₂/SO₂ ratios) of data were obtained during 2020-2023, all listed in Supplementary Table S6.2 and in Figure S6.1. Supplementary Tables S6.3, S6.4 and S6.5 list the statistical parameters of SO₂ and CO₂ concentrations and CO₂/SO₂ ratios, all

calculated on a 3-day basis (the statistical parameters are assigned to the first of the 3 measurement days). We utilize the graphic representation from Aiuppa et al. (2021) solely for illustrative purposes, rather than statistical analysis, to highlight temporal changes in plume chemistry. In these plots, data (Supplementary Table S6.2) are presented as chronograms of normalized frequency distribution diagrams (Figure 6.2c-e). To this aim, sets of individual frequency distribution plots are initially calculated from all concentration/ratio results obtained within each 3-day interval. Next, a centered smoothing function is applied to sets of 5 adjacent frequency distribution plots (covering 15 days in total) to filter out short-term (hourly to daily) fluctuations and emphasise longer term (multi-day to weekly/monthly) trends. In the resulting plots (Figures 6.2c-e, 6.3), warmer tones (dark red) correspond to the median of the frequency distribution, while cooler colours (yellow to green to blue tones) correspond to progressively less represented frequency bins. The plots therefore convey information on temporal fluctuations of: (i) the most-represented concentration levels/ratios, and (ii) on the spread around them.

Finally, the volcanic CO₂ flux (Figure 6.2f and Supplementary Table S6.1) is obtained by combining the CO₂/SO₂ ratio record of Figure 6.2e with the daily-averaged SO₂ flux (Figure 6.2b).

We note that the fraction of data gaps in the Multi-GAS time series is <5% of the total observations. According to theory (Bennet, 2001), this low data gap fraction has marginal impact on the statistical analysis discussed below.

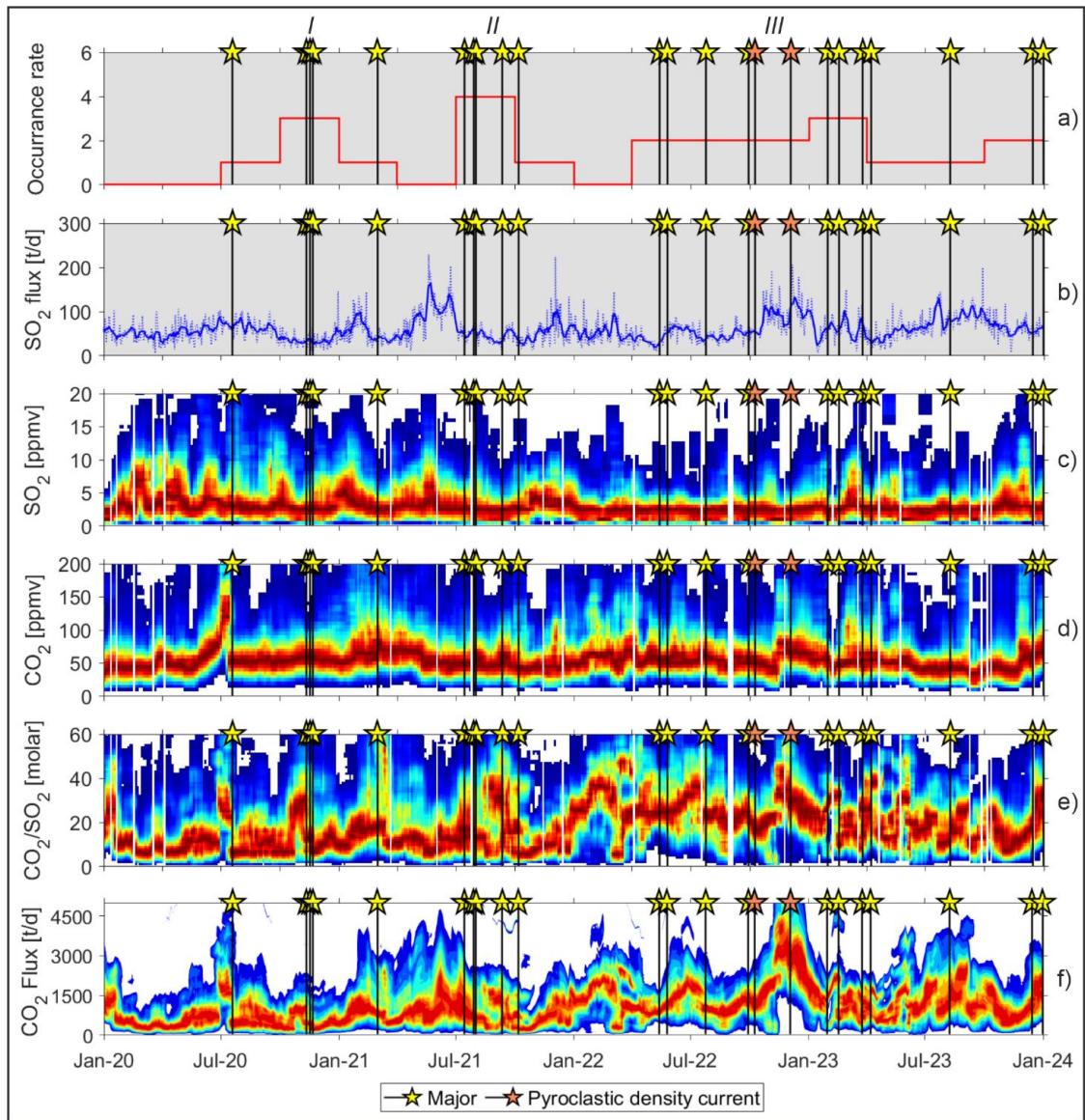


Figure 6.2 - Four years (2020-2023) of gas observations at Stromboli. (a) Rate of occurrence (number of events/3 months) of major explosions. Yellow and orange stars identify major explosions and collapse/pyroclastic density currents, respectively. Three main clusters are identified (labelled I, II and III); (b) Daily averaged SO_2 fluxes (t/d) measured by Stromboli's permanent UV camera (the solid line is 30-day moving average); (c) to (f), temporal plots of (normalised) frequency distribution diagrams of (c) SO_2 and (d) CO_2 concentrations in the plume, CO_2/SO_2 (molar) ratios (e) and CO_2 fluxes (f). Dark red tones show information on the temporal fluctuations of the dominant concentration/ratio levels, whilst orange to blue tones indicate increasingly less represented frequency bins.

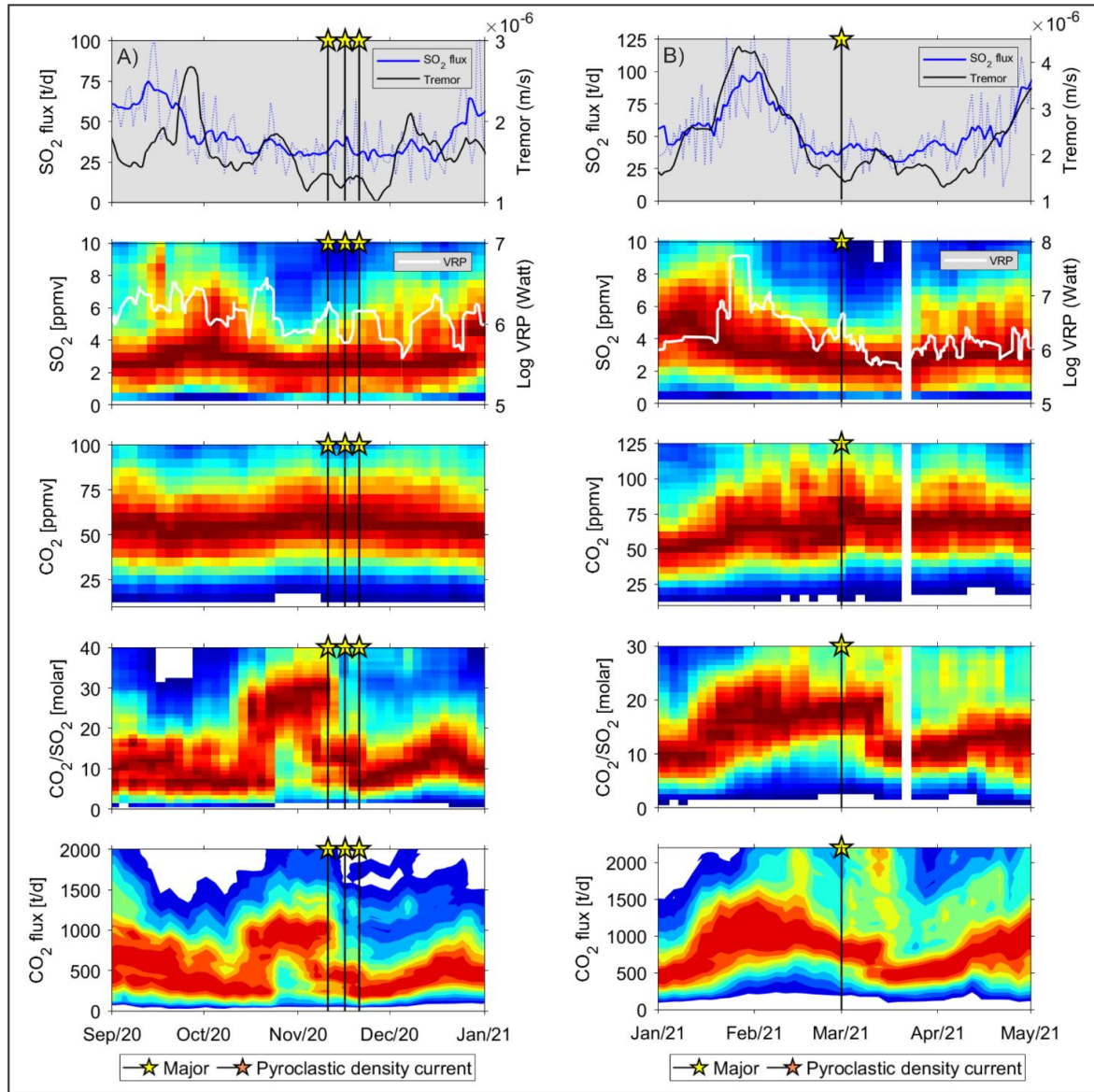


Figure 6.3 - A zoom of the compositional evolution of Stromboli's plume (Figure 6.2) in two selected intervals. The temporal record of volcanic tremor amplitude and the Volcanic Radiative Power (VRP, Watt), derived from satellite thermal observations, are also shown for comparison.

6.3.2 Statistical analysis

We use a Conditional Logit model (Logan, 1983) to investigate the statistical link between the occurrence of major explosions at Stromboli and the temporal changes in gas composition and flux. More specifically, independent models are explored that separately analyse the association between major explosions and: 1) the SO₂ flux (daily averages; Supplementary Table S6.1); 2) the maximum plume SO₂ concentration, 3) the plume Δ CO₂ concentration, 4) the plume CO₂/SO₂ ratio, and 5) the CO₂ flux. For SO₂ and Δ CO₂ concentrations, and for the plume CO₂/SO₂ ratio, the non-averaged data reported in Supplementary Table S6.2 are used.

The Conditional Logit model (Logan, 1983) is commonly used in the context of time series analysis to model categorical outcomes over time. If applied to a dataset consisting of a temporal sequence of repeated observations (as is the case in volcano monitoring), the model can be used to infer how/if different levels of a certain parameter influence the likelihood of a certain event. Lagged variables, in the context of statistical analysis or time series modelling, refer to variables that are delayed in time. In the Conditional Logit model, lagged variables are typically utilized to capture the effect of past exposure on a given event. In our specific case, our analysis aims to estimate if the probability of having a major explosion at a given time t increases when/if a specific geochemical pattern (e.g., high/low levels of gas concentration/ratio/flux) is observed at time t -Lag (where Lag is the lag time). In our case, a lagged variable corresponds to the daily average of the given parameter at t -Lag. The lagged effects are evaluated to up to 15 days (Figure 6.4a) and our models take the following form:

$$\text{logit}(P(\text{event}_t)) = \beta_0 + \beta_1 \text{Lag}_1 + \beta_2 \text{Lag}_2 + \dots + \beta_{15} \text{Lag}_{15} \quad (\text{Eq. 6.1})$$

where $P(\text{event}_t)$ is the probability of having a major explosion at time t , logit is the natural logarithm of the odds of the event occurring, and the β_i are the coefficients corresponding to each lagged variable, representing the impact of that variable on the log odds of the event occurring. To estimate β_i , we solve for the values that maximize the log-likelihood function through an iterative optimization algorithm. To obtain the odds of the event occurring, which indicate how the odds of the event occurring change for a one-unit increase in each lagged variable, holding all other lagged variables constant, we use the following formula:

$$OR_i = e^{\beta_i} \quad (\text{Eq. 6.2})$$

where OR_i is the odds ratio associated with the lagged variable Lag_i (Figure 6.4).

In a separate analysis, the odds ratio (OR) and 95% confidence intervals are calculated using as explanatory variables the right-aligned moving averages (7, 15 and 30 days) of the five parameters analysed (see Figure 6.5), instead of using the lagged variables. Use of the moving average allows testing if the temporal persistence (over timescales of 7 to 30 days) of a specific geochemical pattern impacts the probability of a major explosion happening at time t . This analysis is hence complementary to that obtained using the lagged variables, that are based instead on observations made at a specific t -lag time. Note that, in both types of analysis, each major explosion is treated as a separate case, even if part of a cluster of closely spaced explosions (Figure 6.2a).

Statistical significance is set at $p\text{-value} < 0.05$ (α), where the p -value is a measure of the strength of evidence against the null hypothesis ($\beta_i = 0$ or $OR_i = 1$ in our case). In other words, the p -value tells us if the association between the considered explanatory variable(s) and the event is statistically significant. The α level is the predetermined threshold used to assess the statistical significance of the

p-value and represents a Type I error, i.e., the probability of rejecting the null hypothesis when the null hypothesis is true.

We also perform a ROC (Receiver Operating Characteristic) analysis (Figure 6.6) with the aim of testing the ability of the monitored variables (SO₂ flux, SO₂, CO₂ and CO₂/SO₂) to “predict” the likelihood of a major explosion, and hence select their optimal cutoff values. These correspond to the values of the plume SO₂ flux, SO₂, CO₂ and CO₂/SO₂ closest to the perfect ROC curve (having both sensitivity and specificity equal to 1). The ROC analysis is conducted on both the lagged variables (Lag3, Lag5 and Lag7; Figure 6.6a-d) and on the moving averages of each parameter (moving averages at 7, 15 and 30 days; Figure 6.6e-h). A ROC curve (Figure 6.6) is a graphical tool that illustrates the performance of a binary classifier model, and it is built by plotting the True Positive rate (TPR, or sensitivity) against the False Positive rate (FPR, or 1 - specificity) at varying threshold values. In our application, TPR (sensitivity) and FPR (or 1 – specificity) are defined as the proportion of actual major explosions correctly identified by the model using the optimal cut-off, and the proportion of actual non-major events correctly identified by the model, respectively. Performance of the binary classification model across all possible threshold values is estimated using the area under the curve (AUC) (significance expressed by AUC>0.5; tested using the method described by DeLong et al., 1988) (Supplementary Tables S6.6 and S6.7).

Ultimately, four reduced models based on the significant lagged variables are obtained and the relative risks (RR; Figure 6.7a-d) are derived as follows:

$$RelativeRisk = \frac{P_{event=1}}{P_{event=0}} \quad (Eq. 6.3)$$

Using a similar formula, the RR for the statistically significant (Figure 6.5) moving averages are also obtained (Figure 6.7e-h).

We finally implement a Composite indicator (Figure 6.8) to combine the relative risk of the significant models, using an Additive Value Model (AVM) (Cilluffo et al., 2024). The AVM is a linear approach that quantifies the combined effect of multiple factors. Steps in the analysis include: (i) identifying the relevant dimensions, (ii) assigning their weights, (iii) linearly combining the weighted parameters, and (iv) interpreting the results. The resulting value provides a concise representation for communication and evaluation. In our study, we identify the 15-day moving average SO₂ relative risk (orange in Figure 6.7f) and the 15-day moving average CO₂/SO₂ ratio relative risk (orange in Figure 6.7h) as key attributes for our composite indicator, and the weights are optimised to maximise accuracy. The final Composite indicator assumes values between 0 and 1 (Figure 6.8). High values indicate increased eruption probabilities, while low values identify background activity with low eruption probability.

All the statistical analyses were performed with the R version 4.0.2 (R Foundation for Statistical Computing, Vienna, Austria). Statistical significance is set at $p < 0.05$.

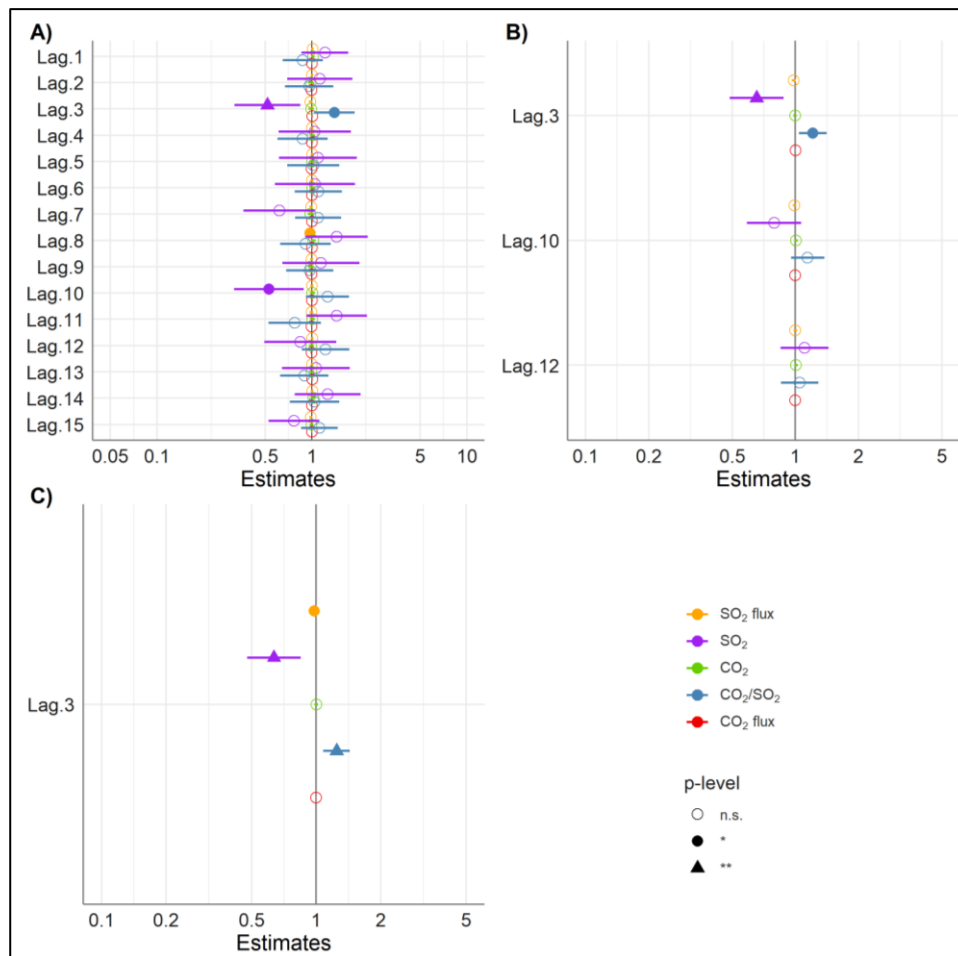


Figure 6.4 - OR and 95% confidence intervals of the four full conditional logistic regression models. *p*-levels: Open symbols, n.s., not significant. Filled circles, statistically significant (circles and tringles indicate increasing levels of significance, denoted by one or two asterisks).

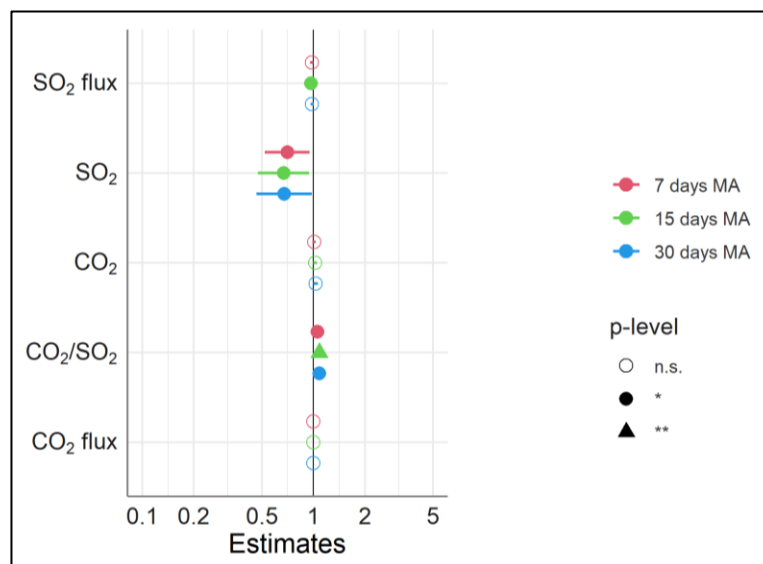


Figure 6.5 - OR and 95% confidence intervals for 7, 15 and 30 days moving averages of the four parameters analysed. p-levels: Open symbols, n.s., not significant; filled circles, statistically significant.

6.3.3 Volcanic tremor and volcanic radiative power (VRP)

To better characterise Stromboli's activity in the run-up to a major explosion, we complement our volcanic gas results with the temporal record of volcanic tremor and volcanic radiative power (VRP). Volcanic tremor, the most typical seismic parameter recorded on active volcanoes, is generally related with the upward migration of magma and gases in the plumbing system and hence provides useful information on volcanic activity evolution (McNutt, 2002; McNutt and Nishimura, 2008). Links between degassing and tremor have been investigated in different volcanic systems (Williams-Jones et al., 2001; Edmonds et al., 2003; Patané et al., 2013; Zuccarello et al., 2013; Delle Donne et al., 2022), and increases in seismic tremor amplitude are typically observed prior to and during eruption (e.g., Brandsdottir and Einarsson, 1992; Fischer et al., 1994; Bonaccorso et al., 2011; Nadeau et al., 2015; Valade et al., 2016; Ripepe et al., 2009; Aiuppa et al., 2023). Tremor at Stromboli is interpreted to be caused by the self-induced resonance oscillations of a bubble cloud (Chouet et al., 1997) in the shallow conduit (<200 m) or by the repeating pressure drop of self-coalesced gas bubbles bursting at the surface of the magmatic column (Gordeev, 1993; Ripepe and Gordeev, 1999). The strong correlation of tremor amplitude with infrasonic pressure energy released in the atmosphere implies a causal link with degassing processes in the shallow portion of the magmatic column (Ripepe et al., 1996). Several studies (e.g., Delle Donne et al., 2022; Aiuppa et al., 2023, Lo Bue Trisciuzzi et al. 2024) have found experimental evidence for the correlation between volcanic tremor and degassing processes, and the temporal increases of both parameters is typically associated with phases of escalating magmatic activity (Ripepe et al., 2009). We use the daily average of RMS (Root Mean Square) tremor amplitude, calculated on 1-minute-long time window of seismic signal filtered in band

0.9–10 Hz (Figures 6.3, 6.9). This frequency band includes the main spectral content of volcanic tremor energy at Stromboli (Chouet et al., 1997). Data streamed from the LGS ROC station is used.

The heat flux produced by volcanic activity is generally measured by means of the Volcanic Radiative Power (VRP), a composite measure of the area and temperature of the observed volcanic source (Coppola et al., 2023). Here we use the data from the MIROVA system (Middle Infrared Observations of Volcanic Activity) which ingests infrared data acquired by the Moderate Resolution Imaging Spectroradiometer (MODIS) and Visible Infrared Imaging Radiometer Suite (VIIRS) sensors to provide near-real time estimates of the VRP (Campus et al., 2022; Coppola et al., 2023). Previous studies at Stromboli have shown that the ordinary, mild-explosive activity is typically associated with VRP in the 1-30 MW range, with periodic fluctuations strictly related to variations in the level of the convective magma column that feeds the craters (Coppola et al., 2014; 2016; Laiolo et al., 2022; Massimetti et al., 2024). Higher VRP values (e.g. close to 100 MW) are typically associated with sustained spattering activity that often precedes short-lived lava overflows or longer effusive eruptions (Laiolo et al., 2022).

6.4 Results

The daily averaged SO₂ fluxes fluctuate widely during 2020-2023, ranging from 6 to 230 tons/day (mean, 60±30 t/d; Figure 6.2b). The maximum in plume SO₂ concentrations also vary widely, from 1 ppmv (the selected concentration threshold) to 38.6 ppmv (Supplementary Figure S6.1). The median SO₂ concentrations (3-day intervals; dark red tones in Figure 6.2c) fluctuate at around 1.9 to 6.8 ppmv level (see Supplementary Table S6.3). SO₂ concentrations > 10 ppmv are hence statistically less frequent (Figure 6.2c) at the Multi-GAS measurement points. The ΔCO₂ concentrations (Figure 6.2d) vary from < 10 ppmv to > 200 ppmv (see Supplementary Table S6.4), with the median level (dark red tones in Figure 6.2d) typically oscillating at ~50 ppmv (range, 35 to 129 ppmv; see Supplementary Figure S6.1). The largest ΔCO₂ concentrations are observed in the weeks prior to the July 19, 2020 major explosion, the largest of this class of events (in the 2020-2023 interval) from instrumental (Ripepe et al., 2021) and physical volcanology (Voloschina et al., 2023) viewpoints. Our extensive dataset confines the mean and median plume CO₂/SO₂ (molar) ratios at 20.9 and 17.7, respectively. The ratio range is extremely wide (1.2 to 809; Supplementary Table S6.2), and the 3-day interval medians (dark red tones in Figure 6.2e) range from ~7 to ~38 (Supplementary Table S6.5). The median CO₂/SO₂ ratio levels are characteristically higher in 2022 to early 2023 (Figure 6.2e), in conjunction with a main cluster of major explosions (Figure 6.2a).

Qualitative, visual inspection of Figure 6.2 highlights non-uniform, case-specific plume composition behaviour prior to each of the 24 studied explosive events. We yet identify some

commonalities, such as the elevated CO₂/SO₂ ratio observed in the days to weeks preceding several major explosions (Figure 6.2e), as previously found (Aiuppa et al., 2011; Voloschina et al., 2023). To illustrate this aspect more clearly, we selected two cases (Figure 6.3a-b and Supplementary Figure S6.2) where the gas plume compositional changes prior to/after an explosive event are more evident. An especially well-behaved gas plume evolution is observed corresponding to three major explosions in November 2020 (Figure 6.3a). The plume CO₂/SO₂ ratio visibly increases starting from ~3 weeks before the first explosion, from ~10 (and certainly <<20) to ≥30. This CO₂/SO₂ ratio increase is also concurrent with a phase of decreased SO₂ fluxes and plume SO₂ concentrations, and of slightly (but appreciably) increasing plume CO₂ concentrations and fluxes (Figure 6.3a). Hence, the three explosions all occurred during a phase of reduced SO₂ release relative to CO₂. Importantly, this behaviour is not unique to the November 2020 explosions. For example, remarkably similar plume evolutions are observed prior to explosions in early 2021 (Figure 6.3b) and in mid-to-late 2021 (Supplementary Figure 6.2a). More complex and ambiguous compositional trends are observed in other temporal intervals (Supplementary Figure 6.2b-d). In the October 2022 to April 2023 period (Supplementary Figure 6.2c), increasing CO₂/SO₂ ratios are still observed weeks prior to all events recorded; however, while SO₂ fluxes and SO₂ concentrations drop down in correspondence to the two winter 2023 explosions, they do not prior to the December 4 2022 event. The January-July 2022 (Supplementary Figure 6.2b) and May-October 2023 (Supplementary Figure 6.2d) events are examples of major explosions which are not preceded by any obvious precursory CO₂/SO₂ ratio change. Rather, in both periods we identify phases (e.g., January to March 2022) in which the CO₂/SO₂ ratio is sustainedly high, and yet no major explosion is observed.

Results of the application of the Conditional Logit model to the volcanic gas compositional time series are illustrated in Figures 6.4 and 6.5. We find a statistically significant association between the occurrence of major explosions and volcanic gas composition (SO₂ concentration and CO₂/SO₂ ratio) at Lag 3 levels (Figure 6.4a,b). Since only Lag 3 shows significant results, we estimate five reduced models (Figure 6.4c) including only Lag 3 of all explanatory variables (SO₂ flux, SO₂, CO₂ and CO₂/SO₂ ratio, CO₂ flux). Of the reduced models (Figure 6.4c), a unit decrease in SO₂ concentration is significantly associated with an increased probability of having a major explosion at Lag 3 (OR = 0.635, 95% CI: 0.476–0.848, p=0.002). Likewise, a unit decrease in the SO₂ flux is significantly associated with an increased probability of having a major eruption. By contrast, a 5 unit increase in the CO₂/SO₂ ratio is significantly associated with an increased probability of having a major explosion over the same (Lag 3) temporal window (OR = 1.243, 95% CI: 1.078– 1.434, p=0.003) (Figure 6.4). CO₂ concentrations and CO₂ flux do not exhibit any statistically significant relationship with the occurrence of major explosions.

The same models applied to moving averages (Figure 6.5) indicate that a unit decrease in the 7-days SO₂ flux moving average is significantly associated with an increased probability of having a major explosion. Likewise, a unit decrease in the 7-15-30 days moving averages of the SO₂ concentration is significantly associated with an increased probability of having a major explosion. By contrast, a unit increase in the 7-15-30 days moving average of the CO₂/SO₂ ratio is significantly associated with an increased probability of a major explosion occurring (Figure 6.5). Again, no statistically significant association is found for the CO₂ flux, which is not considered further in the analysis below.

Figure 6.6a-d illustrates the ROC curves for the variables considered at Lag 3, 7 and 10 days. All considered variables at Lag 3, except for CO₂ concentrations, show moderate ability to predict major explosions (Supplementary Table S6.6), as evidenced by the AUC >0.5 but <0.7 (Swets, 1988). The AUC of the SO₂ flux is 0.63 (95% CI: 0.54-0.71) and the optimal threshold (given by the best combination of sensitivity and specificity) is 47 t/d, associated with a 58% sensitivity and a 61% specificity. The AUC of the SO₂ concentration is 0.706 (95% CI: 0.601-0.807), and the optimal threshold is 3.2 ppmv, associated with a 63% sensitivity and a 71% specificity. The AUC of the CO₂/SO₂ ratio is 0.67 (95% CI: 0.53-0.77), and the optimal threshold is 27.8, associated with a 58% sensitivity and a 69% specificity. At Lag 7, the CO₂/SO₂ ratio optimal threshold is 26.6, associated with a 66% sensitivity and a 67% specificity (Supplementary Table S6.6). Similar results are obtained using the moving averages at 7, 15 and 30 days (Figure 6.6e-f; Supplementary Table S6.7). 15-days moving average of all considered variables, except for CO₂ concentrations, show moderate ability to predict major eruptions. The AUC of the SO₂ flux is 0.62 (95% CI: 0.51-0.73) and the optimal threshold is 54 t/d, associated with a 67% sensitivity and a 54% specificity. The AUC of the SO₂ concentration is 0.65 (95% CI: 0.54-0.75), and the optimal threshold is 4.3 ppmv, associated with a 75% sensitivity and a 57% specificity. The AUC of the CO₂/SO₂ ratio is 0.66 (95% CI: 0.57-0.76), and the optimal threshold is 24.7, associated with a 67% sensitivity and a 59% specificity (Supplementary Table S6.6).

The Relative Risks (RRs) obtained using the lagged variables (Figure 6.7a-d) show spiky behaviour, except for the CO₂ concentration trend that is flat and uniform throughout the entire observation period. The RRs obtained using the moving averages (Figure 6.7e-h) exhibit more well-behaved trends, peaking prior to many of the explosive events.

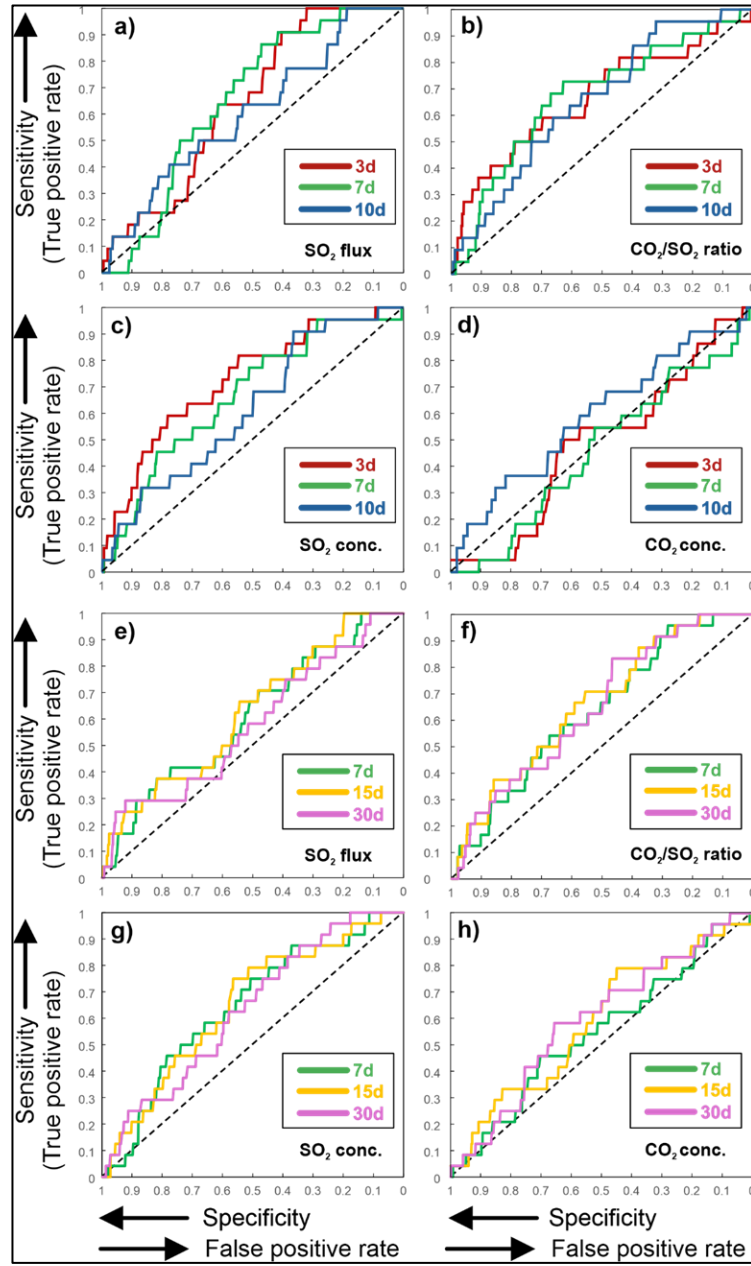


Figure 6.6 - Receiver Operating Characteristic (ROC) curves illustrating the ability of the four monitored volcanic gas parameters to predict the occurrence of major explosions. Plots a) to d) are based on the lagged variables, while e) to h) are based on 7, 15, 30 days moving averages.

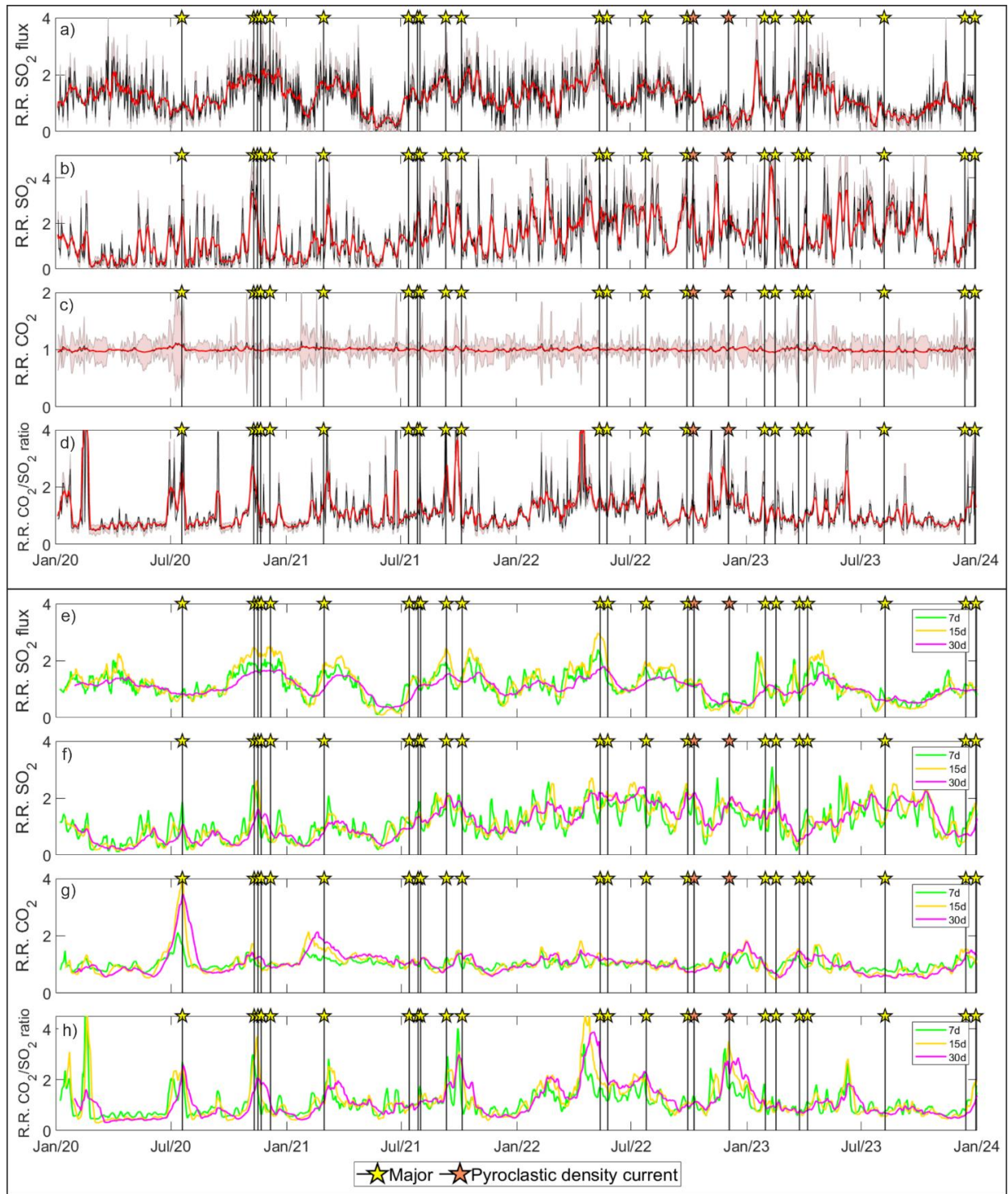


Figure 6.7 – Time series of the Relative Risks (RR) (see. Eq. 6.3) for the four distinct Lagged (3-days) variables (a to d) and for the 7, 15, 30 days moving averages (e to h).

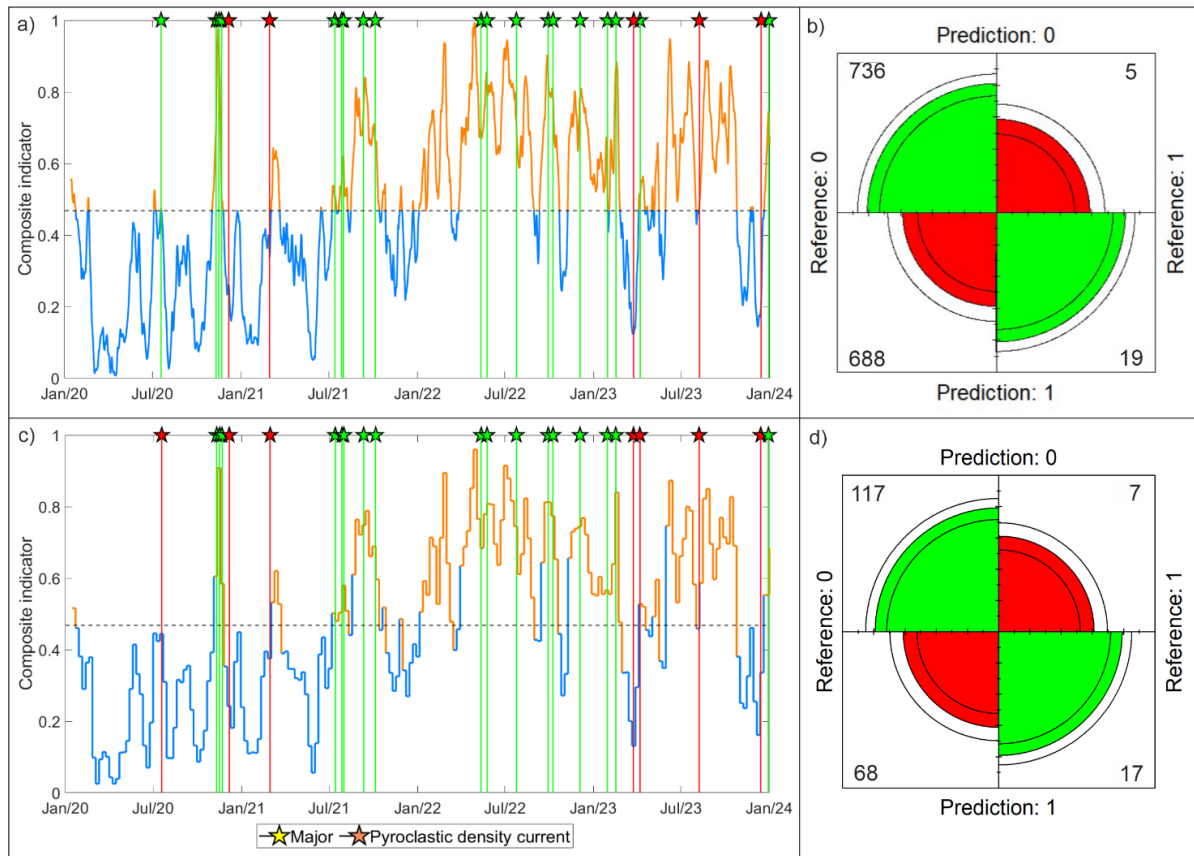


Figure 6.8 - (a) Time series of the Composite indicator, calculated on a daily basis. The Composite indicator is obtained by linear combination of the 15-day moving average SO₂ Relative Risk (light orange in Figure 6.7f) and of the 15-day moving average CO₂/SO₂ ratio Relative Risk (light orange in Figure 6.7h). The blue/red colour tones correspond to values below/above the optimal threshold (0.468). The 24 explosive events are indicated by stars; green stars indicate explosions occurring on days with above threshold composite indicator (True Positives, 19 events), while red stars indicate explosions occurring on days with below threshold composite indicator (False Negatives, 5 events); (b) Confusion matrix computed from the daily values of the Composite indicator (panel a); (c) Weekly averaged time series of the Composite indicator. Blue/red line as in panel a. Green and red stars are explosions identified as True Positives (17) and False Negatives (7), respectively, as based on the confusion matrix of panel (d). This confusion matrix is based on the temporal persistence of the Composite Indicator. True positives are explosions preceded by 7 consecutive days with above threshold Composite indicator, while false negatives are explosions that are not preceded by 7 consecutive days with above threshold Composite indicator.

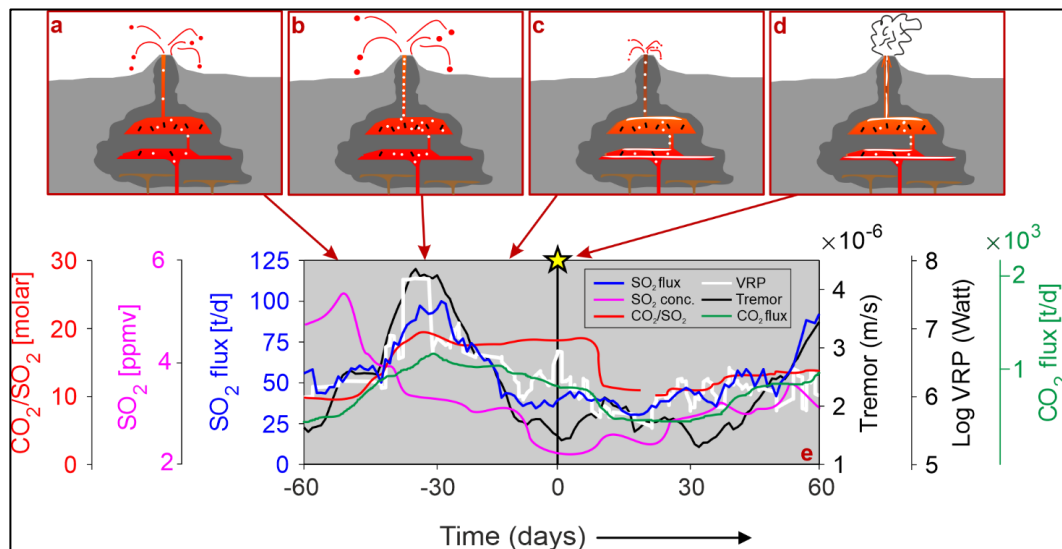


Figure 6.9 - A simplified, observation based, conceptual models of the sequence of events (a to d) leading to a major explosion. See text. The bottom panel (e) illustrates the temporal evolution of gas (data from Figure 6.3b), geophysical (volcanic tremor amplitude; from LGS monitoring network; Ripepe et al., 2009) and thermal output (Volcanic Radiative Power, VRP, from the MIROVA platform, Coppola et al., 2016) records from Stromboli (exemplified by the January to May 2021 event). Time is expressed as days prior to/after the major explosion (set time = 0).

6.5 Discussion

6.5.1 Are gas changes statistically significant?

Our results identify some characteristic patterns in volcanic gas flux/composition time series prior to Stromboli's major explosions. We find that most major explosions in 2020 to 2023 were preceded by periods (of days to weeks) of reduced plume SO_2 concentrations (highly significant), low SO_2 fluxes and increased CO_2/SO_2 ratios (significant) (Figures 6.2, 6.3). The similar association found for SO_2 fluxes and plume SO_2 concentrations corroborates a causal link between reduced SO_2 and increased eruption likelihood. Analysis shows these variations are statistically significant (p values range from 0.002 to 0.02) over temporal timescales of days (Lag 3 in Figure 6.4) to weeks (Figure 6.5). The same timescales are supported by ROC analysis (Figure 6.6), evidencing the repetitive nature of this pattern. Hence, our novel data and analysis provide quantitative statistical validation for the previously reported experimental evidence of a causal link between high CO_2/SO_2 phases and larger-than-ordinary Strombolian explosions at Stromboli found in previous work (Aiuppa et al., 2009, 2010a, 2011), yet in which an higher SO_2 concentration threshold was used (relative to the ≥ 1 ppm level used here and in Aiuppa et al., 2021). In contrast, CO_2 concentrations do not exhibit any statically significant pattern prior to major explosions. We caution that, in addition to reflecting some specific eruption trigger mechanisms (see section 6.5.3), the low significance of CO_2 concentrations may also result from instrumental limitations. The CO_2 -sensing spectrometer has an accuracy of $\pm 2\%$ of the full scale (3000 ppm in our case), resulting in a ± 60 ppm accuracy. Below 4

ppm SO₂, the median Δ CO₂ is approximately 47 ppm, which is below the sensor's accuracy threshold. This indicates that at low concentrations, the CO₂ sensor may lack sufficient precision to accurately resolve CO₂ levels.

Nearly continuous, long-term records, as those available at Stromboli, are now needed at other persistently active volcanoes (that have a frequent enough eruptive activity) to test the general validity of the statistical association found here. For example, Kunrat et al. (2022) recently applied statistical analysis to SO₂ flux time series acquired at Sinabung volcano (Indonesia) and found a systematic drop in SO₂ emissions in the 5 days before individual explosions. This case, with the Stromboli example presented here, shows that statistically significant gas changes may be common at volcanoes, and that the paucity of past examples in the literature likely reflects the lack of continuous, long-term gas records at most degassing volcanoes worldwide.

6.5.2 Evaluating eruption forecast performance

Forecasting volcanic eruptions, a fundamental goal of volcanology (Sparks, 2003), requires that the probability of a future volcanic event is inferred and communicated – together with its associated uncertainty – to governmental authorities and decision makers (Poland and Anderson, 2020). Despite recent major progresses (Acocella et al., 2023), eruption forecasting (deterministic and probabilistic) remains a challenge, especially for sudden, low magnitude (VEI<3) eruptions (Stix et al., 2023).

Our results can be used to quantitatively evaluate the benefits and limitations of volcanic plume geochemical observations as eruption forecasting tools at Stromboli. To this aim, we combine the Relative Risks (RRs) derived from statistical analysis of the individual gas parameters' time series (Figure 6.7) into a Composite gas indicator (Figure 6.8a). This indicator expresses a gas-based prediction - on a 0 to 1 scale - of having a major explosion, and is based on the linear combination of RRs for the statistically significant moving averages of the SO₂ concentrations (relative weight, 75%) and CO₂/SO₂ ratios (relative weight, 25%) (Figures 6.5, 6.6). Notably, although SO₂ concentrations and fluxes are found to have similar association with major explosions (Figures 6.3, 6.6), the latter shows less statistical significance than the former. In fact, tests show that inclusion of the SO₂ flux in the Composite indicator reduces its performance (as inferred by the confusion matrix, see below). As discussed previously (Delle Donne et al., 2017, 2022; Lo Bue Trisciuzzi et al., 2024), the currently operating UV Camera system offers an incomplete view of the Stromboli's bulk plume, because the plumes issuing from the central and southwestern craters are partially hidden by the northeastern crater ridge. This was found to result in a net underestimation of the total SO₂ flux of ~30% (Delle Donne et al., 2022; Lo Bue Trisciuzzi et al., 2024). As many of the major explosions studied here were sourced by vents within the central/southwestern crater terrace, we argue that the UV Camera

may have recurrently failed in capturing in full the pre-explosion drop in SO₂ flux. Additional sources of error do include radiative transfer effects (e.g., plume opacity, light dilution; Kern et al., 2010a) that are not accounted for in the current analysis. Based on the results of our statistical analysis, and considering the Multi-GAS units are exposed to emissions from the entire crater terrace, we hence use the SO₂ concentrations as proxy for the volcano SO₂ emissions. The implications of this assumption will be elaborated upon further.

Results (Figure 6.8a) show that the Composite indicator varies widely during 2020-2023 (range, 0.08-0.90), and that major explosions are recurrently anticipated by increases to > 0.48 (the optimal threshold) days to several weeks before hand. We evaluate the eruption forecast performance of the indicator from its associated confusion matrix (Figure 6.8b), built based on the daily values. This exercise highlights that 19 (TP, True Positives) out of 24 explosions are preceded by increases of the Composite indicator, while 5 are not (FN, False Negatives); hence, the successful prediction rate ($100 \cdot TP / (TP + FN) = 19 / 24$) is 79%. The same analysis, however, additionally demonstrates a 48% ($100 \cdot 688 / 1448$) false positive rate (FPR), and a precision ($100 \cdot TP / (TP + FP) = 100 \cdot 19 / 707$) of only 2.7%. Finally, the overall correct classification rate (also known as accuracy; expressed by the relationship: $100 \cdot (TP + TN) / \text{TOTAL EVENTS} = 720 / 1448$) is 52%. In summary, these results show that our daily geochemical records, although statistically significant ($p\text{-value} < 0.05$; Figure 6.5), have poor predictive behaviour: daily gas-based alerts would be issued during nearly half of the observation interval, but 97.3% of them would in fact not be associated with a major explosion.

This analysis presents the challenges we face in estimating and communicating the hazard. First, the true positive alerts enable us to successfully predict the majority of major explosions (19 out of 24 in 2020-2023). Analysis of the four “unpredicted events” indicated that they are among the smallest in size from instrumental viewpoint, e.g., in terms of their associated ground tilt and seismic VLP (Very Long Period) displacement (Ripepe et al., 2021). Tilt and VLP seismicity are explained as due to volumetric expansion of the upper conduit/dyke feeding system, driven by rapid injection of deep gas+magma shortly (minutes to seconds) prior to the blast (Ripepe et al., 2021); hence, their magnitude scales with the volume of gas and/or gas-rich magma that is to erupt. The lower gas volumes of smaller-scale events, in addition to the high level of false positives (see below), may thus justify the lack of any statistically significant pre-blast pattern.

Secondly, it is evident from our analysis that a large fraction (97.3%) of the gas-based alerts (increases of the Composite indicator; Figure 6.8a) would be False positives (e.g., they would not be followed by a major explosion). False positives are especially problematic as they tend to undermine trust of users (decision makers, population) in the monitoring system. In our case, the high false positive rate likely results from the combined action of several factors. Firstly, it is possible that the

preparatory gas accumulation phase, as tracked by low SO₂ and high CO₂/SO₂ ratios in the plume, is a necessary but insufficient condition for the explosion: rather, we argue that the accumulated gas can be released passively (e.g., without resulting in an explosion) in most cases. Secondly, the presence of multiple vents with distinct gas compositions (Pering et al., 2020) in the crater terrace may render the plume time series noisy, and hence difficult to interpret. Thirdly, and perhaps more importantly, the plume gas concentrations detected by the Multi-GAS units (located at several hundred meters from the degassing vents) are not only modulated by volcanic degassing dynamics, but also by external factors, especially wind speed and direction that govern the extent of plume fumigation at the measurement sites. It is therefore possible that many false positives of the Composite indicator result from low wind speed/unfavourable wind directions, all concurring to determine low SO₂ concentrations. We also cannot exclude that the low SO₂ concentration alerts prior to major explosions are influenced by a combination of volcanological and meteorological effects.

To conclude, when communicating the hazard to authorities, pros and cons of gas observations should be clearly stated. Gas plume monitoring at Stromboli is clearly not yet at a sufficiently mature stage (with the potential of too many false alerts) to be used for daily communicating the hazard of impending major eruptions to authorities with a proper confidence level. Rather than relying on daily acquired data to make inferences on eruption probabilities, a correct use of gas information in decision making should be based on analysing the temporal persistence of anomalous gas changes over longer temporal intervals. We quantitatively explore this possibility by dividing our observational period in 7-day sub-intervals (the weekly averages of the Composite indicator are illustrated in Figure 6.8c). Next, we obtain a confusion matrix (Figure 6.8d) by analysing the temporal persistence (over a 7-day temporal interval) of the Composite indicator. In this case, we count as True Positives (17) explosions preceded by (at least) 7 consecutive days with above threshold Composite indicator, while False Negatives (7) are explosions that are not preceded by (at least) 7 consecutive days with above threshold Composite indicator (Figure 6.8d). With forecasting confined on this 7-day basis, the successful prediction rate ($100 \cdot TP / (TP + FN) = 17/24$) decreases to 71% (relative to 79% in the previous exercise) but, more importantly, the False Positive rate ($FPR = 100 \cdot 68/209$) drastically decreases to ~32%, and precision ($100 \cdot TP / (TP + FP) = 100 \cdot 17/85$) largely improves to 20%. The overall correct classification rate ($100 \cdot 134/209$) also increases to 64%. Our results and analysis then confirm that major explosions cluster in prolonged phases (of week(s)) during which the Composite indicator is above threshold (Figure 6.8c), and that hence any “guess” of eruption likelihood should be based on longer trends (Figure 6.8c) than daily (Figure 6.8a).

Access to Stromboli’s summit (above 400 m a.s.l.) has been unregulated for years, then regulated (2003-2019) and only recently permanently closed to tourists (2019-present). Access today

is limited to monitoring staff personnel since the July 3, 2019 paroxysmal explosion (Giordano and De Astis, 2021). As we wait for research to progress toward the identification of more powerful forecasting tools, to date the gas plume is the only reported mid-term (days to weeks) indicator for an increased likelihood of a major explosion (Figure 6.8). Exploiting the capabilities of gas observations - while bearing in mind their caveats and limitations – may enable safer access for volcanologists and monitoring staff.

6.5.3 Implications for the driving mechanisms of major explosions

Our observations (Figures 6.2, 6.3) and analyses (Figures 6.4, 6.6) also offer novel pieces of information to understand the driving mechanisms of sudden basaltic explosions. These relatively small ($VEI < 1$) but hazardous blasts are not unique to Stromboli (where they are referred as “major explosions” and “paroxysms”, based on their intensity/magnitude), but rather common to open-vent basaltic volcanoes globally (Vergnolle and Métrich, 2022). Owing to their characteristics, such as sudden initiation, short duration (seconds to minutes) and unsteady (unsustained column) behaviour, these blasts are generally thought to be driven by the catastrophic release of accumulated and pressurized volatiles (Stix et al., 2023); however, the processes responsible for driving the volcano to such a critical state can be variable in space and time, and include both bottom-up and top-down mechanisms (Stix et al., 2023).

At Stromboli, paroxysms have typically been explained as triggered by the fast ascent of either gas (Allard, 2010) or gas-rich magma (Métrich et al., 2001, 2010, 2021) from the deep magma plumbing system (bottom-up mechanism), but examples of top-bottom mechanisms, in which paroxysms are favoured by rapid decompression of deep-seated magma by shallow conduit emptying (caused by lateral lava effusion), also exist (Aiuppa et al., 2010a, 2021; Calvari et al., 2011; Ripepe et al., 2015). Conflicting views also exist for major explosions, since resolving between top-bottom (Falsaperla and Spampinato, 2003; Calvari et al., 2012) vs. bottom-up (Andronico and Pistolesi, 2010; Pioli et al. 2014; Voloschina et al., 2023) processes is complicated by more controversial geochemical/petrological evidence (erupted materials are heterogeneous mixtures of denser/degassed stored conduit magma fragments and vesicular/crystal-poor deeper LP magma fragments; see discussion in Voloschina et al., 2023 and Insinga et al., 2024).

One key point here is that our statistical analysis finds no systematic temporal association between major explosions and CO_2 concentrations/fluxes (Figures 6.4a, 6.5, 6.6, 6.7c,g). Owing to very low solubility in silicate melts, shallow stored magma is typically extremely depleted in CO_2 (Edmonds and Wallace, 2017). Hence, increasing CO_2 emissions at open-vent volcanoes (Aiuppa et al., 2010a; Edmonds et al., 2022) have characteristically been interpreted as caused by precursory

CO₂-rich gas bubble leakage from deeply stored, mafic magma, and hence evidence of a bottom-up eruption mechanism. At Stromboli, increasing CO₂ concentrations and fluxes in the weeks/months prior to the July 3 2019 paroxysm (Supplementary Figure S6.3; Aiuppa et al., 2021) have been explained by progressive bubble accumulation into, and passive leakage from, deeply stored LP magma, ultimately causing pressure build-up at depth, and LP magma ascent and eruption. Of the 2020-2023 major explosions studied here, only the July 2020 explosion, the largest of this class of events in terms of geophysical (tilt and VLP; Ripepe et al., 2021) and volcanological (Voloschina et al., 2023) parameters, is associated with a CO₂ concentration increase similar, or even larger, than that observed prior to the July 2019 paroxysm (Supplementary Figure S6.3). This, in combination with the CO₂ flux increase (Figure 6.2f and Voloschina et al., 2023), points to a bottom-up mechanism for the July 2020 event (and potentially for a few others in 2009, Aiuppa et al., 2011; Insinga et al., 2024). However, the lack of any distinctive CO₂ concentration/flux trend for the remaining 2020-2023 explosions (Figure 6.2), and the generally lower CO₂ fluxes relative to those observed in concomitance with the 2019 events (Supplementary Figure S6.3), require a distinct mechanism.

Our analysis shows that the pre-major explosion CO₂/SO₂ ratio increases in 2020-2023 (Figures 6.4, 6.6) are systematically caused by decreasing SO₂ concentrations (Figures 6.4, 6.5) rather than increasing CO₂ concentrations. Major explosions are also systematically clustered in periods of reduced SO₂ fluxes (Figures 6.2, 6.7; although with the limitations discussed above). This association is especially intriguing as the SO₂ flux is a proxy for the volume of magma that is ascending (and degassing) in the topmost ~3-4 km of the Stromboli's magma feeding system (Allard et al., 1994; Laiolo et al., 2022). Hence, the recurrent (Figures 6.2, 6.3) drop in SO₂ concentrations/fluxes prior to most of Stromboli's major explosions is suggestive of a process in which slowing down of shallow magma circulation (acting over timescales of days to weeks) creates the favorable conditions for major explosions to occur.

Our preferred hypothesis of a top-down control on major explosions is schematically illustrated in Figure 6.9. This simplified model, exemplified by the gas observations taken in January-May 2021 (Figure 6.3b), also builds on temporal records of (i) volcanic tremor amplitude (data from the Laboratorio Geofisica Sperimentale, UniFi network: Ripepe et al., 2009) and (ii) thermal output (as based on estimates of Volcanic Radiative Power obtained from infrared satellite data; Coppola et al., 2016). These two parameters are typically used to parametrize the level of activity of the shallow magmatic column at Stromboli, and they consistently increase when gas-rich, more buoyant magma circulates in the volcano's upper magma feeding system, hence sustaining more vigorous regular (Strombolian) activity (e.g., Valade et al., 2016), and decrease when the shallow gas±magma supply is reduced.

In our hypothesis, regular Strombolian activity (panels a and b) is fed by sustained circulation and degassing of magma in the upper conduit/dyke feeding system (Allard et al., 2008). The convecting magma column is relatively permeable to deeply sourced gas bubbles. If the upper feeding system is increasingly percolated (Burton et al., 2007) by deeply-sourced bubbles (panel b), as tracked by increasing CO₂ fluxes (panel e), Strombolian activity intensifies, volcanic tremor amplitude increases (because of an increased gas bubble coalescence and bursting; Ripepe and Gordeev, 1999), and SO₂ flux and thermal output (VRP) concurrently increase because bubbly magma is more buoyant and faster convecting, releasing more gas and heat to the surface (Laiolo et al., 2022). With time, however, flushing by deeply sourced (CO₂-rich) gas causes the shallow resident magma to dehydrate, to become more viscous and stagnant (Caricchi et al., 2024). This slowing down of the upper magma column (panel c) causes a waning of Strombolian activity, as tracked by decreasing seismic and thermal activity, and by declining SO₂ and CO₂ fluxes. Notably, the CO₂/SO₂ ratio remains high in this phase (c). We propose that the viscous magma acts as a rheological (permeability) barrier below which deep gas bubbles can accumulate. If so, the high CO₂/SO₂ ratios in phase (c) would result from a combination of ineffective SO₂ release from the stagnant (degassed) magma column (see decreasing SO₂ fluxes and concentrations) and passive leakage of some accumulated gas (Aiuppa et al., 2021), as corroborated by the CO₂ flux decreasing less than the SO₂ flux (panel e). The depth of the gas accumulation zone (the base of the rheological barrier) is difficult to constrain. However, the CO₂/SO₂ ratios of >23.6-31 that are statistically recorded in the run-up (3-15 days prior) to the “predicted” major explosions (Figure 6.8) imply source depths of ~2-4 km below the summit vents (based on the modelled pressure dependence of the magmatic gas CO₂/SO₂ ratio at Stromboli; Aiuppa et al., 2021). If this interpretation is correct, then the “unpredicted” 2020-2023 major explosions (Figure 6.8) may reflect shallower gas accumulation, a condition where the accumulated gas bubbles would have CO₂/SO₂ ratio unresolvable from background (passive) degassing. The fact these 5-7 events (Figure 6.8) are volumetrically less important (based on geophysical evidence) supports this shallower origin. Ultimately, as some overpressure level is reached, catastrophic release and rise of previously accumulated gas ($\pm$ some deep LP magma) takes place during one (or in a sequence of) major explosion(s) (panel d). Removal of viscous stagnant magma (as HP magma fragments) in the major explosion(s) re-establishes permeable gas flow and active magma circulation in the upper conduit/feeding dyke, ultimately rejuvenating Strombolian activity (return to phase a).

We caution that this model is only based on observational evidence (Figure 6.9), while quantitative (fluid-dynamic) modeling would be needed to critically test its feasibility. Nevertheless, the mid-term (days/weeks) feedback mechanism between the shallow and deep portions of Stromboli’s magma plumbing system it is based upon explains well the temporal fluctuations in

regular (Strombolian) activity (Ripepe et al., 2008), the clustering of major explosions in sequences of closely spaced events (Figure 6.2a), and their concentration in periods of reduced shallow degassing activity (Figure 6.9).

6.6 Conclusions

The statistical significance of plume compositional/flux records as potential forecasting tools of sudden, larger-than-normal (major) explosions at Stromboli has been tested. Our results and analyses identify a recurrent volcanic gas temporal pattern prior to Stromboli's major explosions (in 2020-2023), in which these events are statistically preceded (over timescales of days to weeks) by: (i) low SO₂ concentrations and fluxes and (ii) high CO₂/SO₂ ratios. We have integrated these results into a gas-based Composite indicator, from which we obtain a success prediction rate (as expressed by the TPR) of 71% on a weekly basis (e.g., by counting the number of explosions preceded by above threshold Composite indicator for at least 7 consecutive days). The TPR increases to 79% if daily values of the composite indicator are used instead (but precision is only 2.7 % in this case). The weekly Composite indicator, based on the weighted linear combination of statistically significant parameters (SO₂ concentrations and CO₂/SO₂ ratios), can be implemented in real-time from monitoring data and, if additionally tested and improved to improve the currently low precision (20%), may evolve into a potentially exploitable monitoring and decision-making tool in the future.

We interpret these variations in view of a top-down mechanism (Figure 6.9), in which: (a) convective overturning of shallow magma in the feeding conduit/dyke system slows down (potentially due to the dehydration effect of CO₂ flushing; Caricchi et al., 2024), causing (b) gas bubbles to accumulate below a rheological discontinuity (the base of the viscous plug, formed by stagnant shallow HP magma), and hence (c) the catastrophic release of accumulated gas (plus a mixture of some deep LP magma and fragments of the HP magma plug) in a (or sequence of) major explosion(s). We stress that our results do not preclude other driving mechanisms for major explosions. For example, a bottom-up mechanism involving ascent of mafic LP magma from depth has been identified on a petrological basis for the July 19 2020, major explosion (Voloschina et al., 2023). This event consistently exhibits the most evident Δ CO₂ variation (Figures 6.2, 6.7g) in our dataset, and fails to be “predicted” by our Composite indicator (Figure 6.8), suggesting a distinct source mechanism. This is in agreement with its larger magnitude and with petrological and geophysical characteristics almost overlapping with those of paroxysmal eruptions at Stromboli.

One very critical aspect to consider is that, as noted, our Composite indicator is associated with a high (32%) false positive rate and hence low precision, which limits its applicability as an operational forecasting tool (at least in its present form). Identifying new processing tools and/or new

parameters that can more effectively monitor overpressure development in the explosion generation zone is critical to more robust forecasting of the small scale ($VEI < 1$) class of eruptions studied here. For example, use of artificial intelligence (machine learning) is now under test to identify any hidden pattern in our dataset. A second UV Camera system has been recently deployed at Stromboli to mitigate against incomplete coverage of the bulk plume by the currently operating UV Camera system (Lo Bue Trisciuzzi et al., 2024). New data processing tools are being explored to filter out from the Multi-GAS dataset observational periods with wind speed/direction unsuitable for plume fumigation at the measurement sites (Tamburello et al., 2023). Hopefully, this expanded dataset and analysis will help reducing the currently high false positive rate.

Ultimately, it is important to recall that Stromboli's major explosions rank at the lower end of the range of volcanic explosive eruptions and better eruption forecasting success rates are expected for larger-scale explosive events (Aiuppa et al., 2021; Acocella et al., 2024).

SUPPLEMENTARY MATERIAL

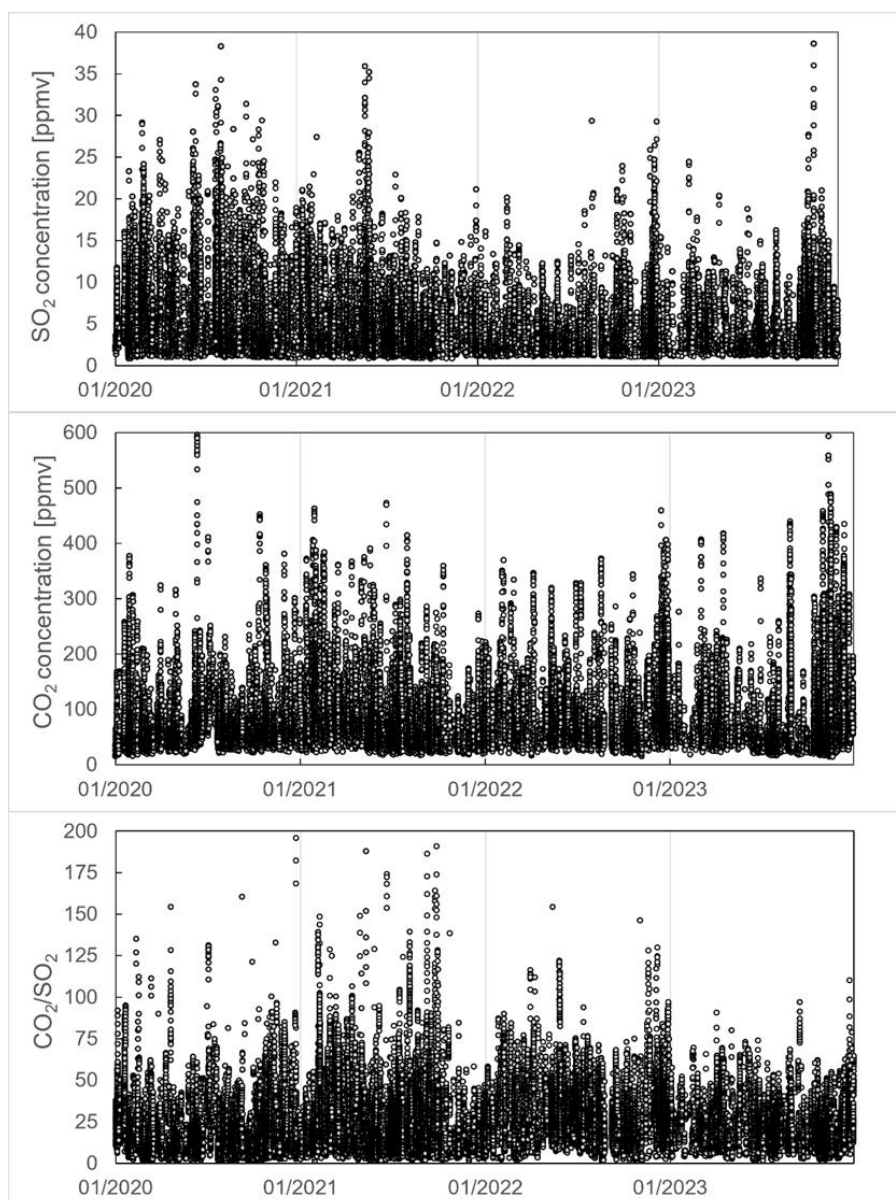


Figure S6.1 – Temporal record (2020–2023 period) of peak SO_2 and CO_2 concentrations, and CO_2/SO_2 ratios, data from Supplementary Table S6.2. Results are obtained by use of a R-based script that sequentially scans the dataset using a 60s long moving window, searching for intervals with (i) high temporal coherence (Pearson correlation coefficient ≥ 0.9) between CO_2 and SO_2 concentrations time series, and (ii) SO_2 concentrations at/above a ≥ 1 ppm threshold. These data are used to generate the plots of Figures 6.2 and 6.3.

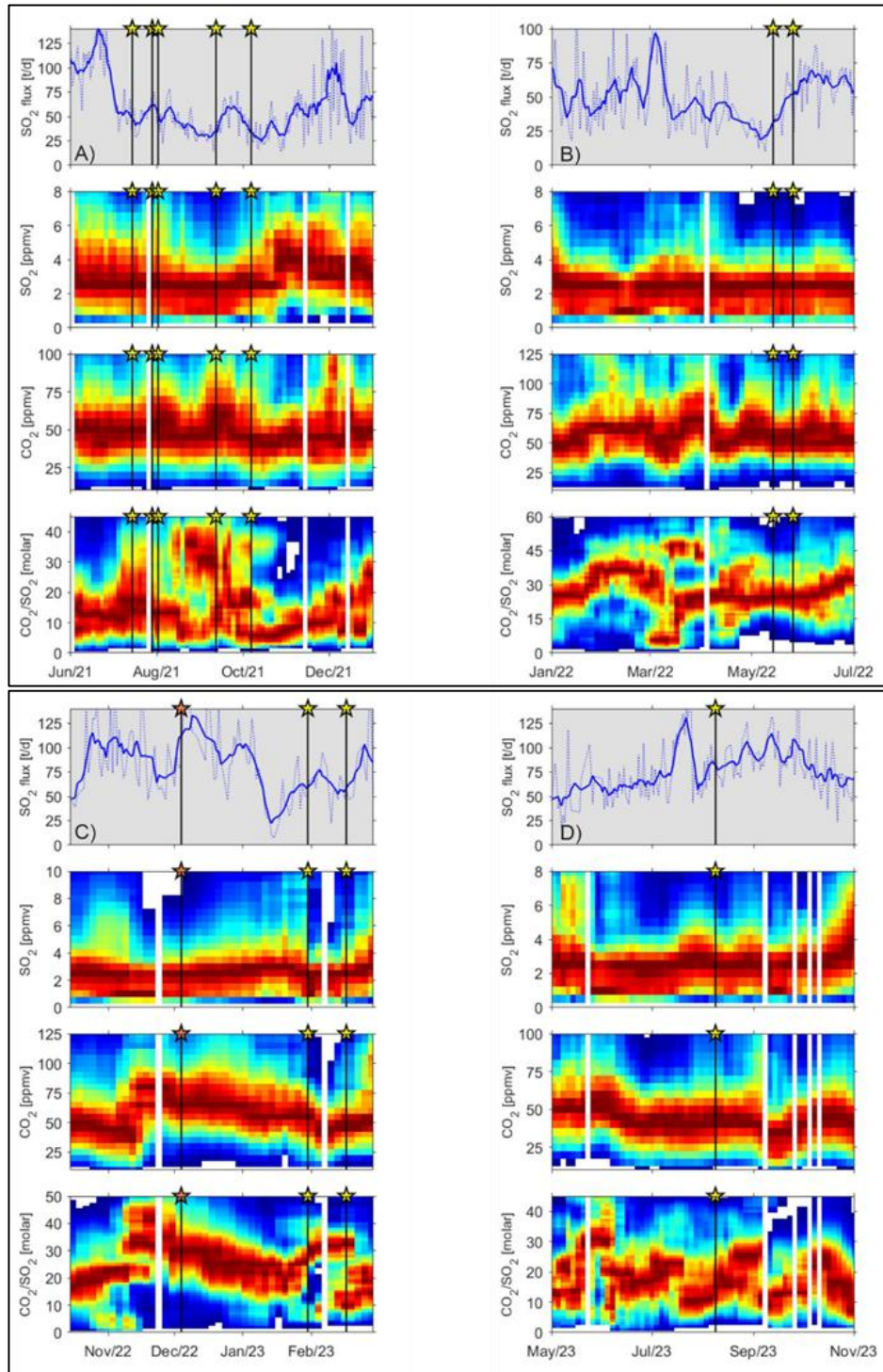


Figure S6.2 – ZOOMS of the compositional evolution of Stromboli's plume (Figure 6.2) in four selected temporal intervals. Yellow and orange stars identify major explosions and collapse/pyroclastic flow events, respectively. For each selected interval, we plot; (i) the daily averaged SO_2 fluxes measured by the Stromboli's permanent UV Camera, (ii) the temporal plots of (normalised) frequency distribution diagrams of SO_2 (c) and CO_2 (d) concentrations in the plume, and (iii) the corresponding CO_2/SO_2 (molar) ratios. Dark red tones convey information on the temporal fluctuations of the dominant concentration/ratio levels, whilst orange-to yellow-to green-to blue tones indicate increasingly less represented frequency bins.

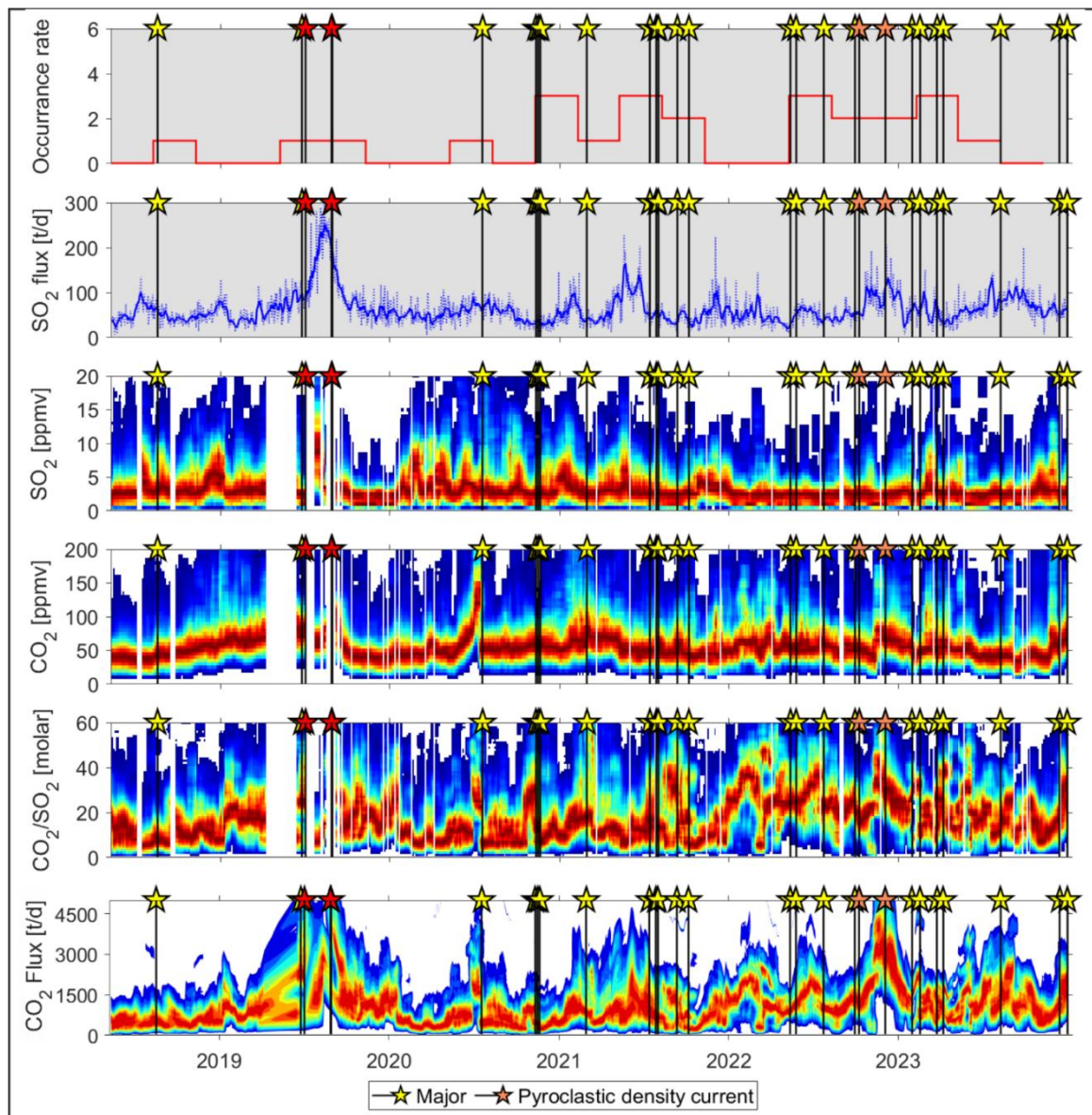


Figure S6.3 – Temporal record of gas observations at Stromboli. Same as Figure 6.2 in the main text, but with start time set at May 2018 to encompass the summer 2019 paroxysms (data from Aiuppa et al., 2021).

SUPPLEMENTARY TABLE:

Due to the large size, the Supplementary Tables S6.1 to S6.7 for this article can be found online at:

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CHAPTER 7

A DEEP CO₂ DRIVER FOR THE STROMBOLI'S SUMMER 2024 ERUPTIVE SEQUENCE

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Software, Methodology, Visualization, Resources. **Marco Pistolesi**: Writing – review & editing, Visualization, Software, Methodology, Formal analysis, Validation. **Maurizio Ripepe**: Visualization, Software, Methodology, Formal analysis, Data curation, Validation. **Marija Voloschina**: Software, Methodology, Visualization, Resources. **João Lages**: Writing – review & editing, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization, Supervision.

Abstract

A brief but intense eruptive sequence took place at Stromboli volcano on July 4-11, 2024. The sequence started with an event of landslide-generating partial failure of the summit North-East crater, causing opening of an eruptive fracture and hence rapid emptying of upper conduit magma into a developing lava flow field with the Sciara del Fuoco (SdF) scar. The sequence closed with a violent paroxysmal explosion during vanishing effusive activity. Here, we use compositional (CO_2/SO_2) and flux (SO_2 and CO_2) results for the volcanic gas crater plume, obtained with a permanent instrumental network, to propose a CO_2 -driven, deep trigger for the eruption. We show that the eruption was anticipated by ~60 days of above background ($>> 900$ metric and up to 3800 tons per day, t/d,) CO_2 fluxes, totalling an excess CO_2 mass of 2.18×10^5 tons at eruption onset. With geophysical data, this precursory CO_2 increases contributed to effective forecasting of the eruption. At the eruption onset, the plume CO_2/SO_2 molar ratio, that had remained high (25-40) for two months, abruptly dropped to <10 , consistent with degassing being dominated by CO_2 -poor shallow conduit magma upon its eruption as lava. After 8 days of uninterrupted effusion, the deeper magma that had sourced the precursory phase of heightened CO_2 degassing was ultimately erupted in the July 11 paroxysm. Comparison with previous (2007, 2014, 2019) Stromboli's eruptive sequences support the systematicity of excess CO_2 prior to paroxysm-generating eruptive sequences. Yet, the mid-term (timescales of days to weeks) prediction of the exact timing of a paroxysm, and of the exact sequence of events (if the paroxysm will be preceded by a precursory effusive phase as in 2024, or not), remains a challenge.

7.1 Introduction

Monitoring volcanic gases provides critical information to understand volcano behaviour, and to track – and potentially anticipate – the quiescence-to-eruption transition (Fischer and Chiodini, 2015; Edmonds, 2021; Edmonds et al., 2022, Aiuppa et al., 2025a, 2025c). At open-vent, persistently degassing mafic volcanoes, volatiles are primarily released from summit crater plumes (Oppenheimer et al., 2014). Measuring the composition and flux of such magmatic gas plumes is hence instrumental

to infer the role of volatiles in magmatic processes, such as (i) in sustaining active convective magma circulation during passive degassing (Shinohara, 2008), (ii) in causing the gas bubble coalescence events that drive mild explosive activity (Burton et al., 2007), and (iii) in triggering the more massive foam accumulation events (Jaupart and Vergnolle, 1989; Menand and Phillips, 2007) that are thought to be implicated in the triggering of larger-scale, explosive basaltic explosions (Allard et al., 2005; Aiuppa et al., 2021).

Over the past two decades, significant progress has been made in instrumental monitoring of volcanic gas plume composition and fluxes (Aiuppa et al., 2025a, 2025c; Campion, 2015). Among volcanic gas species, sulphur dioxide (SO₂) is the most widely measured, given its high abundance in volcanic emissions and low atmospheric background, which allows precise detection by spectroscopic remote sensing techniques (Kantzas and McGonigle, 2008; Oppenheimer et al., 2011; Kern, 2025). Continuous SO₂ flux monitoring has become a standard tool for surveillance, providing high-resolution datasets with direct relevance for eruption forecasting (Oppenheimer et al., 2014; Aiuppa et al., 2005). However, increasing evidence points to the crucial role of CO₂ in driving explosive basaltic volcanism (e.g., Aiuppa et al., 2010a; Edmonds and Wallace, 2017; Caricchi et al., 2018, 2024; Allison et al., 2021; La Spina et al., 2022; Barth et al., 2024). Basaltic magmas are often CO₂-rich, and owing to the very low solubility of CO₂ in silicate melts, exsolution typically initiates at mid- to deep-crustal levels (Carroll and Holloway, 1994; Edmonds, 2008; Edmonds and Wallace, 2017). The segregation and separate ascent of such exsolved CO₂-rich magmatic fluid phase (Edmonds et al., 2022) may lead to foam accumulation at the roof of magma storage zones and/or at structural/rheological discontinuities (Caricchi et al., 2024), causing pressure buildup, and ultimately eruption (Aiuppa et al., 2021). The CO₂-rich nature of basaltic magmas (e.g., Blundy et al., 2010; Barth et al., 2024) is also likely implicated in sustaining fast magma decompression rates during violent basaltic explosions (Allison et al., 2021; Barth et al., 2024). Numerous studies have highlighted increases in CO₂ fluxes and CO₂/SO₂ molar ratios prior to eruptions at basaltic volcanoes (Aiuppa et al., 2007, 2010a, 2017, 2021; Acocella et al., 2023). These variations are commonly interpreted as evidence of precursory segregation of CO₂-rich magmatic gas from soon-to-erupt mafic magma at depth, and hence represent increasingly robust precursory signals of magmatic unrest, and eruption (Aiuppa et al., 2025a, 2025c).

At Stromboli (Southern Italy), one of the most persistently active basaltic volcanoes, violent paroxysmal explosions punctuate the otherwise continuous Strombolian activity 1-2 times per decade on average (Rosi et al., 2013; Bevilacqua et al., 2020). These violent events (VEI $\leq$ 3) generate eruptive columns 3-8 km high, disperse coarse ballistic fallout across inhabited areas, and occasionally produce tsunamigenic pyroclastic density currents (Pioli et al., 2014; Giordano and De

Astis, 2021). Their sudden onset, short duration, and hazardous impacts underscore the importance of early identification of eruption precursors (Ripepe et al., 2021). Petrological evidence indicates that Stromboli's paroxysms originate from depths of 7-10 km within the plumbing system (Métrich et al., 2005, 2021). While different triggering mechanisms have been proposed (Métrich et al., 2005, 2021; Allard, 2010; Calvari et al., 2011; Aiuppa et al., 2021; Ripepe et al., 2017), there is broad consensus that paroxysms are initiated by the rapid ascent and injection of gas-rich, low-porphyricity (LP) magma from depth into the shallow conduit system (Métrich et al., 2021). Volatile saturation models reveal that such LP magma coexists with a CO₂-rich gas phase characterized by high CO₂/SO₂ ratios both during storage and ascent (Aiuppa et al., 2010a), whereas the shallow high-porphyricity (HP) magma feeding ordinary Strombolian activity is CO₂-poor and in equilibrium with a gas phase showing low CO₂/SO₂ ratios. Accordingly, models predict an escalation in CO₂ release during decompression-driven degassing of the LP magma prior to large explosions (Aiuppa et al., 2010a, 2021).

The most recent of such deviations from ordinary activity occurred at Stromboli in July 2024, when a brief but intense lava flow emission from the summer craters, started on July 4, preluded to a paroxysmal explosion on July 11. This eruptive sequence produced a substantial morphological transformation of Stromboli's crater terrace (Civico et al., 2024; Zuccarello et al., 2025), but fortunately caused no damage to humans and infrastructures. Here, we analyse the volcanic gas plume compositional (CO₂/SO₂ molar ratios) and flux (SO₂ and CO₂ fluxes) time series acquired, with a permanent network of one UV Camera and two Multi-GAS instruments (same as in Aiuppa et al., 2021), prior, during and after the eruptive sequence. Our novel results corroborate the role of CO₂ as a trigger of explosive basaltic eruptions, and the CO₂ flux an useful monitoring tool in volcanology.

7.2 Volcanic activity and hazards at Stromboli

Stromboli is the easternmost volcanic island of the Aeolian Archipelago, located in the southern Tyrrhenian Sea, Italy (Figure 7.1). Its persistent activity is dominated by frequent low-energy Strombolian explosions, typically occurring at a rate of approximately 7 to 10 events per hour (Ripepe et al., 2008; Calvari et al., 2010), sustained by continuous magma overturning in the shallow conduits (Giberti et al., 1992; Allard et al., 1994; Laiolo et al., 2022). These events eject a mixture of gas, spatter, scoriae and ash to modest heights, generally reaching a few tens of meters above the crater rim (Chouet et al., 1974; Gouhier and Donnadieu, 2010). Occasionally, more powerful major explosive episodes occur, resulting in the ejection of tephra beyond the crater terrace and affecting the upper part of the island (Pioli et al., 2014; Voloschina et al., 2023; Insinga et al., 2025). These explosions are associated with eruptive columns that may rise a few hundreds of meters above the

craters and are documented to occur, on average, a few times per year (Calvari et al., 2012, 2021b, 2022b; Bertagnini et al., 1999; Rosi et al., 2013; Giudicepietro et al., 2022; Bevilacqua et al., 2020). During periods of heightened activity, Stromboli may also produce paroxysmal explosions (Rosi et al., 2013) that generate eruptive columns exceeding 4 km in height, eject ballistic blocks up to 2 m in diameter, and causing substantial collapse of the summit crater areas. These events can be accompanied by the emplacement of pyroclastic density currents along the Sciara del Fuoco (SdF), which can enter the sea and travel up to 2 km from the shoreline, with the potential to trigger tsunamis (Rosi et al., 2006; Calvari et al., 2006; D'Auria et al., 2006; Pistolesi et al., 2020; Ripepe and Lacanna, 2024). Although less frequent than major explosions, paroxysms represent the most hazardous eruptive scenario for Stromboli (Rosi et al., 2013; Giudicepietro et al., 2020; Giordano and De Astis, 2021; Andronico et al., 2021a; Métrich et al., 2021).

In addition to explosive activity, Stromboli also experiences effusive eruptions. These may involve the slow extrusion of lava from the summit craters (the so-referred overflows) or the emplacement of more voluminous lava flows in the SdF depression. In some cases, lava is emitted from vents or fissures along the SdF, generating flows that reach the coast (Calvari et al., 2005, 2010, 2020a, 2022b; Di Traglia et al., 2018a; Nappi, 1976; Capaldi et al., 1978; Marsella et al., 2012; Casalbore et al., 2021). A genetic link between effusive eruptions and some paroxysmal explosions has been proposed (Aiuppa et al., 2010a, 2021; Calvari et al., 2011; Ripepe et al., 2015, 2017; Laiolo et al., 2022), in which lava effusion may act as a trigger for paroxysms by promoting decompression of the shallow plumbing system, for instance through magma-level drops in the conduit. This mechanism is interpreted as a top-down process, in which the onset of surface effusion destabilizes the shallow system promoting gas-magma ascent from depth, culminating in a paroxysm (Aiuppa et al., 2021). However, not all paroxysms are preceded by effusion. Some occur without clear precursory lava outflow and are instead likely associated with a bottom-up mechanism (Métrich et al., 2021), in which the rapid ascent and accumulation of volatile-rich magma from depth drives the explosive climax (Métrich et al., 2005; Pichavant et al., 2009). More recently, Caricchi et al. (2024) proposed that the collapse of gas-rich foam layers at the base of the magma column may also act as a trigger for paroxysmal explosions.

The structural evolution of the volcano is also shaped by gravity-driven phenomena. Landslides frequently occur along the SdF, often in association with eruptive events (Falsaperla et al., 2008; Rosi et al., 2019; Pistolesi et al., 2020; Turchi et al., 2020), occasionally triggering small tsunamis (Maramai et al., 2005; Tinti et al., 2006; Fornaciai et al., 2019). The progressive accumulation of material and the rise of magma levels within the conduit system are contributing factors to the

instability of the summit cone, particularly in the presence of lateral magma intrusions (Calvari et al., 2016, 2020a, 2021b, 2022b; Di Traglia et al., 2014, 2018b; Tioukov et al., 2022).

7.2.1 The Stromboli's 4-11 July 2024 eruption

The chronology of the July 2024 eruptive sequence is reconstructed based on monitoring reports dispatched by Istituto Nazionale di Geofisica e Vulcanologia, INGV (<https://cme.ingv.it/bollettini-e-comunicati/bollettini-ingv-stromboli>) and Laboratorio Geofisica Sperimentale, LGS (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1>). The eruptive sequence, started on July 4, was preceded, since mid-April 2024, by a general intensification of the regular (Strombolian) activity, as indicated by a progressive escalation in seismic tremor and infrasonic amplitude, by more frequent and shallower very long seismicity (VLP), and by a concentration of the degassing/eruptive activity at the North-east crater (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1>). These geophysical signals were interpreted to reflect an increased supply of magmatic volatiles (as discussed in this study) into the Stromboli's shallow conduit feeding system, causing a general escalation in volcanic activity. Based on these monitoring results, LGS raised to orange (high) its volcanic activity level on April 30, 2024 crater (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1/2270-bollettino-unifi-lgs-stromboli-20240502/file>), and the orange (high) Alert Level was declared by the Italian Civil Protection (Dipartimento di Protezione Civile, DPC) on June 24. Followed a further escalation in geophysical and geochemical signals, LGS raised its volcanic activity level to red (very high) on June 28 (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1/2368-bollettino-unifi-lgs-stromboli-20240704/file>).

The eruption commenced on July 4 between ~ 12:11 and ~1:00 pm (time in UTC), after a further increase in volcanic activity caused gravitational failure of the North-East crater outer flank, triggering hot avalanches in the SdF (Civico et al., 2024; Zuccarello et al., 2025). A major explosion took place during the initial phase of the eruption (at 12:11 pm), followed by the partial collapse of the crater at ~4.40 pm and consequently intensification of landslide activity in the SdF, and the opening (at ~5 pm) of an eruptive fracture at circa 700 m asl (above sea level) at the base of the North-East cone. Concurrently, explosive activity at the summit craters ceased. The red (very high) Alert Level was declared by DPC (<https://emergenze.protezionecivile.gov.it/it/vulcaniche/eruzione-stromboli-2024/>). Initiation of the lava effusion was accompanied by an intense phase of deflation phase (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1>; see also Zuccarello et al., 2025). Lava effusion continued, with variable but overall milder intensity, until July 11, forming a lava delta along the shoreline. As previously observed at Stromboli (e.g., Aiuppa et al., 2010a, 2021), a paroxysmal eruption occurred on July 11 at 12:08 (UTC) during this waning effusive activity phase. A large

inflation was detected by LGS tiltmeters in the minutes before the paroxysm, as systematically observed at Stromboli (Ripepe et al., 2021). Following cessation of effusive activity on the same July 11, Strombolian activity resumed at the summit craters on 17 July. This post-eruptive phase was punctuated by four major explosions on 26 July, 25 August, 6 October, and 2 November 2024.

The July 2024 eruption represents the most substantial morphological transformation of Stromboli's crater terrace in recent decades. The most considerable topographic variation was recorded in the northern sector, where the elevation of the lower northern crater vent decreased by more than 100 meters (Civico et al., 2024).

7.3 Methods

The volcanic gas time series reported in this study (Figures 7.2, 7.3) have been acquired from records of the geochemical instrumental network operated by Università di Palermo (UniPa) under the umbrella of the LGS university partnership (also including the universities of Firenze, Pisa and Torino), and funded (for the 2022-2024 period) within the context of collaboration agreement with INGV. The instrumental network is the same as previously described in Aiuppa et al. (2021).

The SO₂ flux data used in this study are obtained using records from a permanent UV Camera system installed in May 2014 at the Roccette site (Figure 7.1), located on the northeastern upper flank of Stromboli at approximately 750 meters above sea level. Measurement principles of the UV Camera station are detailed in Mori and Burton (2006), Kantzas et al. (2010) and Kern et al. (2010a, 2010b). Situated at only ~500 meters northeast of the active vents, this vantage point provides an optimal view of the northeastern portion of the crater terrace, while potentially missing a substantial (~30%; Delle Donne et al., 2022) fraction of the gas emitted by the partially hidden South-West and Central craters (Lo Bue Trisciuzzi et al., 2024). The UV Camera system is designed to output high-rate (0.5 Hz) SO₂ flux records in continuous mode during daylight for ~6 h per day. It is equipped with two UV-sensitive cameras, each fitted with a distinct bandpass optical filter, which operate respectively in the range 300-320 nm (where there is the maximum absorption of sunlight by SO₂) and 320-340 nm (where the absorption is significantly reduced, see also Vandaele et al., 1994). A detailed list of components and their specifications is provided in Delle Donne et al. (2017, 2019, 2022). Pairs of simultaneously acquired images are post-processed by combining them into a single absorbance image using the Lambert-Beer law. This process enables the quantification of the relative absorption of scattered UV sunlight by any volcanic SO₂ present within the cameras' field of view. Column densities are then derived over the entire images from image calibration (McGonigle, 2007; Delle Donne et al. 2017). Plume speed is calculated by applying the Lucas and Kanade optical flow algorithm (Lucas and Kanade, 1981) to sets of consecutive images. These plume speed measurements

are conducted on a restricted image portion, capturing rectangular subregions located just above the crater rim, where the SO_2 absorption signal is maximized while atmospheric effects, such as air entrainment in the plume and complexities in the local wind field, are minimised. By combining the plume speed and SO_2 column density, a SO_2 flux density profile is derived along an ideal transect, that connects the centres of the rectangular subregions (Delle Donne et al. 2017). Subsequently, a time series of SO_2 flux is computed at a temporal resolution of 0.5 Hz by integrating the flux density along the full length of the profile (Lo Bue Trisciuzzi et al., 2024).

Daily SO_2 fluxes are calculated by averaging flux values corresponding to optimal observational conditions. These conditions are identified using a visibility index, defined as the absolute ratio between the mean pixel intensities of the portions of the camera's Field Of View (FOV) directed toward the sky and the ground, respectively (Delle Donne et al., 2019). Depending on environmental and meteorological conditions, a variable number of measurements (with a visibility index below a predefined threshold) are excluded from the analysis. The entire data processing workflow is implemented in MATLAB, which produces near-real-time time series of SO_2 fluxes (Delle Donne et al., 2017, 2019).

Given the short distance of the UV camera from the target plume (~ 500 m), and the small error ($\pm 5\%$) associated with plume transport speed measurements derived from tracking plume motion with an optical flow algorithm (Delle Donne et al., 2019), we estimate that SO_2 flux uncertainty at Stromboli is primarily controlled by radiative transfer effects (typically $\sim \pm 30\%$, see Table 1 in Kern et al., 2010b and Lo Bue Trisciuzzi et al., 2024) and by the incomplete plume coverage ($\sim \pm 30\%$, Delle Donne et al., 2019). The sum of these individual errors, as square root of the sum of the squares of the individual errors, contribute to a total uncertainty of $\sim \pm 43\%$.

Real-time measurements of the CO_2/SO_2 ratio are retrieved from in situ measurements of in-plume CO_2 and SO_2 concentrations using two permanent and fully-automated Multi-GAS devices (Aiuppa et al., 2005, 2021, 2025b; Shinohara, 2005), deployed in the Pizzo Sopra la Fossa area (Pizzo in Figure 7.1) since mid-2018 and late-2020 respectively. The instruments, assembled at the Dipartimento di Scienze della Terra e del Mare (DiSTeM), Università di Palermo (UniPa), are configured to acquire in-plume gas concentrations at 1 Hz using a set of pre-calibrated, gas-specific sensors, mounted in series and exposed to the target gas plume (see Aiuppa et al., 2021). In particular, each unit employs a Campbell CR6 data logger to command operations of (and retrieve signals from) an infrared spectrometer (Gascard NG, Edinburgh Sensors; calibration range: 0-3000 ppmv, accuracy: $\pm 1.5\%$, resolution: 0.1 ppmv) for CO_2 detection and an electrochemical sensor (SO_2 T3ST/F-TD2G-1A, City Technology; calibration range: 0-200 ppmv, accuracy: $\pm 2\%$, resolution: 0.1 ppmv) for SO_2 detection. Measurements are automatically triggered at fixed intervals (typically every

4 h) by the datalogger, with acquisition durations ranging from ~300 to 1800 s, depending on SO₂ levels, in order to optimize power consumption. Data acquisition is interrupted when SO₂ concentrations fall below a pre-defined threshold, to avoid collecting measurements with poor volcanic signal. Otherwise, the instrument continues operating during four daily cycles (each 30 minutes long), during which data are acquired and stored locally on the data logger.

The CO₂ and SO₂ concentration time series are processed in near-real time by a R-based script that sequentially scans the dataset using a 60 s-long moving window, searching for intervals with (i) high temporal coherence (Pearson correlation coefficient ≥ 0.9) between CO₂ and SO₂ concentrations time series, and (ii) SO₂ concentrations at/above a 1 ppm threshold (same as Aiuppa et al., 2021, 2025b). For each temporal window in which both conditions above are met, the algorithm records the maximum SO₂ concentration, the range (maximum - minimum) Δ SO₂ and Δ CO₂ concentration levels, and calculates the corresponding CO₂/SO₂ molar ratio. Use of the Δ CO₂ and Δ SO₂ concentration helps to resolve the volcanic concentration signal above the atmospheric background. The CO₂/SO₂ ratio is obtained from the slope of the best-fit linear regression function calculated from the Δ SO₂ and Δ CO₂ concentration time series within the processing window (Aiuppa et al., 2021, 2025b). The overall uncertainty in SO₂/CO₂ measurements from Multi-GAS instruments has been reported to be $\leq 20\%$ (Aiuppa et al., 2009) depending on sensor performance and environmental conditions, while laboratory experiments indicate errors of $\leq 15\%$ (Pering et al., 2014).

By combining the daily CO₂/SO₂ ratio distributions with the daily-averaged SO₂ flux, a volcanic CO₂ flux can be retrieved. The associated uncertainty, resulting from the propagation of errors in both CO₂/SO₂ ratios and SO₂ fluxes, is estimated at $\leq 44\%$.

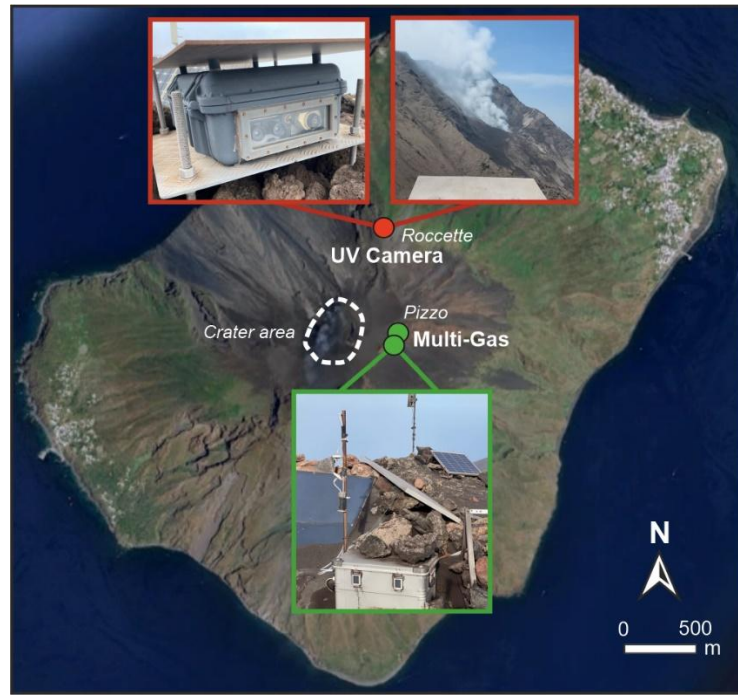


Figure 7.1 – The UniPa monitoring network at Stromboli.

7.4 Results

Figure 7.2 illustrates our volcanic gas records obtained during a 1-year period (January 1 - December 31, 2024) encompassing the July 2024 Stromboli's eruption.

The SO₂ flux time series, derived from processing of the UV Camera images, exhibits a well-behaved temporal behaviour (Figure 7.2a). The SO₂ fluxes are generally at levels <50 t/d during January to April 2024. Since late April, the SO₂ flux increases by a factor ~2, reaching levels of ~100 t/d. Onset of the eruptive sequence on July 4 marks a step change in the SO₂ degassing regime, with the SO₂ flux sharply increasing to values > 200 t/d, peaking at ~555 t/d on 08 July. The SO₂ flux average for the 8 days of the effusive unrest (4-11 July) is 281 ± 181 t/d. In the post-eruptive phase, the SO₂ flux oscillates at levels of 100-200 t/d during a 4-month period (mid-July to mid-October) during which 4 major explosions take place. These levels are well above the decadal (2014-2023) SO₂ flux time-average of 67 ± 49 t/d (Lo Bue Trisciuzzi et al., 2024). Then after, the SO₂ fluxes gently declines to values comparable to those observed prior to the onset, reaching values <50 t/d by December 2024.

Figure 7.2b shows a chronogram of the normalized frequency distribution diagrams of the CO₂/SO₂ (molar) ratio, calculated on a 3-day interval (same graphic representation as in Aiuppa et al., 2021, 2025b). A centered smoothing function is applied to sets of 3 adjacent frequency distribution plots to reduce high-frequency variability related to wind field or and other nonvolcanic phenomena, further emphasizing longer term trends. In the resulting plots, the warmer tones (dark

red) indicate the most represented values (the mode of the distribution), while cooler tones (yellow to blue) reflect progressively less frequent concentration/ratio bins, thereby capturing both central tendencies and data spread. The CO₂/SO₂ ratio time series (Figure 7.2b) shows the persistence of high (>20; Aiuppa et al., 2021) values during most part of the measurement interval. After a 2-month long (January to February 2024) period with values dominantly <20, from March 2024 the CO₂/SO₂ ratio increases to values of ~25 to 40, that persist until the onset of the 4 July eruption. Notably, a bimodal distribution of CO₂/SO₂ (molar) ratios, with the coexistence of two populations with modal values of ~10 and ~30, is observed in the weeks before eruption onset. A similar bimodal distribution was observed prior to the 2019 eruption (Aiuppa et al., 2021 and Figure 7.3b). Onset of sustained lava effusion on July 4 marks a sharp decrease of the ratio to values <10 that persist until the paroxysmal explosion of 11 July. The syn-eruptive CO₂/SO₂ ratio average is 4±0. After the abrupt termination of the eruptive sequence with the July 11 paroxysm, the high (>20) CO₂/SO₂ ratio signature is rapidly recovered, and maintained until the end of the analysed period.

The CO₂ flux time series, obtained by combining the CO₂/SO₂ ratio chronogram (Figure 7.2b) with daily-averaged SO₂ fluxes (of Figure 7.2a), exhibits a prominent peak prior to the eruption onset (Figure 7.2c). The CO₂ flux is initially at ~1000-1500 t/d in January-March; then after, it progressively increases to ~2000 t/d in April 2024, to finally reach a sustained level of ~3000-4000 t/d in May, and until the onset of the eruption (Figure 7.2c). For context, the 2018-2023 average is 981±611 t/d when results for the 2019 eruptive sequence (Aiuppa et al., 2021) are omitted to approximate background levels during ordinary activity (Figure 7.3). The eruption onset is marked by a sharp drop in CO₂ fluxes that lasts for the entire duration of the effusive phase. This reflects the low CO₂/SO₂ ratio compositions described above (Figure 7.2b). In the post-eruptive phase, the CO₂ flux recovers to the pre-eruptive values within ~2 weeks, anticipating the first (of a cluster of 4) major explosion on July 26 2024. This elevated CO₂ flux phase continues throughout the following month, with fluxes peaking at ~6100 t/d before the second major explosion on 25 August 2024. CO₂ emissions then remain elevated, in the range of 3000-3500 t/d, until late September, when they decrease to ~2500 t/d in October to early November, during which two further major explosions occurred (6 October and 2 November 2024). Following these last explosive events, the progressive CO₂ flux vanishing trend continues, finally reaching values of < 2000 t/d.

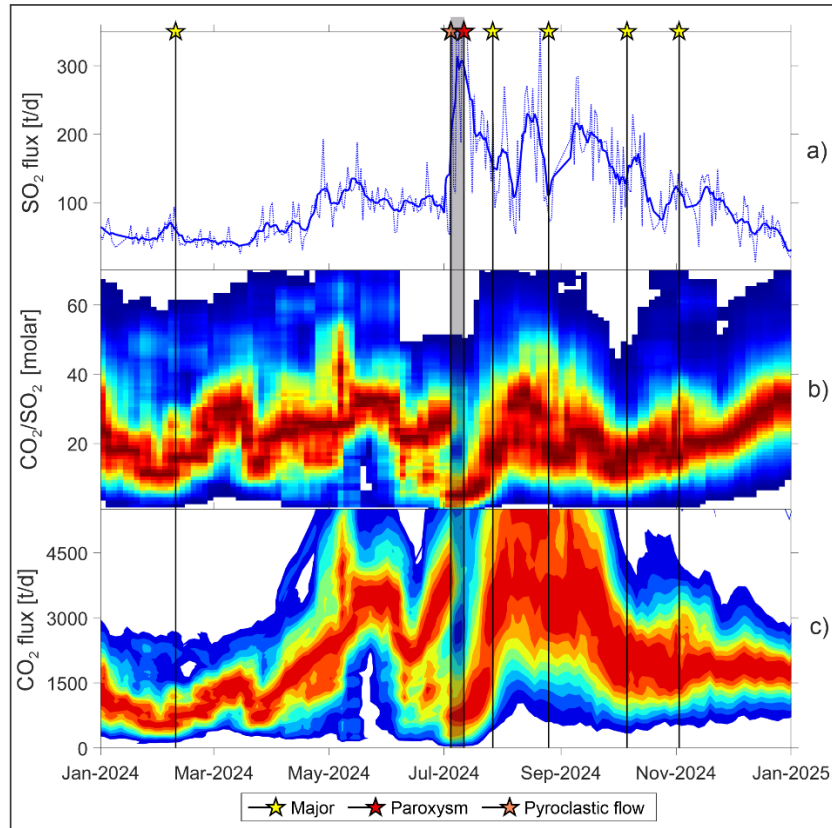


Figure 7.2 – CO_2/SO_2 ratio and gas fluxes in a 12-month period encompassing the July 2024 eruption (grey area). a) Daily averaged SO_2 fluxes from UV Camera; b) Normalized frequency distribution diagrams of the in-plume CO_2/SO_2 ratios on a 3-day interval, obtained processing data from Multi-Gas. To fill gaps in the time series resulting from unfavourable wind directions or instrumental issues, data has been linearly interpolated (see also Aiuppa et al., 2021); c) Daily CO_2 flux matrix retrieved by combining SO_2 fluxes and CO_2/SO_2 frequency distribution. In b) and c) the modal CO_2/SO_2 and CO_2 flux values, corresponding to the dominant bins of the respective frequency distributions, are indicated by dark red tones. The key volcanic events are indicated by stars (see legend).

To put our results in context, the 2024 time series is compared with previous results (Aiuppa et al., 2021, 2025b), yielding the extended (2018-2024) time series of Figure 7.3.

Figure 7.3a demonstrates that similar syn-eruptive excursions to high values (of 250-300 t/d) are common to the 2019 and 2024 eruptions. A similar syn-effusive SO_2 flux increase, above background activity levels (e.g., Allard et al., 2008), has also been observed prior to the 2003 (Ripepe et al., 2005) and 2007 (Aiuppa et al., 2010a) eruptions.

The CO_2/SO_2 ratio time series (Figure 7.3b) is characterised by large variability. Notably, the CO_2/SO_2 ratio oscillates at higher values of 22 ± 8 during 2022-2024, relative to the 2018-2021 period (13 ± 6). The 2022-2024 period is consistently associated with a higher recurrence rate of major explosions (14 events; ~ 4.7 events/year on average; for context, the recurrence rate is 3.25 events/year in 2018-2021) (Figure 7.3). The causal link between high CO_2/SO_2 ratio phases and major explosions is discussed elsewhere (Aiuppa et al., 2025b).

Figure 7.3c demonstrates that the modal (dark red colour tones) CO₂ flux values of > 2000 t/d are exclusively observed in the weeks/months before large-scale (non-ordinary) explosive events. In particular, large, above background (981±611 t/d) CO₂ flux emissions are observed, in addition to the 2024 eruptive sequence (Figure 7.2c), prior to the 2019 paroxysms on July 3 and August 28 (Aiuppa et al. 2021), and before a major failure events of the North-east summit crater rim on 4 December 2022, which produced the fast spreading of hot pyroclastic density currents inside the SdF (Di Traglia et al., 2024; Casalbore et al., 2025). Notably, the 2024 paroxysmal eruption is characterized by the highest CO₂ fluxes on record, with values twice as much higher than those measured prior and during the 2019 eruptive sequence.

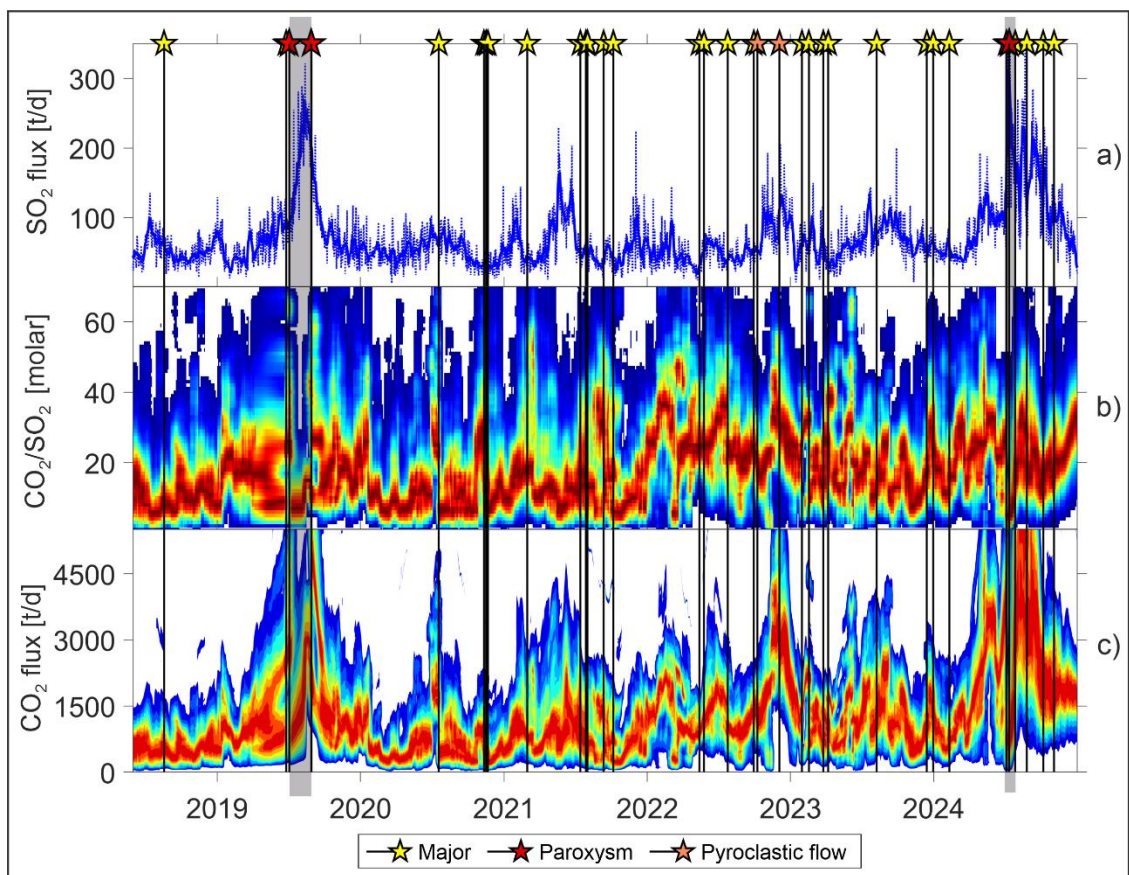


Figure 7.3 – CO₂/SO₂ ratio and gas fluxes from 2018 to 2024. The 2018 to 2023 results are taken from Aiuppa et al., (2021, 2025b). The 2018 and 2024 eruptions are marked as grey areas.

7.5 Discussions

7.5.1 A “forecasted”, CO₂-driven eruption

Our results for the 2024 Stromboli’s eruption corroborate the utility of CO₂ fluxes as a robust volcano monitoring tool. We show that the eruption was preceded by a large, well-above background precursory escalation in CO₂ emissions (Figures 7.2, 7.3). To better illustrate this pre-eruptive CO₂

increase, we calculate (as in previous work; Aiuppa et al., 2021) the cumulative excess CO₂ trend of Figure 7.4b, in which excess CO₂ is quantified by subtracting the pre-unrest (e.g., January-mid April 2024) CO₂ flux average of 900 ± 283 t/d. Results demonstrate a clear break-in-the-slope in the cumulative excess CO₂ curve at ~60 days before eruption onset. This corresponds to the time at which the shallow Stromboli's conduit feeding system was first supplied with CO₂-rich, deeply sourced bubbles (Aiuppa et al., 2010a, 2021), at an above-than-normal rate. We suggest that this anomalous deep gas supply caused the shallow magma column to become more effervescent and buoyant, as indicated by the overall acceleration in regular (Strombolian) activity tracked by volcanological observations (<https://cme.ingv.it/bollettini-e-comunicati/bollettini-ingv-stromboli>), and by escalating volcano seismicity and infrasound (<https://cme.ingv.it/bollettini-e-comunicati/categoria-1/2270-bollettino-unifi-lgs-stromboli-20240502/file>). This heightened Strombolian activity ultimately reached a critical condition at which the North-East crater had become gravitationally unstable, up to its failure at the onset of the eruption on July 4. At that point, the cumulative excess CO₂ flux had reached 2.18×10^5 tons (Figure 7.4b).

The important conclusion we can draw here is that, while the 2024 eruption was ultimately caused by the rapid (days) and shallow (crater terrace and upper SdF flank) build-up of gravitational instability (Di Traglia et al., 2024), the ultimate driver of the eruption was acting over longer timescales (2 months) and deeper in the system (e.g., caused by fluxing of deeply sourced CO₂-rich bubbles). In terms of eruption forecast potential, the 2024 eruption example shows that we can track and anticipate the quiescence-to-eruptive transitions months to days in advance (the LGS Volcanic Activity Level was raised to orange and then red on April 30 and June 28, respectively) when CO₂ and geophysical monitoring tools are used in concert. In this context, the bimodal nature of both CO₂/SO₂ (Figure 7.2b) and CO₂ fluxes (Figure 7.2c) in the weeks before eruption onset on July 4 is intriguing, and considering it was also observed before the 2019 eruption (Aiuppa et al., 2021 and Figure 7.3), it may find usage (if confirmed by further observations) as an indicator of imminent eruption. We interpret this bimodality as reflecting a transient condition in which deeply degassing volatile-rich magma (initially sustaining the high CO₂/SO₂ ratio, high CO₂ flux phase) finally finds its final ascent route toward the surface (shallow-pressure, decompressional magma degassing is associated with decreasing CO₂/SO₂ ratios, as models suggest; Aiuppa et al., 2007, 2010a, 2021, 2025a, 2025c; Edmonds, 2008; Edmonds et al., 2022)

The SO₂ flux also increased prior to eruption onset in 2024 (Figure 7.2a), but the cumulative excess SO₂ trend is flatter than for CO₂ (compare Figures 7.4a, 7.4b). CO₂ has limited solubility in mafic silicate melts (Wallace et al., 2015), which causes its early exsolution from mafic magmas at mantle to deep crustal conditions (Edmonds and Wallace, 2017). This justifies the precursory CO₂

flux escalation at volcanoes in which eruption is deeply initiated (Aiuppa et al., 2006, 2008, 2010a, 2021; Poland et al., 2012). In contrast, sulphur is partitioned into the exsolved magmatic gas phase as magmas are decompressed/stored at shallower crustal conditions (Oppenheimer et al., 2011; Spilliaert et al. 2006; Edmonds and Wallace, 2017; Métrich et al., 2021; Ding et al., 2023), which causes pre-eruptive SO₂ flux signals to be often more subtle relative to CO₂ (e.g., Aiuppa et al., 2008). In contrast, efficient shallow magma transport during eruption causes the large syn-eruptive SO₂ flux increases, seen at Stromboli (e.g., Laiolo et al., 2022) and at volcanoes globally (Carn et al., 2016, 2017). Our results for the July 2024 eruptive sequence at Stromboli reflect these mechanisms, as SO₂ and CO₂ fluxes exhibited temporally shifted trends, in which the SO₂ fluxes peaked during the effusive phase, whereas the CO₂ fluxes well before (and after) (Figure 7.2a,c). Rapid, shallow magma emplacement and eruption explain well the dominantly low CO₂/SO₂ ratios observed during the 2024 eruptive phase (Figure 7.2b).

7.5.2 Comparison with other eruptions

At Stromboli, a close temporal association between phases of excess CO₂ degassing and paroxysmal activity has been observed in occasion of *all* the instrumentally monitored eruptions of the last two decades, such as in 2007 (Aiuppa et al., 2010a), 2019 (Aiuppa et al., 2021) and 2024 (this study) (no CO₂ sensing device was in place at the time of the 2003 paroxysm, unfortunately). This systematicity is promising in terms of eruption forecast potential. However, predicting the exact timing of a paroxysm remains a challenge, if not in the very last minutes beforehand (Ripepe et al., 2021). During the last two decades, paroxysmal eruptions have generally occurred in association with effusive activity in the SdF (e.g., Barberi et al., 2009; Rosi et al., 2013). However, while some events have occurred during (2003, 2007) or in the vanishing phases (28 August 2019, 2024) of an effusive phase, the July 3 2019 paroxysm occurred right at its opening stage (Figure 7.4). This poses the question of why the 2019 and 2024 were both anticipated by excess CO₂ degassing, but had such inconsistent (shifted relative to eruption onset) timings for the paroxysms (Figure 7.4). Also, while syn-effusive paroxysms have been interpreted to be caused by a top-to-bottom process of slow decompression of the deep plumbing system, induced by lava drainage (Aiuppa et al., 2010a, 2021; Calvari et al., 2011; Ripepe et al., 2015, 2017; Laiolo et al., 2022), this mechanism is obviously not applicable to the July 3 2019 event, which rather requires a bottom-up process (Métrich et al., 2021). Clearly, in spite of the many commonalities (Métrich et al., 2021), each of the Stromboli eruptions has its own distinct behaviour, dynamics, and timing of the events, likely reflecting different modes and rates of magma pre-eruptive storage and ascent.

We compare our 2024 gas record with those for the 2007 (Aiuppa et al., 2010a) and 2019 (Aiuppa et al., 2021) eruptive sequences, as well as for the 2014 effusive eruption (in which no paroxysm occurred; Liuzzo et al., 2015), in the attempt to identify any recurrent degassing pattern, or rather the existence of eruption-specific behaviours (Figure 7.4). It is important to keep in mind, while performing this comparison, that the 2007-2014 results are based on the SO₂ fluxes obtained from the UV scanning spectrometers of the FLAME Network (Burton et al., 2009; Salerno et al., 2009a), which have been shown to exhibit a systematic positive offset of ~ 130 t/d relative to the UV Camera (Lo Bue Trisciuzzi et al., 2024). Also, a different CO₂/SO₂ ratio processing routine was used for the pre-2018 Multi-GAS dataset (for example, a different SO₂ threshold was in use), making a direct comparison of the CO₂ flux datasets (derived from SO₂ flux and CO₂/SO₂ from them) inappropriate. Hence, the comparison can only be attempted for the excess SO₂ (Figure 7.4a) and CO₂ (Figure 7.4b) masses, in which the eruption-specific pre-unrest averages are subtracted (hence mitigating against the differences in the datasets). More specifically, we calculate the SO₂ excess by subtracting a background SO₂ flux corresponding to pre-unrest averages of 143 ± 49 t/d (2007), 145 ± 72 t/d (2014), 52 ± 21 t/d (2019) and 50 ± 14 t/d (2024). Likewise for calculation of the CO₂ excess, we subtract the averaged CO₂ fluxes (during the same intervals above) of respectively 400 ± 230 t/d, 645 ± 419 t/d, 680 ± 246 t/d and 900 ± 283 t/d.

The excess SO₂ curves (Figure 7.4a) demonstrate similarly mild accelerations in SO₂ degassing regime in the weeks to months before eruption onset. We find no obvious difference in pre-eruptive excess SO₂ mass for eruptions with (2007, 2019 and 2024) or without (2014) a paroxysm. In all four examples, the most remarkable feature is the abrupt, steep change-in-the-slope of the cumulative excess SO₂ curve at the onset of the effusive phase (identified by the dashed portions of curves in Figure 7.4). Manifestly, accelerating (excess) SO₂ is primarily syn-effusive in nature, and is the largest for the 2007 event (Figure 7.4a).

The cumulative excess CO₂ flux curves (Figure 7.4b) exhibit more diverse behaviours. In 2007, the cumulative CO₂ curve exhibits a marked step change at ~ 80 days before effusion onset, followed by a slower and more gradual rise that persists until then. The 2019 eruption is preceded by a more continuous (constant rate) increase in excess CO₂, starting ~ 180 days prior to onset (Aiuppa et al., 2021). Finally, the 2024 event stands out for exhibiting the largest excess CO₂, which starts escalating ~ 60 days before and reaches $2.18 \cdot 10^5$ tons at onset. By contrast, the purely effusive 2014 eruption exhibit very minor ($\sim 0.35 \cdot 10^5$ tons) pre-eruptive excess CO₂ mass.

Overall, the conclusion we can take from Figure 7.4 are:

- (i) An excess CO₂, at levels of $0.78\text{--}2.18 \cdot 10^5$ tons, is observed prior to all 4 paroxysmal explosions in our record, and hence appears as a necessary pre-requisite for the event to occur. The largest excess is associated with the 2024 event;
- (ii) The quiescent-to-effusive transition marks a discontinuity (a change-in-the-slope) in the excess CO₂ mass trend, implicating that the discharge of shallow conduit magma during an effusion phase causes a general perturbation (depressurization) of the Stromboli's magma feeding system (Ripepe et al., 2005, 2015, 2017; Laiolo et al., 2022) that extends deep in the magma storage zone(s) that act(s) as source of CO₂ (Aiuppa et al., 2021).
- (iii) This perturbation is delayed in time the 2024 case, in which the excess CO₂ mass increases *after* rather than *at onset* and *during* the effusive unrest (Figure 7.4b). The occurrence of four major explosions (Figure 7.4) in this phase indicates that the post-eruptive periods of heightened CO₂ degassing are favourable conditions for major explosions to occur. Eventually, such “perturbed” deep plumbing system may continue to supply CO₂-rich deep gas for months, hence favouring the accumulation events of gas foams at rheological/structural discontinuities (a proposed mechanism for major explosions; Aiuppa et al., 2025b);
- (iv) An excess CO₂ mass, in the order of $1\text{--}2 \cdot 10^5$ tons, is also reached in the terminal stages (e.g., from ~40 days after effusion onset) of the 2014 eruption, and yet no paroxysm is observed. Clearly, there is not an exact threshold at which a paroxysm will necessarily occur. Rather, the 2014 example shows that it is rate at which this threshold is achieved that may control if a paroxysm is to happen or not. If the deep Stromboli's plumbing is decompressed at slow enough rate, as in eruption characterised by low lava effusion rate as in 2014 (Ripepe et al., 2015, 2017), then the excess CO₂ mass can be delivered quiescently by deeply stored magma, without resulting in a paroxysm;
- (v) The 2007 and 2019 eruptive sequences started at similar cumulative excess CO₂ masses, well below those seen in 2024. And yet, the 2019 eruptive sequence started with a paroxysm without a precursory effusive phase.

This latter point is the most problematic for volcanic hazard management. If an excess CO₂ phase starts, how can we predict the expected sequence of events? will the eruptive sequence be opened by a paroxysm (as in 2019), or by crater failure and effusion (as in 2024)? There is no easy answer to these critical questions. We yet tentatively note that the 2019 event is associated with the smallest excess SO₂ mass (see Figure 7.4a) in the dataset. This may reflect a reduced transport of magma at shallow levels, and hence that this specific eruption was initiated deeper in the system than the others. In such conditions, the precursory lava effusion phase is perhaps less likely to occur, and

the system is more prone for an early paroxysmal phase. The characteristics of the pyroclastics erupted during the July 11 2024 are now being analysed. If our interpretation is correct, a larger fraction of shallow-derived magma fragments is to be expected in 2024, relative to the LP magma fragment-rich July 3 2019 event (Métrich al, 2021).

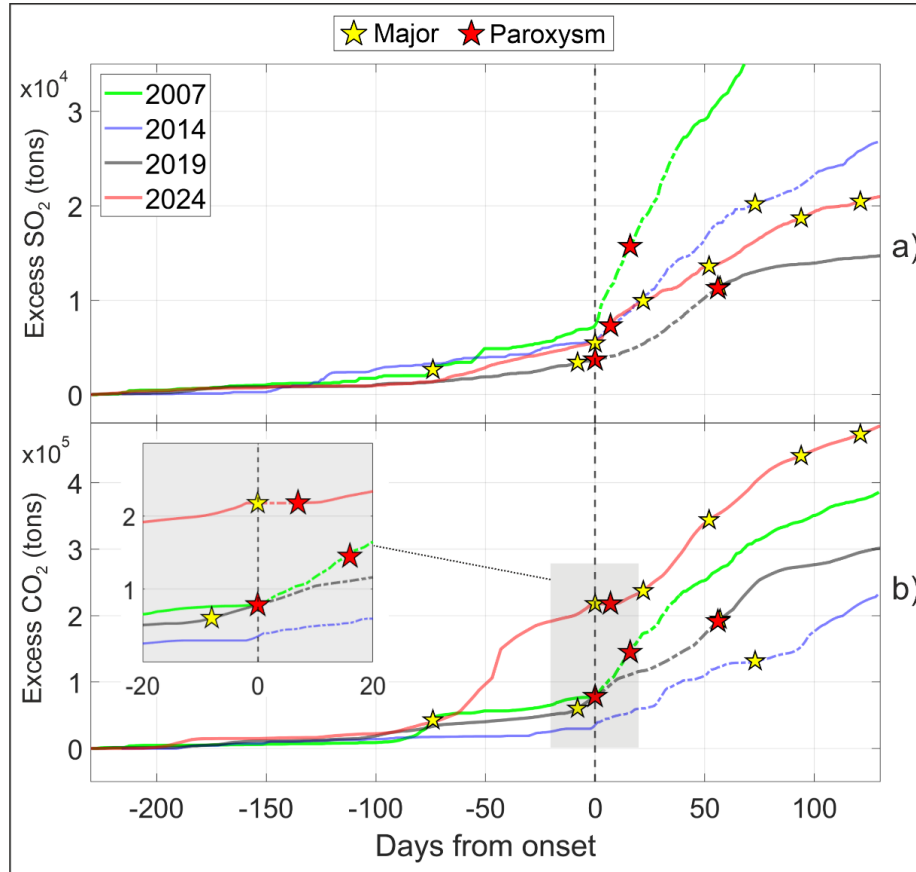


Figure 7.4 – Cumulative excess degassing trends for both SO₂ (a) and CO₂ (b). Time is expressed in days before/after eruption onset (set at 0). Dashed portions of the cumulative curves represent the effusive phases.

7.6 Conclusions

We have reported on a comprehensive analysis of SO₂ and CO₂ fluxes at Stromboli during the July 4-11 2024 eruptive sequence, highlighting the critical role of volcanic gas monitoring for understanding eruptive dynamics. We have shown that the eruptive sequence was anticipated by a sustained escalation in CO₂ flux, from background levels of ~900 t/d to > 2000 t/d in the ~ 60 days before event, hence peaking at 3800 t/d at onset. We conclude that a heightened supply of CO₂-rich, deeply sourced bubbles was the causal factor in triggering the eruption. We propose that such as above-normal CO₂-bubble supply caused the resident conduit magma to become more buoyant and effervescent, hence accelerating the frequency/energy of strombolian explosions and spattering until the summit North-east crater (partially) failed, opening of the effusive phase. Because degassing of

erupting shallow conduit magma dominated during the effusive phase, as indicated by high SO_2 fluxes and low CO_2/SO_2 ratios, we argue that the deep magma – source of the excess CO_2 precursory phase – may have stalled at some point in the plumbing system for 8 days (4-11 July), before being erupted in a violent paroxysm. Petrological and geochemical information on erupted pyroclasts of the July 11 paroxysm is critically needed to test this hypothesis. Comparison with previous eruptions (2007, 2014, 2019) reveals that pre-eruptive excess CO_2 is systematically observed prior to paroxysmal activity, confirming the utility of CO_2 plume monitoring as eruption forecasting tool at open-vent mafic volcanoes. In spite of these progresses, we are still far from a deterministic mid-term forecast of the exact timing of a paroxysm, and of the sequence of events that are to be expected.

CHAPTER 8

COUPLING UV CAMERA-DERIVED SO₂ FLUXES AND THERMAL OUTPUT AT MOUNT ETNA: A DECADE ANALYSIS ACROSS ERUPTIVE PHASES

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original draft, Software, Validation, Supervision, Resources, Methodology, Investigation, Formal analysis, Conceptualization, Data curation, Visualization.

Abstract

Volcanic gas emissions represent a fundamental expression of magmatic activity, and provide essential information on the state and evolution of a volcanic system. At persistently degassing, open-vent volcanoes, for example, volcanic SO₂ fluxes typically increase at, and at times before to, the transition from quiescent to eruptive activity. However, despite their critical importance, accurately quantifying volcanic SO₂ emissions remains challenging, owing to the complex and variable interaction of the volcanic plume with the surrounding atmosphere, and the often limited spatial and temporal resolution of observations. The UV Camera method, for example, potentially offers SO₂ flux time series of unprecedented temporal and spatial resolution, but difficulties in automation and caveats and limits in accurate SO₂ retrieval have so far limited to a few volcanoes the acquisition of robust, long-term SO₂ flux time series with fully autonomous instruments. Here, we present a long (~10-years) record of SO₂ flux emissions from Etna volcano in Italy, obtained using a permanent UV Camera system. We combine these SO₂ flux results with satellite-derived thermal flux data, in the attempt to investigate the link between SO₂ degassing dynamics and volcanic activity mode and style. We initially use thermal radiance to identify the main eruptive phases during the decade and we examine in detail the variations in SO₂ and thermal regimes, exploring their temporal trends during the pre-, syn- and post-eruptive periods. Our analysis demonstrate that, despite the challenges posed by variations in eruption source location, intensity, and instrumental limitations, the UV Camera system can successfully resolve a clear (above background) increase in eruptive SO₂ flux, with detection efficiencies of 61%, 72% and 72% for the pre-eruptive, syn-eruptive, and post-eruptive periods, respectively, and with improved performance during eruptions involving larger volumes of magma. We identify a characteristic (averaged) SO₂ flux increasing pattern, from ~1957 t/d during inter-eruptive periods, to ~2300 t/d and ~2600 t/d in the days before and after the main eruptive phases, culminating into syn-eruptive SO₂ emissions of ~3800 t/d. Coupling the thermal-derived magma output rate with the SO₂ flux-derived magma supply rate within the shallow plumbing system, we also quantify, for each stage of eruptive phases, the volumetric imbalance between degassed and erupted magma. Our volumetric analysis confirms the large imbalance between degassed and erupted magma at Etna, with only ~27% of the ascending magma being effectively discharged during eruptions, and the remaining magma fraction (~73%) remaining stored in the intrusive magmatic system. We calculated that ~120 Mm³ of magma may have been stored in the 2015-2021 interval. Overall, our combined analysis highlights the strong variability in Etna's eruptive behaviour and

underlines the importance of integrating gas and thermal observations to assess the magmatic balance and to improve the interpretation of monitoring data.

8.1 Introduction

The emission of volcanic gases at active volcanoes is the ultimate effect of magma degassing, caused by decompression and/or crystallization of magma in the subsurface (Oppenheimer et al., 2014; Edmonds and Wallace, 2017; Edmonds et al., 2022). Monitoring volcanic gases hence plays a crucial role for understanding magma behaviour at depth, and ultimately how likely (and if so, when) magma is to erupt (Edmonds, 2021; Aiuppa et al., 2025a, 2025c). After water vapor and carbon dioxide, sulphur dioxide (SO₂) represents the third most common gas emitted by volcanoes (Oppenheimer et al., 2011), and its release is strictly linked to magma migration in the upper portion of magmatic systems (Scaillet et al., 1998; Shinohara, 2008), and/or to gas bubble separate ascent from stored magma in the upper crust (Edmonds et al., 2022). As such, monitoring the mass flux of SO₂ provides essential insights for the evaluation of volcanic hazards (Kern et al., 2022; Arellano et al., 2021; Lo Bue Trisciuzzi et al., 2024; Theys et al., 2019). Unlike many other gases, SO₂ has very low natural background levels and shows distinct absorption features in the ultraviolet (UV) spectral ranges, making it relatively easy to detect with spectroscopic techniques (Oppenheimer et al., 2011).

The relevance of SO₂ in volcanic systems has promoted continuous technical efforts to improve precision and accuracy of SO₂ flux records via ground-based and satellite-based remote sensing (e.g. Galle et al., 2003; Kantzas and McGonigle, 2008; Oppenheimer et al., 2011, 2014; Carn et al., 2017). The improved spatial and temporal resolution of today SO₂ flux measurements has unveiled the intrinsically nonstationary nature of volcanic degassing (Mori et al., 2013; Shinohara, 2013; Carn et al., 2017; Fischer et al., 2019; Arellano et al., 2021), in which periods of heightened emission alternate with period of reduced degassing (Kazahaya et al., 2004; Mori et al., 2013; Carn et al., 2017). While such fluctuations complicate the reconstruction of long-term emission inventories and the estimation of global volcanic gas budgets (Werner et al., 2019), they also offer the opportunity to identify and interpret unrest at restless volcanoes (Aiuppa et al., 2025a). Sustained increases in SO₂ emissions have often been detected in the run-up to eruption, confirming the pivot role of this gas as a precursor of magma ascent and decompression (Boichu et al., 2015; Aiuppa et al., 2023; Laiolo et al., 2022; Kunrat et al., 2022). Moreover, integrating SO₂ flux measurements with thermal or geophysical data (e.g., Coppola et al., 2019; Laiolo et al., 2022; Aiuppa et al., 2023) provides valuable insights to better understand the volcanic dynamics, hence contributing to more effective hazard assessment.

With the advent of UV Cameras (Mori and Burton, 2006; Bluth et al., 2007), SO₂ flux measurements improved their spatial and temporal resolution, hence with large potentials for volcanic

monitoring (Burton et al., 2015). However, these instruments typically require deployment in the proximity of the active vents that, while reducing light dilution effects (Campion et al., 2015), cause targeting the proximal and often most optical opaque volcanic plume portions, in which radiative transfer errors are larger (Kern et al., 2010a, 2010b; Kern et al., 2012; Kern et al., 2013). Furthermore, the proximity to the active vents often causes the plume to be only partially imaged by UV Cameras, due to morphological complexity of volcano summits or unfavourable wind directions (Delle Donne et al., 2022; Lo Bue Trisciuzzi et al., 2024). This, combined with challenges in site conditioning and maintenance, has made UV Cameras predominantly used for discrete surveys (Tamburello et al., 2011; Ilanko et al., 2020), while applications of permanent deployments for semi-continuous observations have so far been restricted to a limited number of volcanoes (Burton et al., 2015; D'aleo et al., 2016; Delle Donne et al., 2017; Kern et al., 2015; Wilkes et al., 2023).

Mount Etna, on the eastern coast of Sicily (Figure 8.1a), is one of the few volcanoes on Earth instrumented with one of such permanent UV Camera system, since 2014 (Delle Donne et al., 2019). Etna is the largest active volcano in Europe, and is well recognised for its nearly persistent activity (Branca et al., 2011). The volcano currently reaches a maximum elevation of 3329 m, and has a composed summit crater terrace consisting of four active main summit craters (Figure 8.1b), which have formed and expanded over the past decades: the Voragine Crater (VOR), the Bocca Nuova Crater (BN), the Northeast Crater (NEC), and the Southeast Crater (SEC) (Branca et al., 2011). Etna's activity is primarily characterized by continuous degassing from its summit craters, driven by passive gas release from shallow convecting magma (Allard, 1997), and usually accompanied by mild Strombolian activity. However, this background activity is often interrupted by effusive eruptions (Branca and Del Carlo, 2004; Branca and Del Carlo, 2005; Andronico et al., 2005; Branca et al., 2011) and more energetic, explosive (lava fountaining) episodes (Andronico et al., 2021b) that can last from a few hours to over 24 hours. These short-lived episodes are generally anticipated by a progressive intensification of Strombolian activity (Viccaro et al., 2014) that then evolves into lava fountains forming eruptive columns reaching heights of up to 14 km above the craters (e.g. Corsaro et al., 2017). Since the 1980s, these explosive events have become more frequent, often occurring in clusters of multiple lava fountains (Andronico et al., 2021b). These events are believed to be driven by an increasing influx of volatile-rich, mafic magma into a shallow magma storage zone located about 2 to 4 km beneath the summit. The interaction of this mafic magma with more evolved, shallower resident magma (Kahl et al., 2015; Corsaro et al., 2017; Giuffrida and Viccaro, 2017) is believed to trigger gas-driven pressure buildup over periods ranging from days to months (Giuffrida and Viccaro, 2017), ultimately resulting in eruption (Bonaccorso et al., 2013; Patanè et al., 2013; Corsaro et al., 2017; Cannata et al., 2018; Bonaccorso et al., 2021).

Here, we review and discuss the utility and implications of UV Camera observations at Etna, by presenting a nearly decade-long analysis (November 2015 to 2024) of SO₂ flux results obtained by the permanent UV Camera system (Figure 8.1b,c). The SO₂ flux time series is combined with satellite-derived thermal data obtained using the SEVIRI sensor, and with volcanic activity records (Figure 8.2a) extracted from the INGV-OE (Istituto Nazionale di Geofisica e Vulcanologia - Osservatorio Etneo) monitoring reports ([INGV-OE monitoring reports](#)). The aim of this integrated analysis is to evaluate the utility of UV Camera records for the understanding of degassing dynamics at Etna, and the possible forecast of the quiescent-to-eruption transition.

8.2 Methods

8.2.1 SO₂ flux measurements from UV Camera

The daily-averaged SO₂ flux time series used in this study (Figure 8.2b) was obtained through the manual processing of images recorded by a permanent UV Camera system (here referred also as UV3 system) installed on the “La Montagnola” ridge (Figure 8.1b), at an elevation of 2600 m. The system has been operational since 2014 and provides an optimal view of the southern sector of the summit crater terrace, located approximately 3 km away. The system employs two JAI CM-140GE-UV Cameras sensitive to UV radiation and fitted with band-pass optical filters (Full Width at Half Maximum, FWHM, of 10 nm). Because SO₂ absorption of solar radiation decreases sharply around ~320 nm (Vandaele et al., 1994), the system employs two narrow-band filters: one covering the 300-320 nm range, corresponding to the region of maximum absorption, and one covering the 320-340 nm range, located outside the absorption band. These co-aligned cameras capture daily image sequences (520x676 pixels at 10-bit resolution) at a frame rate of 0.5 Hz during optimal lighting conditions (e.g., from 7:30 a.m. to 12:30 p.m. UTC). Image acquisition is automated using a Fujitsu RX100 Workstation. Details of hardware and software have been described in previous work (D’Aleo et al., 2016, 2019; Delle Donne et al., 2019).

The daily acquired images are processed using a MATLAB custom code (Delle Donne et al., 2019), where pairs of images taken simultaneously by the two cameras are merged into single absorbance images, applying the Lambert-Beer law. This method allows for the quantification of the relative absorption of scattered UV sunlight by volcanic SO₂ within the camera’s field of view (FoV). The resulting SO₂ absorption images are then converted into SO₂ column density images by applying a proportionality ratio between absorbance and SO₂ column density. This ratio is established using an Ocean Optics USB 2000+ spectrometer focuses on a specific subregion of absorbance images, along with periodic field calibration campaigns using gas cells of known SO₂ concentration, using the methodology described in McGonigle (2007). The SO₂ column densities are then integrated along

a rectangular sub-area positioned above the crater terrace and capturing the full cross-section of the imaged plume. The so obtained integrated SO₂ column amounts (kg/m) are multiplied by plume speed (m/s) obtained by applying an optical flow algorithm tracking the movement of gas fronts in consecutive frames (Lucas and Kanade, 1981; Delle Donne et al., 2019), to obtain a 0.5 Hz SO₂ flux daily time series (kg/s). The SO₂ fluxes reported here represent daily-averaged values (t/d) obtained from averaging all SO₂ fluxes associated to the best conditions during the day. For this aim, our SO₂ flux time series is manually retrieved and validated by an operator, which allows manually excluding (from calculation of the daily average) all images taken under not-optimal conditions (e.g., presence of clouds, ash-rich plumes, etc.). These are relatively common during the highly variable weather conditions at the summit of the Etna volcano. To this aim, image sequences are visually checked by a co-exposed visible camera.

It is well known that cloud coverages and, even more, the presence of ash inside the imaged volcanic plume, are capable to reduce the in-plume SO₂ signal (e.g., Kern et al., 2010b, 2013), resulting in artificially low flux estimates. Moreover, UV Cameras focus on the nearby and often optically dense part of the plume, making them more vulnerable to errors related to multiple scattering of light within the plume, which is caused by aerosols and water droplets (Kern et al., 2010a, 2012, 2013). These radiative transfer effects can be somewhat alleviated by choosing frames with visibly clear (uncondensed or ash-free) plumes, as determined by the operator's review of visible camera images, but cannot be completely removed from measurements. Light dilution can also affect the measurements, as some photons may be scattered toward the instrument without traversing the volcanic plume. Since these photons do not contain spectral absorption structures, they generate a dilution of the measured column density. This effect is primarily dependent on the distance between the observation point and the target plume (Campion et al., 2015; Kern et al., 2010a) and can weaken the SO₂ signal, resulting in a significant underestimation of SO₂ column densities. The light dilution effect at the UV3 site was quantified (using the methodology of Campion et al. 2015) at ~32% by Aiuppa et al. (2023). The same authors also inferred an average random error of about 20% for each measurement.

The UV Camera's field of view (Figure 8.1b,c) also limits the detectable gas emissions to those released from the southern portion of the crater terrace, which encompasses mainly the South-East Crater (SEC) and the southern portion of central craters (VOR+BN) (D'Aleo et al., 2019). Hence, the system's configuration prevents from capturing emissions from the North-East Crater (NEC), and underestimates emissions from the central craters, except during intense explosive events that generate high sustained plumes (D'Aleo et al., 2019). As a result, our SO₂ flux measurements do not

reflect the total plume emissions from Etna, which are regularly monitored with scanning UV spectrometers located at greater distances (Salerno et al., 2009a).

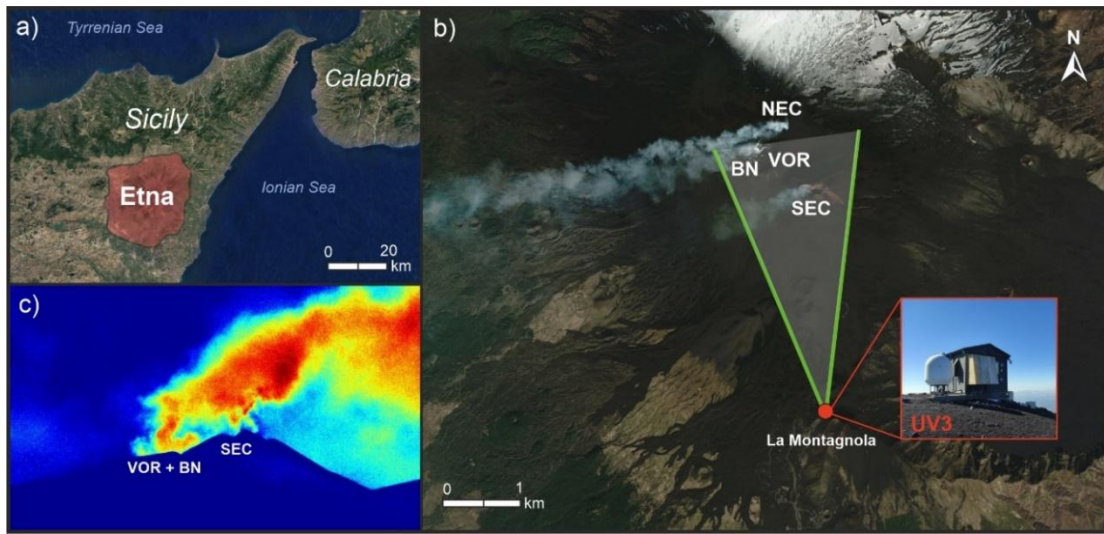


Figure 8.1 – a) Geographical location of Mount Etna; b) View of the Etna's summit craters and location of the UV Camera system (UV3) at "La Montagnola" ridge. The UV Camera field of view is represented as the area encompassed between the green lines; c) An example of absorbance image taken from the UV3 system.

8.2.2 Radiant heat flux from SEVIRI

Eruptive episodes at Mount Etna are also characterized by satellite-based thermal observations (Figure 8.2c). Here we use the radiant heat power (VRP, in Watt) derived from space-borne images acquired by the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) on board Meteosat Second Generation (MSG) satellite. The SEVIRI sensor acquires images in the middle and thermal infrared region of the electromagnetic spectrum, that are particularly useful in detecting and tracking hot spots associated with fires and effusive activity (Hirn et al., 2008; Bonaccorso et al., 2011). Thanks to the high frequency of acquisition (~ 15 min), SEVIRI is also the most suitable space-based instrument for observing and tracking short-lived eruptive events such as Etna's lava fountains (Ganci et al., 2011, 2016, 2018a, 2018b; Calvari et al., 2021a, 2022a). However, because of the low spatial resolution of SEVIRI images (3 km at nadir), most of the lower intensity thermal activity, such as those related to strombolian activity, spattering or small lava flows cannot be detected. Another issue is that, in case of intense eruptive activity, the SEVIRI channel ($3.9 \mu\text{m}$) used to observe high temperature features often saturates, causing a possible underestimation in the recorded signal.

SEVIRI multispectral images are retrieved via the CL-HOTSAT system, a new version of the HOTSAT system (Ganci et al., 2011; Ganci et al., 2016; Ganci et al., 2018a, 2018b) implemented in the OpenCL standard to obtain near-real-time monitoring of volcano-related thermal activity. The CL-HOTSAT system developed at INGV-OE allows for automatic detection of thermal anomalies

and quantification of thermal activity through calculation of the radiant heat flux. The system automatically downloads the SEVIRI images in near real-time using the API service provided by EUMETSAT (the Data Access API Client) using a Python-based routine, and locates the thermal anomalies (hotspots) within each image, computing the radiant heat flux for each hotspot pixel. Finally, the total radiant heat flux is obtained by summing the individual radiant heat flux of all hotspot pixels for each image.

8.2.3 Magma input rate inferred from UV Camera measurements

Open-vent volcanoes are globally significant emitters of heat and volatiles that, during their persistent regular activity, produce more gas than explained by degassing of erupted magma (Francis et al., 1993; Kazahaya et al., 1994). Such excess degassing implies either the existence of an efficient magma convection mechanism within the volcanic plumbing system that allows huge parcels of magma to degas without being erupted (Shinohara, 2008), or an effective gas separate ascent from stored magma (Edmonds et al., 2022). As a direct consequence, during ordinary activity the magma supply/degassing rate (Q_{in}) into the shallower portion of magmatic plumbing system exceeds the magma discharged rate (Q_{out}) from the active vents.

Calculation of the magma supply (input) rate to the volcanic system can be performed starting from SO_2 flux measurements (Aiuppa et al., 2023; Delbrel et al., 2024; Coppola et al., 2019). The conversion of SO_2 flux into magma input (degassing) rates is commonly achieved using a petrological approach based on the following equation:

$$Q_{in} = \frac{\phi_{SO_2}}{2\Delta X_S \cdot (1-x) \cdot p_m \cdot 86.4} \quad (\text{Eq. 8.1})$$

where Q_{in} represents the magma input rate (in m^3/s) above the SO_2 exsolution level (the depth at which sulphur starts to degassing from melt), ϕ_{SO_2} is the SO_2 flux (in t/d), ΔX_S represents the sulphur loss during degassing (inferred from the difference between parental melt and residual post-eruption sulphur contents in Etna's magmas), and x and p_m correspond to melt crystallinity and density (in kg/m^3), respectively. The value of 86.4 is the unit's conversion factor from t/d to kg/s.

In brief, the equation links the SO_2 emissions with the volume of magma feeding the volcanic system, providing an estimate of magma supply/degassing rates. The method relies on knowledge of the initial (pre-degassing) and final (post-emplacement) sulphur content in the melt and has been extensively applied at Mount Etna to constrain magma degassing budgets over various timescales (Allard, 1997; Allard et al., 2006; Coppola et al., 2019; D'Aleo et al., 2019).

The application of Equation 8.1 requires a model that accurately describes sulphur (S) behaviour during the magmatic degassing of Etna's alkali basalts. Such a model should, starting from the known initial S content in parental magmas, predict the fraction of S lost to the vapour phase

along the magma decompression path down to surface pressure (Allard, 1997). Deeply trapped primitive melt inclusions are commonly used to estimate the initial (pre-degassing) S content of parental melts (Wallace and Edmonds, 2011). At Etna, melt inclusion studies (Métrich et al., 2004; Spilliaert et al., 2006) indicate a parental melt S content of approximately 3000 ppm (0.3 wt%), along with a systematic decrease in dissolved S with decreasing pressure. However, more recent work (Gennaro et al., 2019) has found more variable parental melt S contents in magmas. Sulphur loss to the vapour phase is observed to initiate at Etna at pressures ≤ 150 MPa (Spilliaert et al., 2006). This 3000 ppm estimate, along with an assumed 95% S extraction from the melt upon degassing at pressures ≤ 150 MPa ($\Delta X_S = 0.00285$), has been used in previous estimates of magma input rates (Allard, 1997; Allard et al., 2006; Coppola et al., 2019; Aiuppa et al., 2023).

We used Equation 8.1 to convert our daily averaged SO_2 fluxes (ϕ_{SO_2}) into a daily averaged magma input rates (Q_{in}) by taking into account the possible full spectrum of ΔX_S and crystallinity x values in Etna's magmas. For this aim, we used a 3000 ppm parental melt as an upper limit scenario (ΔX_S at 0.00285 and $x = 0$), and a 2200 ppm parental melt (from Gennaro et al., 2019) ($\Delta X_S = 0.00205$ and $x = 0.3$) as a lower limit scenario, following the same conditions used in Aiuppa et al. (2023). An intermediate scenario is then obtained from averaging the minimum and maximum estimates (see Supplementary Figure S8.2b). In all calculations, melt density (ρ_m) is fixed at 2700 kg/m^3 , as a typical density of basaltic magmas, used in various previous works at Mount Etna (e.g., Andronico et al., 2018; Negro et al., 2013; Greco et al., 2022). Note that, as the SO_2 fluxes measurements reported here do not correspond to the total bulk plume (see Paragraph 8.2.1), the obtained magma input rates can approximate the total magma input rates only if /when activity is concentrated at SEC. Based on these magma supply rates, the cumulative volume of degassed magma over time can be established by integrating the supply rates over a selected period.

8.2.4 Magma output rate using SEVIRI thermal anomalies

During an eruption, the radiant heat flux radiated by lava is calculated from SEVIRI images, giving as result a heat flux curve describing the event. This heat flux curve can be converted in time-averaged discharge rate (TADR or output rate, in m^3/s) adopting a straightforward relationship between the radiant heat loss from an active lava flow and the effusion rate (e.g., Harris et al., 1997; Wright et al., 2001):

$$TADR = \frac{Q}{\rho(C_p \Delta T + C_L \Delta \phi)} \quad (\text{Eq. 8.2})$$

where Q is the total thermal flux obtained by summing up the radiant heat flux computed for each hotspot pixel, ρ is the lava density, C_p is the specific heat capacity, ΔT is the difference between the

eruption temperature and the temperature at which flow stops, C_L is the latent heat of crystallization, and $\Delta\phi$ is the volume percent of crystals that form while cooling through ΔT .

The conversion from radiant heat flux to volumetric flux depends on such lava parameters and has to be determined depending on flow conditions (Harris and Baloga, 2009; Pieri and Baloga, 1986). Since we do not usually have these source parameters for the application of Equation 8.2, the most appropriate approach is to adopt a range of possible values. For the aim of this work, we report a minimum and maximum TADR estimates by adopting the extreme values reported in Harris et al. (2000, 2007), which are considered suitable for Etna lavas (see also Del Negro et al., 2016). An intermediate estimate was then derived by averaging these boundary cases (see Supplementary Figure S8.2b). However, this approach is based on the implicit assumption that the heat flux is generated by an active lava flow (Coppola et al., 2013). Consequently, the model fails when the lava is not extruded but rather retained in the vent, as in the case of open-vent activity. In such circumstances, the thermal proxy provides a minimum estimate of the magma circulation (convection) at very shallow depth, rather than effusion rates, as long as the top of the magma column is sufficiently shallow to produce thermal anomalies on the surface (Werner et al., 2017; Aiuppa et al., 2018).

For effusive events, calculation of TADR is based on a thermal budget that assumes equilibrium between the heat retained in the eruptive volume and that radiated from the surface. This approach relies on two key assumptions: (i) the volumetric flux is proportional to the surface area of the lava flow, and (ii) the lava surface cools exponentially with time and distance from the vent (Harris and Baloga, 2009). However, these assumptions are not valid for short-lived, transient events such as Etna's paroxysmal episodes, during which a steady-state regime, and thus thermal equilibrium, cannot be established (Garel et al., 2012). During short-lived paroxysmal events, thermal anomalies are often affected by the tephra or ash column produced during the eruptive climax, which can attenuate or obscure the signal. Conversely, when the anomaly is not obscured, the high radiant power released can easily saturate the sensor. As a result, TADR cannot always be reliably calculated during the peak fountaining phase. For such events, estimation of event intensity can instead be obtained by modelling the radiant heat flux curve recorded during the cooling-dominated phase of activity (Ganci et al., 2012, 2013), by incorporating the radiating area and applying constraints on the minimum and maximum expected lava thickness for these short-lived episodes (see also Ganci et al., 2023). This technique has been successfully applied several times at Etna volcano for modelling purposes (e.g., Cappello et al., 2018), with an uncertainty of $\pm 30\%$, derived from the radiant heat flux measurements (Ganci et al., 2016, 2023).

A cumulative volume of erupted magma over time can be retrieved for each estimated scenario by integrating the respective TADR values.

8.3. Results

8.3.1 Characterization of volcanic activity using SO₂ fluxes, thermal anomalies and INGV-OE monitoring reports

Throughout the ~10-year observation period, Etna exhibited a wide spectrum of volcanic activity behaviours, with all summit craters showing intermittent or simultaneous activity ranging from passive degassing to Strombolian explosions and lava effusion. Activity was primarily concentrated at the South-East Crater (SEC) and Voragine (VOR), while Bocca Nuova (BN) and the North-East Crater (NEC) played a secondary role (Figure 8.2a). Passive degassing was frequently punctuated by episodes of strombolian activity from one of the summit craters, by effusive eruptions of variable duration, and by numerous paroxysmal lava fountain episodes (Figure 8.2a). Lava fountains are highly energetic but short-lived events, usually lasting only a few hours, that can either occur as isolated episodes or in clusters (eruptions periodically repeating at intervals of days/weeks) (Andronico et al., 2021b). From the INGV-OE weekly reports, we identify 75 paroxysmal lava fountain events, representing the most intense yet transient form of magma eruption and 32 lava flows of variable duration. We calculate that the majority of the eruptive events show small inter-event times (< 15 days, Figure 8.3a), particularly due to the clustering of lava fountains (Andronico et al., 2021b).

Our nearly decade-long dataset reveals large temporal fluctuations in SO₂ fluxes (Figure 8.2b), with daily mean values averaging 2468 ± 2090 t/d (range: 30 to ~20,000 t/d). Although the UV Camera system employed may underestimate the total SO₂ output due to its preferential view of the southern sector of the summit craters, our reported SO₂ fluxes are within the range reported previously (Carn et al., 2016, 2017; Boichu et al., 2015; Grandin et al., 2024; Corradini et al., 2020; Salerno et al., 2009a; Queißer et al., 2019). Despite the oscillating and complex nature of the time series, observation of Figure 8.2 demonstrates a broad link between SO₂ levels and volcanic activity state. The largest (> 3000 t/d) SO₂ fluxes are typically observed in correspondence to eruptive episodes. In particular, a systematic pattern of gradual SO₂ increase prior to eruption onset, followed by an abrupt decrease immediately after eruption cessation, is evident before the 2017, late-2018, 2021 and mid-2022 eruptions (see also Supplementary Figure S8.1). This pattern is, however, less evident in some other events. The lowest fluxes, typically < 2500 t/d, are recorded during phases of passive degassing and mild Strombolian activity, in what we refer as the background persistent activity level of Etna.

The distribution of SO₂ fluxes (Figure 8.3b) shows an asymmetric distribution, with a peak at ~1000 t/d. The frequency of observations decreases progressively toward higher flux values, with a long tail extending beyond 10,000 t/d. This skewed distribution indicates that most of the measured

fluxes cluster between a few hundred and a few thousand tons per day, whereas extreme values are comparatively rare.

Thermal anomalies detected by SEVIRI imagery (~15 min of temporal resolution) provide clear evidence of lava outpouring from vents or scoria emission from sustained Strombolian activity (Spina et al., 2025). These anomalies account for nearly 10% of the total observation time and are consistently associated with phases of elevated volcanic activity (Figure 8.2c). The mean VRP value during the decade is 0.15 GW, with values spanning from 0 to 36 GW. Major eruptive phases are characterized by sharp increases in thermal output, with lava emissions typically exceeding 10 GW, whereas inter-eruptive periods show relatively stable VRP values below 1 GW. Minor radiative peaks scattered throughout the record correspond to episodes of variable lava output that are consistently documented in daily monitoring reports.

The distribution of daily-averaged radiative power shows a skewed profile (Figure 8.3c), with a first class of high frequency and low VRP values, and a second class of less-abundant higher VRP values at ~0.3 GW, likely marking the transition from quiescent degassing or weak Strombolian activity to more energetic eruptive behaviour.

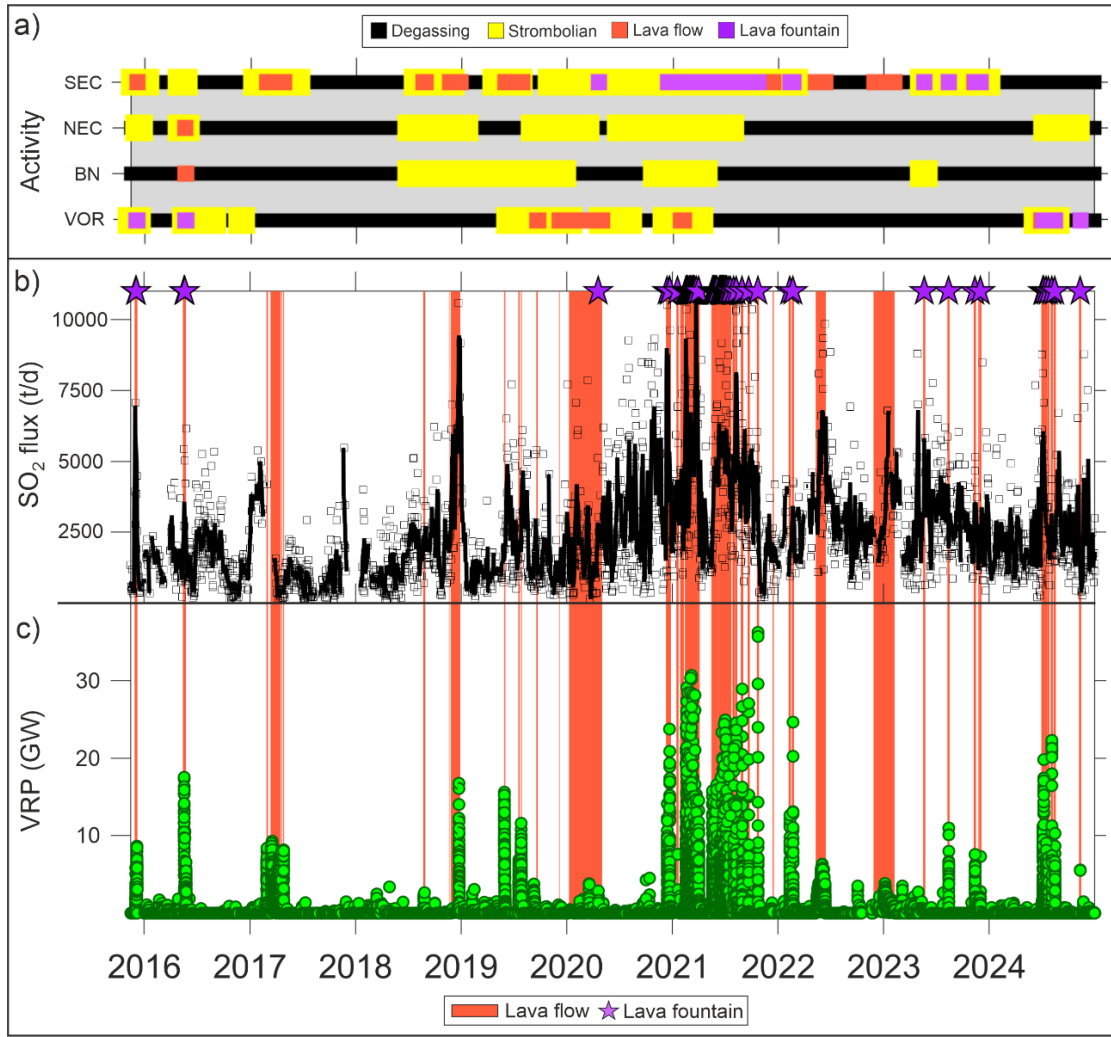


Figure 8.2 – a) Eruptive activity at Etna’s summit craters, inferred from INGV-OE monitoring reports; b) SO_2 daily-averaged flux from UV Camera. Black squares represent the raw data, and the black solid line represents a 7-day moving average. Here and in the following panel we also show all the not-ordinary eruptive events (effusions and lava fountains) reported in panel “a”; c) Volcanic radiative Power (VRP) from SEVIRI images.

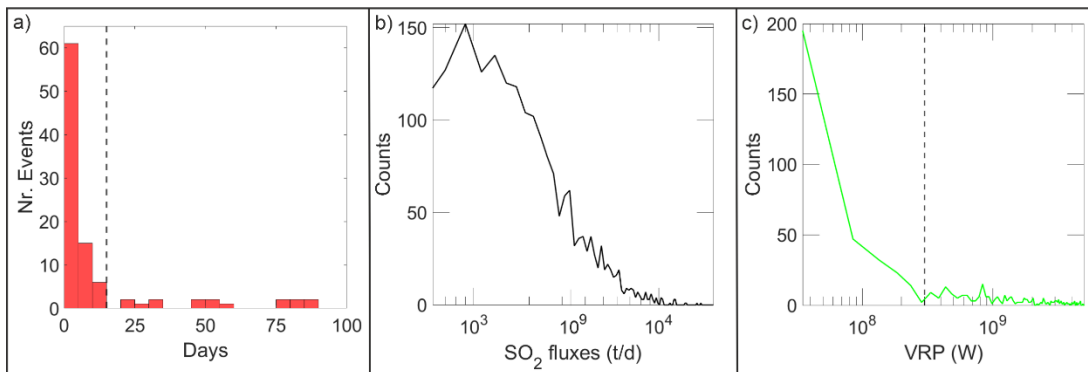


Figure 8.3 – a) Occurrence time between eruptive events, defined as the difference between the onset of an event and the termination of the previous one. The majority of events ($\sim 77\%$) show inter event times < 15 days, as shown from the dashed line. b) Distribution of daily-averaged SO_2 fluxes. c) Distribution of daily-averaged VRP values. The dashed line marks two different populations, separating abundant low-VRP values from less-abundant high-VRP values.

8.3.2 Magma input/output rates

The two distinct (upper and lower scenario) magma input rates (cfr. 8.2.3) are illustrated as a grey area in Supplementary Figure S8.2b, together with the intermediate scenario (black line) obtained by averaging the maximum and minimum estimates. Based on the intermediate scenario, the mean magma input rate (Q_{in}) during the observed period (late-2015 to 2024) is $2.63 \pm 0.87 \text{ m}^3/\text{s}$, with a range from 0.03 to $22.30 \text{ m}^3/\text{s}$. The magma input rate sensibly accelerates in proximity of eruptive events, generally exceeding values $> \sim 4 \text{ m}^3/\text{s}$, accordingly to previous studies at Etna (Allard, 1997; Allard et al., 2006; Coppola et al., 2019; Aiuppa et al., 2023).

The intermediate scenario of the magma output rate is illustrated in Supplementary Figure S8.2b as green triangle marks, while the upper and lower scenarios are depicted as green vertical bars (see 8.2.4). While the magma input rate generally exceeds the magma output during background (persistent) activity, this trend reverses during most eruptive events. Our output rates exhibit notable variations across the wide range of volcanic activity, spanning from 0 to $>20 \text{ m}^3/\text{s}$ during more energetic phases of lava emission (max value: $49 \text{ m}^3/\text{s}$), reaching the highest values during paroxysmal lava fountain events. The mean magma output rate (intermediate scenario) during the observed period is $0.40 \pm 0.11 \text{ m}^3/\text{s}$, which is consistent with previous annual-averaged values (Wadge et al., 1975, Wadge and Guest, 1981; Harris et al., 2011).

By integration in time, the cumulative volume of erupted (output) and degassed (input) magmas can be calculated.

8.4. Discussion

UV Cameras have increasingly been exploited in volcanology in the last two decades (Burton et al., 2015). Applications have been the most diverse, including the quantification of eruptive gas masses from small explosive events (Mori and Burton 2009), the resolution of different degassing modes (Tamburello et al., 2012), the spatial resolution of different degassing sources at multi-vent volcanoes (Pering et al., 2020; Delle Donne et al., 2022), the expedite quantification of SO_2 budgets during campaigns (Tamburello et al., 2014), and the correlation with geophysical parameters to examine the source processes controlling volcano seismicity and infrasound (Nadeau et al., 2011). In contrast, the contribution of UV Cameras to volcano monitoring has been more modest, especially because volcanoes instrumented with permanent instruments are relatively few (e.g., Stromboli: Delle Donne et al., 2017, Lo Bue Trisciuzzi, 2024; Kīlauea: Kern et al., 2015; Lascar: Wilkes et al., 2023). Our Etna results here, with their continuity of observations, can hence help evaluating the potential of UV Cameras in volcano monitoring.

8.4.1 Can we track Etna's eruptive behaviour using SO₂?

Our ~10-year record opens the way to quantitatively testing the possible link between SO₂ flux and volcanic activity state (cfr. 8.3.2). To achieve this task, we use a procedure in which the largest (most voluminous) eruptive events in the Etna's eruption catalogue ($n = 107$, 75 lava fountains + 32 lava flows; Figure 8.2a) are selected and then grouped into main eruptive phases. Given the tendency of Etna's eruptions to concentrate in clusters (Andronico et al., 2021b), grouping closely spaced events into single eruptive phases let us to analyse the degassing signals prior, during and after each phase, preventing overlaps between pre- and post-eruptive signals of successive events and allowing a clearer assessment of degassing trends. Specifically, we identify the dominant events in the eruption record using the volcanic radiative power (VRP; Figure 8.2c) as a proxy for eruption magnitude. This stands on the assumption that a larger thermal output corresponds to a more sustained and voluminous eruption of lava, as the intensity of thermal emissions reveals the fractional pixel areas invaded by active flows (Wright et al., 2001; Harris and Baloga, 2009), under given compositional, insulation, rheological and ambient conditions (Harris et al., 2000, 2007, 2010).

We first use a VRP threshold of 0.3 GW (Figure 8.3c) to mark the transition between quiescent and eruptive regimes and fix the onset of an eruptive phase at the specific day where the 0.3 GW VRP threshold is first reached. Termination of an eruptive phase is chosen at the last day with VRP above the threshold, once no positive detections are encountered in the following 30 days, grouping all the positive detection within this interval into a unique eruptive phase (see Supplementary Figure S8.3). Use of such 30-day interval is motivated by the inter-event (repose) periods being <15 days for the majority (~77 %) of the Etna's eruptive events (Figure 8.3a). Hence, a ± 15 -day (30 day) interval encompasses the duration of the pre- and post-eruptive periods of most of the eruptive events in the INGV-OE catalogue. Use of this threshold makes resolution of quiescent vs. eruptive behaviours well consistent with eruptive activity reports in INGV-OE bulletins and can be considered as a minimum transition value, since the distribution of daily-averaged VRP (Supplementary Figure S8.3) shows eruptive peaks consistently above 1 GW. This procedure allows excluding from further analysis the eruptive events of shorter duration and/or reduced volume, which are unlikely to leave a detectable signal in the SO₂ flux record.

Applying this approach, and clustering into a single phase events repeating at short intervals, we identify 18 eruptive phases (Supplementary Table S8.1 and Supplementary Figure S8.3). These eruptive phases (marked in yellow in Figure 8.4) broadly coincide with the main steps of the cumulative thermal curve (green line in Figure 8.4; derived from the thermal flux time series of Figure 8.2c). Each phase, reported in Supplementary Table S8.1 together with its cumulative thermal energy and erupted volume, corresponds to a significant short-term increase in thermal flux. Collectively,

these phases account for more than 90% of the total thermal energy released during the study period. Their durations range from 1 to 126 days, with released energies between 2.18×10^9 and 1.18×10^{11} J.

We next quantify the background SO₂ flux levels by considering all the SO₂ flux results taken in the inter-eruptive periods (Figure 8.5a, top panel), e.g., outside the eruptive phases define above. The average background (e.g., inter-eruptive) degassing flux of Etna for the study period is 1957 ± 1470 t/d (range, from 30 to 9328 t/d; Figure 8.5a). In contrast, the syn-eruptive SO₂ flux (e.g., that corresponding to all flux results obtained with each of the 18 eruptive phases) is twice as much higher, averaging at 3828 ± 2686 t/d (Figure 8.5a, third panel from top). This analysis confirms that eruptions at Etna are effectively associated with enhanced SO₂ emissions. If we concentrate on the 15-day intervals before (pre-eruptive) and after (post-eruptive) the 18 eruptive phases, we find intermediate flux levels of respectively 2302 ± 1481 t/d and 2595 ± 1929 t/d (Figure 8.5a, second and fourth panels from top), suggesting that major eruptive phases at Etna are broadly preceded and followed by above background SO₂ levels.

To further illustrate the variability of the SO₂ flux at, and around (before and after), an eruption, we examine the degassing trends for each of the selected eruptive phases, shown individually in Supplementary Figure S8.4. This figure reveals that each of the eruptive phases considered is associated with its distinctive degassing signal. More importantly, we find that the pre-, syn- and post-eruptive flux increase is not systematic, but rather event-specific. As shown by Figure 8.5a, for some eruptive phases, the pre-, syn-, and post-eruptive mean fluxes fall below the quiescent (inter-eruptive) flux mean. Furthermore, the quiescent flux distribution (top panel of Figure 8.5a) overlaps substantially with the eruptive flux ranges (lower 3 panels of Figure 8.5a). Taken together, these results indicate that the quiescent vs. eruptive SO₂ flux distributions are not distinctly separated, but rather broadly overlapping. This means that while the SO₂ flux allows resolving the “averaged” quiescent vs. eruptive behaviour, only a fraction of the eruptive phases does exhibit an SO₂ degassing regime well resolvable above the background.

We quantify this fraction by estimating the “success rate” of the SO₂ flux trends in anticipating or accompanying eruptions. Success is defined here by the condition in which the mean value of a given pre-, syn-, or post-eruptive period exceeds the inter-eruptive SO₂ mean of 1957 t/d. Results (Figure 8.5b) indicate that, across all selected eruptive phases, success rates are ~61%, ~72% and 72% for the subpopulations of pre-, syn- and post-eruption period, respectively. Nicely, we find that success rate increases for all categories with increasing eruptive volumes (Figure 8.5c) (e.g., when eruptive phase associated with low eruptive volumes are progressively excluded from calculation), peaking at 85% (syn-eruptive) and ~83% (pre- and post-eruptive) for eruptive volumes between 2

and 5 Mm³, where the majority of phases fall (~70%, see also Supplementary Figure S8.5). We conclude that, while our UV Camera-derived flux record may fail to resolve from the (large) SO₂ background (sustained by passive degassing) most of the less voluminous eruptive phases, it can nicely detect more voluminous eruptive phases at Etna.

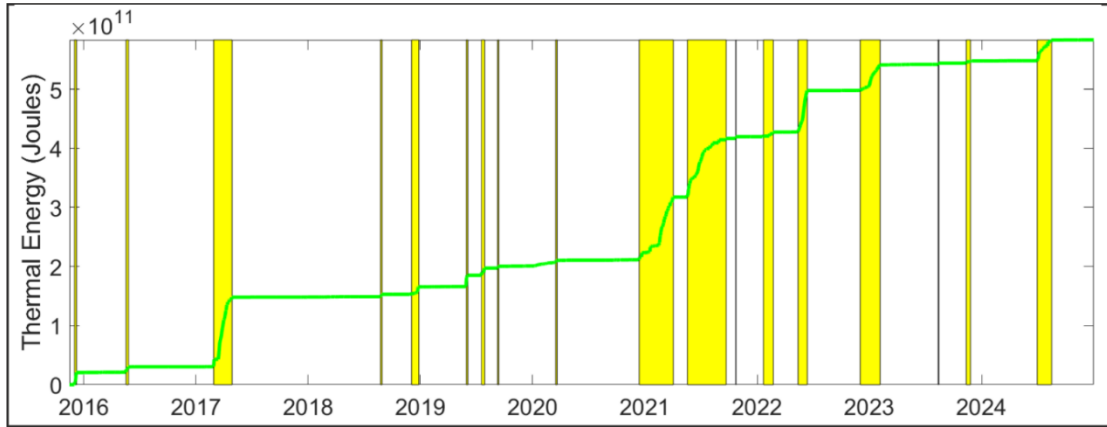


Figure 8.4 – Main eruptive phases detected following variations in thermal radiance. The green line represents the cumulative radiant heat curve obtained by cumulating our daily-averaged VRP values. The selected eruptive phases broadly follow the main steps of the cumulative thermal curve.

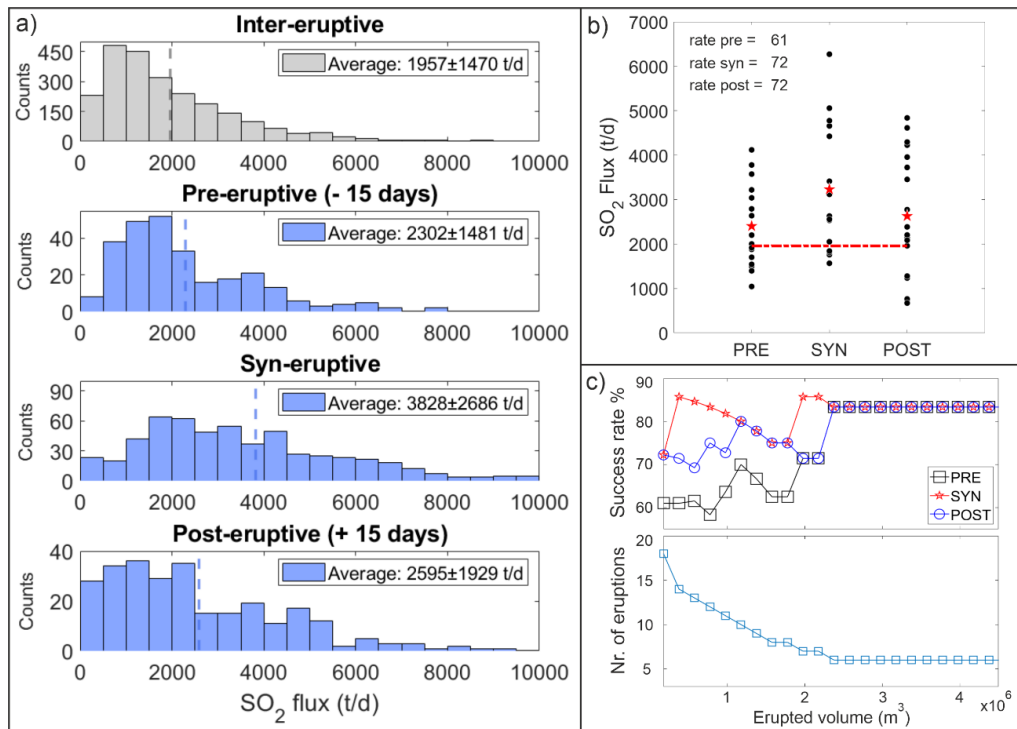


Figure 8.5 – a) Distribution of SO₂ fluxes from UV Camera during inter-event and pre-, syn-, post-eruptive periods. The dashed lines show the mean value of the distribution. b) Success rate of the employed system to resolve a mean degassing value above the background value prior, during and after each eruptive phase. Black dots show, for each eruptive phase, the mean degassing value during the pre-, syn- and post-eruptive period. Red stars show the average value between all eruptive phase across the corresponding period. c) Success rate in function of erupted volumes. The success rate generally increases progressively excluding smaller eruptions.

8.4.2 A look into longer-term (monthly to yearly) thermal and SO₂ trends

Etna's activity is known to alternate periods of heightened and reduced activity over a variety of timescales (Behnke and Neri, 2003). In this section, we investigate if, and to what extent, our combined SO₂ and thermal records can capture the oscillations in volcanic activity state taking place at Etna over timescales of months to years, e.g., over longer timescales than days, as investigated above (cfr. 8.4.1 and Figure 8.5). For the scope, we analyse the temporal changes in the slopes of the cumulative trends of thermal (Figure 8.6b) and SO₂ (Figure 8.6c) output, and their correlation with eruptive activity (Figures 8.6a, 8.6d).

During the late-2015 to 2024 interval, the thermal and SO₂ outputs have varied widely, as indicated by large spread of slopes of the cumulative curves (values displayed in panels b and c). The changes in the slope of the cumulative thermal energy curve (Figure 8.6b) match well the changes in eruptive style and energy (from INGV-OE monitoring bulletins, sequence reported in Figure 8.6a), supporting the ability of satellite-based thermal records to resolve between inter-eruptive and eruptive regimes. From this, and in tandem with SO₂ flux record, we can broadly identify during the investigated period the alternation of three main activity styles, shown with different colour gradation in Figure 8.6.

Firstly, we identify a year-long (from mid-2017 to mid-2018) phase of quiescence (green shaded area in Figure 8.6), characterized by passive degassing from summit craters (Figure 8.6a), and being consistently associated with the lowest thermal output (mean slope: ~1.84 MW). Nicely, the cumulative SO₂ trend is also the flattest in this period (corresponding to an average flux of 1054 t/d) (Figure 8.6c). We take this quiescent degassing period as representative of baseline (background) persistent open-vent degassing activity at Etna.

Thermal and SO₂ output excess relative to this baseline is typically associated with escalating eruptive activity. For example, periods of slightly higher thermal output (and time-averaged values under 0.3 GW; yellow shaded areas in Figure 8.6) contribute modestly to the overall thermal budget, and are linked to transitional eruptive behaviour. This is characterized by relatively modest/infrequent eruption of magma (in which effusive and paroxysmal events are relatively modest in size), interspersed within a dominant passive degassing mode associated with mild Strombolian activity episodes from summit craters (Figure 8.6a). These periods include the years 2016, 2019-2020, most part of 2022, and 2023-2024. In general, intermediate SO₂ fluxes of 2000-2700 t/d are observed during these phases of moderate thermal energy and subdued eruptive activity.

Finally, periods of high thermal output (overall higher than 0.3 GW), characterized by averaged thermal fluxes exceeding 1 GW (red areas in Figure 8.6), are clearly associated with more vigorous

eruptive episodes. These periods encompass the 2017 and mid-2022 effusions and the 2021 large eruptive sequence in which two main clusters of lava fountains took place (Aiuppa et al., 2023; Bruno et al., 2022; Carleo et al., 2023). The 2021 sequence represents the most energetic phase of activity during the decade, and is consistently associated with a sustainedly high SO₂ mass output (average SO₂ flux, 4257 t/d; Figure 8.6c). During this ~9 months-long period, the largest fraction (71%) of the lava fountains occurred, with an average rate of ~5 events for month (Figure 8.6d).

We calculate the cumulative excess SO₂ mass (black solid line in Figure 8.6c) by removing from the SO₂ degassing trend the averaged flux (of 1054 t/d) characteristic of the 2017-2018 quiescent degassing period. This cumulative excess curve exhibits a segmented behaviour that is broadly reminiscent of that identified from the thermal output trend (of Figure 8.6b). However, careful scrutiny of Figure 8.6 shows that the high-thermal phases of volcanic activity (red areas in Figure 8.6) are systematically anticipated by precursory slope increases in the cumulative excess SO₂ (at relatively invariant thermal output regime. These transitional phases (marked in orange shades in Figure 8.6) typically occur ~1 month prior the 2017 and mid-2022 eruptive phases, and up to ~6 months prior onset of the larger 2021 eruptive sequence (Figure 8.6c), and are associated to average SO₂ flux values well above the background value of 1054 t/d (Figure 8.6c). We conclude that phases of heightened SO₂ degassing do predate the escalation in volcanic activity during major eruptive periods, as in 2021. This precursory SO₂ phase is interpreted to reflect the decompressional degassing of magma, resupplying the Etna's shallow plumbing system, and accumulating at depth prior to surface emplacement in a major eruptive phase/sequence (Aiuppa et al., 2023).

INGV-OE reported the occurrence of 75 paroxysmal events from November 2015 to 2024. These short-lived eruptions are characterized by the formation of ash-laden eruptive clouds several kilometres high (Calvari et al., 2021a). These transient phenomena, typically lasting only a few hours, are not randomly distributed, but rather occur in clusters (Figure 8.6d) during periods of heightened thermal and SO₂ degassing activity (such as in 2021). Our analysis shows that our UV Camera based SO₂ flux observations have the potential to anticipate the onset of these main lava fountain cluster, with obvious benefits for risk management in this highly touristic area.

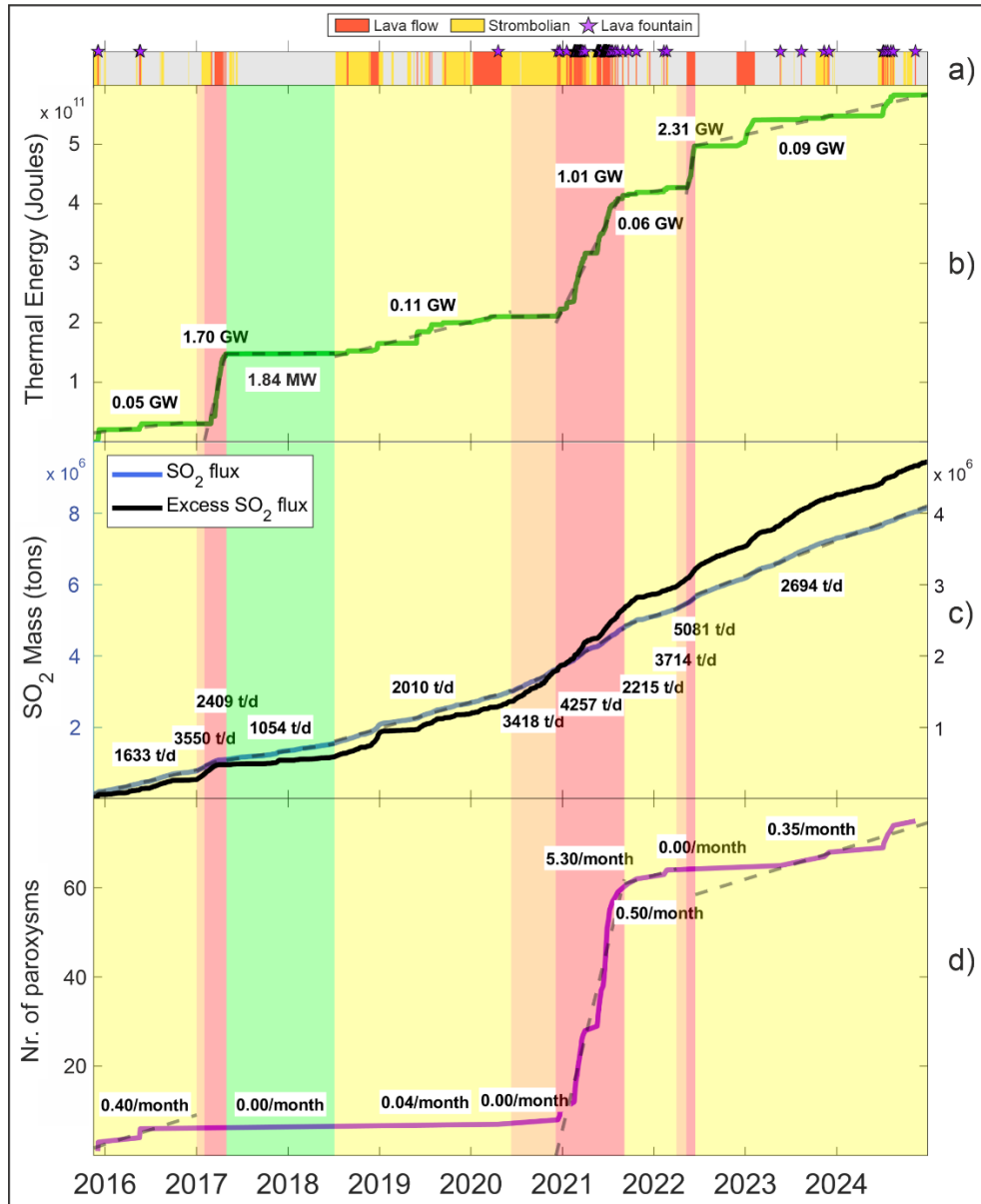


Figure 8.6 – Main phases of volcanic activity derived from thermal radiance and SO_2 flux trends: the green area represents a phase of minor thermal energy and passive degassing from summit crater; yellow areas represent phases of moderate thermal energy and weak and/or short-lived lava emissions; red areas represent high thermal flux periods associated with more vigorous eruptive episodes. Orange areas represent transition phases preceding these vigorous high-thermal phases, characterized by high SO_2 fluxes. Numbers represent the average trend between phases. a) Eruptive activity at all summit craters derived from INGV-OE monitoring reports. b) Thermal energy derived by cumulating the daily-averaged VRP values. c) SO_2 mass obtained by cumulating the SO_2 flux over the entire period (blue line) and excess SO_2 mass (black line) obtained removing the quiescent value of 1054 t/d. d) Cumulative number of paroxysms and number of monthly episodes in each phase of activity.

8.4.3 Volumetric magma budget across different eruptive phases

It has long been known that a general imbalance exists between the rates of magma degassing (input) and eruption (output), at Etna (Allard, 1997) and at other volcanoes globally (Shinohara, 2008). The added value in our study is that we can characterise this imbalance not only over

timescales (decades) characteristic of eruptive cycles (Allard et al., 2006), but also at the scale of individual eruptive phases, and keeping separated the different activity styles (e.g., inter-, pre-, syn- and post-eruptive periods).

We reported in Supplementary Figure S8.6 the cumulative volume of erupted (output) and degassed (input) magmas during the whole analysed period. The volumetric magma input and output rates for all the pre, syn and post periods of the thermal-derived eruptive phases (of Figure 8.4) are reported in Supplementary Figure S8.7. Results indicate that the input rates (Supplementary Figure S8.7a) are on average higher during the pre- and post-eruptive periods (averages of $2.58 \pm 0.85 \text{ m}^3/\text{s}$ and $2.91 \pm 0.96 \text{ m}^3/\text{s}$) than during inter-eruptive periods (average $2.20 \pm 0.73 \text{ m}^3/\text{s}$). The largest input rates are consistently observed during the syn-eruptive periods ($4.30 \pm 1.42 \text{ m}^3/\text{s}$), reflecting the same trends of Figure 8.5a. The output rates exhibit similar distributions, albeit with some distinctive features (Supplementary Figure S8.7b). The pre- and post-eruptive periods are characterised by very low output rates (average values of $0.01 \pm 0.00 \text{ m}^3/\text{s}$ and $0.04 \pm 0.01 \text{ m}^3/\text{s}$ respectively). In contrast, the output rates are 1 to 3 orders of magnitude larger during the syn-eruptive periods (average $2.54 \pm 0.78 \text{ m}^3/\text{s}$), when the main extrusion of lava takes place. This distribution confirms that the eruptive phases selected on the basis of thermal output (Figure 8.4 and Supplementary Figure S8.3) are well constrained, as the onset and cessation of lava extrusion closely match the duration of the syn-eruptive period. The inter-eruptive magma output rate is very low ($0.01 \pm 0.00 \text{ m}^3/\text{s}$, Supplementary Figure S8.7b), indicating that strombolian activity contributes little to the overall erupted magma budget.

This exercise shows that, during inter-eruptive periods (passive degassing $\pm$ mild strombolian activity), only a very minor fraction ($\sim 0.5\% = 100 \times 0.01/2.20$) of the degassed magma is ultimately erupted, implying long-term endogenous volcano growth and deformation. As the magma input rate increases (to $\gg 2 \text{ m}^3/\text{s}$), the magmatic system becomes increasingly unstable, with magma over-pressure build-up ultimately driving eruption (Aiuppa et al., 2023; Paonita et al., 2021). As eruption starts, the fraction of erupted magma (relative to degassing magma) then increases by more than an order of magnitude ($\sim 60\% = 100 \times 2.54/4.30$).

The same magma imbalance can be demonstrated (Figure 8.7) by analysing the cumulative volumes of degassed (input) and erupted (output) magma associated to the subpopulations of inter-eruptive, pre-, syn- and post-eruptive periods (same as Figure 8.5). We find the largest imbalance during the inter-eruptive intervals (Figure 8.7), when lava extrusion from the craters is significantly reduced to the sporadic and mild Strombolian activity. In these periods, the volumetric budget is largely dominated by degassing of magma ($449 \pm 149 \text{ Mm}^3$), little of which is erupted ($1.33 \pm 0.32 \text{ Mm}^3$). The pre- and post-eruptive (15-days) periods show degassed volumes of $60 \pm 20 \text{ Mm}^3$ and $68 \pm 22 \text{ Mm}^3$ respectively, and eruptive volumes of $0.48 \pm 0.12 \text{ Mm}^3$ and $1.16 \pm 0.32 \text{ Mm}^3$, respectively.

Finally, during the syn-eruptive intervals, the erupted magma volume increases significantly ($\sim 121 \pm 33 \text{ Mm}^3$), but still remains lower than the degassing magma volume ($\sim 213 \pm 70 \text{ Mm}^3$).

We conclude that magma imbalance is common to any of the Etna's activity states, including during eruption.

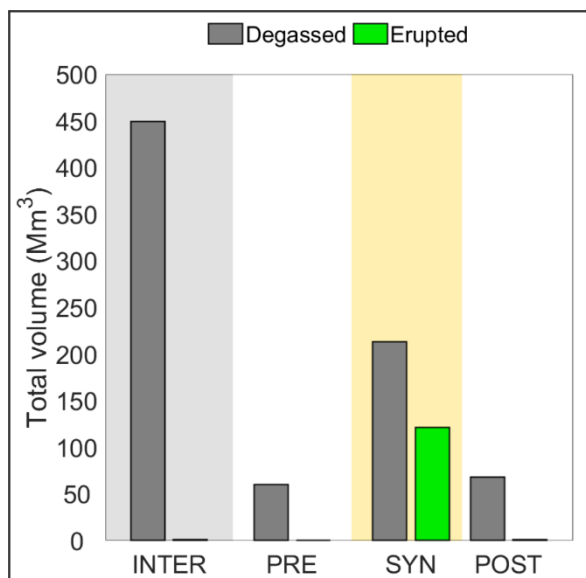


Figure 8.7 - Total degassed and erupted magma volumes obtained by cumulating the daily input and output magma rates during the inter-event and pre-, syn- and post-eruptive periods.

8.4.4 Tracking volume changes in the magma reservoir

The cumulative volumes of erupted (output) and degassed (input) magma during late-2015 to 2024 are respectively $124 \pm 34 \text{ Mm}^3$ and $781 \pm 261 \text{ Mm}^3$ (Supplementary Figure S8.6b,c). If we subtract a baseline (background activity) magma input/degassing rate of $1.18 \text{ m}^3/\text{s}$, corresponding to the quiescent phase of mid-2017-2018 (green shaded area in Figure 8.6 and Supplementary Figure S8.6, when both thermal and SO_2 fluxes were at their lowest), the magma input volume reduces to $458 \pm 150 \text{ Mm}^3$ (black solid line in Supplementary Figure S8.6c). This “eruptible” magma volume (see Laiolo et al., 2022) corresponds to the volume of magma in excess to that sustaining background (baseline) activity (as exemplified by the quiescent phase of mid-2017-2018), and that is hence available to sustain any eruptive activity. With these numbers, we find that only 27% of the input magma volume is ultimately erupted, with the remaining magma fraction ($\sim 73\%$) remaining stored/stalled within the plumbing system. For comparison, Palano et al. (2024) inferred the magma eruption efficiency during 2001-2022 to be $\sim 29\%$.

The difference between the eruptible magma volume ($458 \pm 150 \text{ Mm}^3$; calculated above) and the erupted magma volume ($124 \pm 34 \text{ Mm}^3$; from thermal records) corresponds to the “excess” magma volume, e.g., the volume that would have accumulated within (or being withdrawn from) the Etna's

magma plumbing system. This “excess” magma volume, shown in Figure 8.8a, reveals a steady increase, implying that magma accumulation prevails over magma withdrawal (e.g., that more magma enters the system than it is emitted during eruption). The “excess” magma volume reaches $\sim 120 \text{ Mm}^3$ at the onset of the 2021 eruption, at which point an inversion in this trend is observed: the negative slope of the excess curve Etna implies eruption of more magma than being supplied to the plumbing system, causing a net mass deficit.

Interestingly, this excess magma budget trend correlates with volcano geodesy evidence for deformation of the volcanic edifice, as inferred from the elevation changes measured by the GPS network (Palano et al., 2024) (Figure 8.8b). This shows that, between 2016 and the start of the 2021 paroxysmal sequences, the edifice underwent a net uplift of $\sim 60 \text{ mm}$, in general agreement with the volumetric accumulation trend inferred from our SO_2 -thermal budget (Figure 8.8a). The 2021 February to April period of high-clustered paroxysmal sequence (Marchese et al., 2021; Amadio et al., 2024; Phase S₁ in Carleo et al., 2023) (F1 in Figure 8.8) produced both a reversal in the elevation trend ($\sim 25 \text{ mm}$ of subsidence) and in the magma budget curve, corresponding to a withdrawal of $\sim 11 \text{ Mm}^3$, which matches closely the geodetic volumetric estimates for the event ($\sim 12 \text{ Mm}^3$, Palano et al., 2024). According to Palano et al. (2024), these deformation changes can be explained by a deflating source located at $\sim 8 \text{ km}$ depth. After a short period of quiescence (~ 2 months), Etna experienced another second intense phase of high-clustered lava fountains (F2 in Figure 8.8) from May to August 2021 (Phase S₂ in Carleo et al., 2023). Nicely, our observations reveal that this second energetic eruptive period produced a further subsidence of the volcano edifice of $\sim 21 \text{ mm}$, which corresponds to a new withdrawal of $\sim 9 \text{ Mm}^3$. After this period, the occurrence of more sporadic lava fountains interspersed by sustained passive degassing, produced a less pronounced subsidence of the volcano, resuming an excess volume accumulation due to the predominance of inter-event degassing phases. This correspondence underscores the value of the combined thermal-gas approach in identifying endogenous accumulation processes of eruptible magma in volcanic systems, with clear implications for hazard assessment.

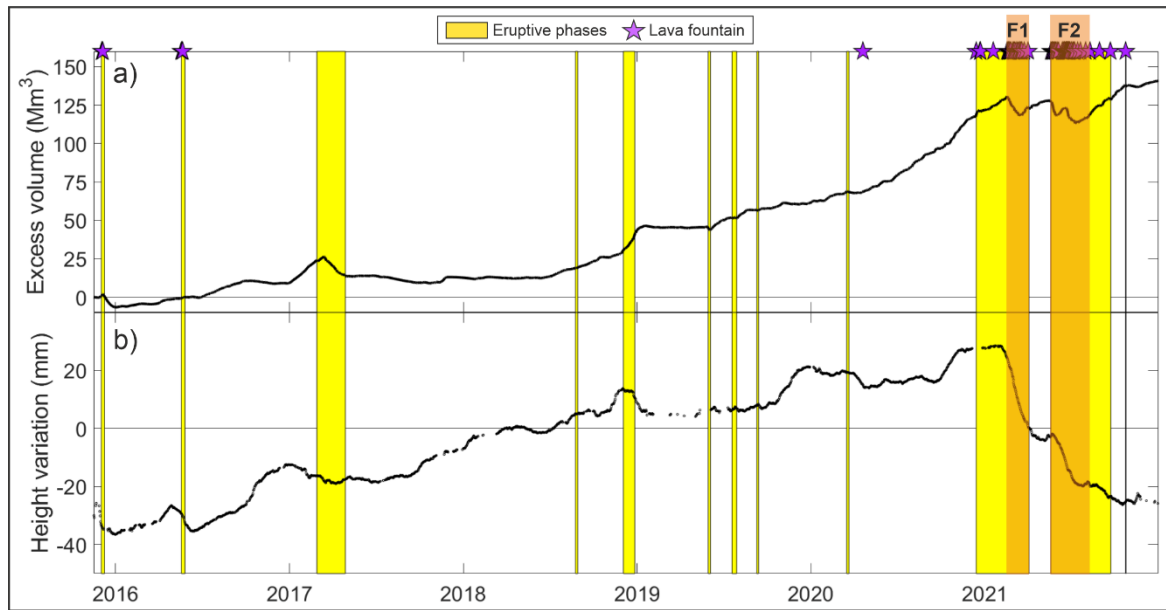


Figure 8.8 – a) Magma accumulation within Etna’s shallow plumbing system during the late-2015 to 2021 period. This excess volume has been obtained by removing from the magma input rate the fixed recycling rate of $1.18 \text{ m}^3/\text{s}$ and the magma output rate. b) Height variations of the volcanic edifice measured by the GPS network (data from Palano et al., 2024). Yellow areas represent the main eruptive phases obtained from thermal radiance (same as Figure 8.4), while orange portions represent the more intense phase of clustered lava fountain activity (see Carleo et al., 2023).

8.4.5 A valuable monitoring tool – with caveats and limitations

We have provided evidence for the utility of a combined SO_2 -thermal approach to understand Etna’s behaviour, and to track (and potentially anticipate) the quiescence-to-eruption transition. This combined approach is also relevant to understanding the volcano magma budget (Cfr. 8.4.3 and 8.4.4). Our analysis, however, also demonstrates caveats and limits of the UV Camera-derived SO_2 flux time series as a volcano monitoring tool.

We have shown that the UV Camera-derived SO_2 flux records can resolve *some*, but not *all*, the Etna’s eruptive phases (Figure 8.5). The eruption detection efficiency of our SO_2 records scales with eruption size (Figure 8.5c). This means that while major swings in volcanic activity can be effectively tracked (and potentially anticipated; see also Figure 8.6 and related discussion on “orange” periods), SO_2 is not able to identify *any* change in Etna’s activity state, especially those associated with small ($< 1 \text{ Mm}^3$) erupted volumes (Figure 8.5c). This limit may not only apply to UV Camera records, but to SO_2 flux time series in general. In particular, the relatively large Etna’s background SO_2 emissions during quiescent periods, make the detection of subtle perturbations in the magma feeding system challenging.

Some caveats and limits are characteristic of the UV Camera technique, however. For example, our camera system is blind to any degassing/eruption process occurring in the central/norther portion of the crater terrace; hence, any eruption at NEC and (partially) at VOR would remain undetected.

Given the preferential clustering of volcanic activity at SEC during late-2015 to 2024 (Figure 8.1a), this is unlikely to be severe limit in our specific case. More difficult to evaluate is the impact of processes related to radiative transfer issues. SO₂ fluxes derived from UV Cameras are affected by multiple factors, including multiple scattering in the plume and in the background atmosphere, light dilution, in-plume condensation processes, and changes in atmospheric conditions and wind pattern, all of which do introduce significant uncertainties and lead to potential underestimation of SO₂ (Kern et al., 2010a, 2010b; Kern et al., 2013; Varnam et al., 2021; Delle Donne et al., 2022; Lo Bue Trisciuzzi et al., 2024). These problems are exacerbated by the use of a single high-elevation station, where plume geometry and atmospheric conditions can strongly influence the acquired signal, especially in a multi-vent and high-elevation volcano like Mount Etna. In our specific case, the use of operator-reviewed images can mitigate against errors associated with multiple scattering, by selecting frames associated to the best viewing conditions, such as ash-poor plumes or favourable (cloud-free) meteorological settings. Nevertheless, this operation addresses only a limited fraction of the uncertainties, while additional processes, such as in-plume condensation/scattering and light dilution, may still significantly affect the measurements. Moreover, under strong-wind conditions, the complex morphology of the SEC ridge can cause the UV Camera-imaged plume to represent only part of the degassing process rather than its full extent. For these reasons, evaluating the SO₂ flux dynamics using a single UV Camera station remains a complex task, particularly when comparing different eruptive phenomena in terms of source characteristics and duration.

With these limitations in mind, the ability of the UV Camera system to efficiently resolve the eruptive degassing escalation from the quiescent level (Figures 8.5, 8.6) proves its utility as volcanic gas monitoring tool.

8.5 Conclusions

This study has provided a nearly decade-long synthesis of the degassing and thermal dynamics across the main eruptive phases of Mt. Etna, obtained through the joint analysis of SO₂ fluxes from a permanent UV Camera system and satellite-derived thermal radiance data.

We have shown that the thermal and SO₂ fluxes vary coherently across the different volcanic activity states. By analysing the SO₂ and thermal records in proximity of 18 major eruptive phases from November 2015 to 2024, accounting for >90% of the erupted lava volume during the decade, we have inferred a characteristic progression of increasing SO₂ fluxes, from 1957 ± 1470 t/d during the inter-eruptive periods, to 2302 ± 1481 t/d in the (15 day-long) pre-eruptive periods, and up to 3828 ± 2686 t/d during eruption. The SO₂ fluxes then decay to 2595 ± 1929 t/d in the post-eruptive period. Likewise, the transition from quiescence activity to more energetic behaviour is associated

with thermal fluxes >0.3 GW, while the most energetic eruptive episodes typically radiating more than 1 GW.

We tested the ability of the UV Camera-derived SO_2 fluxes to resolve the different Etna's activity states, finding detection efficiencies of 61%, $\sim 72\%$ and 72% for the pre-, syn- and post-eruptive periods. Performance scales with eruption magnitude, implying that larger eruptions are associated to major swings in activity that can be more effectively captured (and frequently anticipated by several days) in the SO_2 record, while less voluminous eruptive phases more frequently escape from detection. This analysis reveals benefits and applicability limits of UV Cameras as monitoring tools.

We have also resolved the magma input (degassing) and output (eruption) volumetric fluxes associated with the inter-, pre-, syn- and post-eruptive periods. Results indicate that eruptive phases are initiated by magma input rates increasing well above the inter-eruptive average of $2.20 \pm 0.73 \text{ m}^3/\text{s}$. Our volumetric analysis confirms that the significant imbalance between degassed and erupted magma extends across all activity phases, and is highest during inter-eruptive periods ($\sim 0.5\%$ of degassing magma being erupted) and lowest during eruption ($\sim 60\%$ of degassing magma being erupted). Over a decade of activity, the erupted magma fraction is $\sim 27\%$, with the residual fraction of unerupted magma ($\sim 120 \text{ Mm}^3$ from 2016 at the onset of the 2021 large lava fountain sequences) being stored in the plumbing system and contributing to volcano deformation.

The in-tandem analysis of SO_2 and thermal fluxes data can contribute to deeper understanding of gas and magma budget at open-vent volcanoes globally.

SUPPLEMENTARY MATERIAL

Start	End	Duration (days)	Energy (Joules)	Volume (m ³)	Max rate (m ³ /s)	Mean rate (m ³ /s)
02/12/2015	09/12/2015	7	2.06E+10	2.20E+06	8.10	3.65
18/05/2016	26/05/2016	8	9.11E+09	9.89E+05	5.96	1.43
27/02/2017	28/04/2017	60	1.18E+11	1.41E+07	10.11	2.73
24/08/2018	29/08/2018	5	4.02E+09	4.09E+05	1.78	0.95
03/12/2018	27/12/2018	24	1.26E+10	1.47E+06	9.12	0.71
30/05/2019	04/06/2019	5	1.90E+10	1.94E+06	13.64	4.48
19/07/2019	29/07/2019	10	1.20E+10	1.33E+06	6.35	1.54
09/09/2019	13/09/2019	4	3.05E+09	2.98E+05	1.99	0.86
16/03/2020	21/03/2020	5	2.95E+09	3.01E+05	1.33	0.70
13/12/2020	03/04/2021	111	1.06E+11	3.30E+07	26.98	3.45
19/05/2021	22/09/2021	126	9.93E+10	4.50E+07	27.36	4.13
23/10/2021	24/10/2021	1	2.78E+09	7.15E+05	16.56	8.28
21/01/2022	22/02/2022	32	7.66E+09	9.07E+05	4.04	0.33
14/05/2022	13/06/2022	30	6.98E+10	8.25E+06	6.47	3.18
02/12/2022	05/02/2023	65	4.31E+10	5.19E+06	2.90	0.92
13/08/2023	15/08/2023	2	2.18E+09	1.78E+05	2.42	1.03
12/11/2023	26/11/2023	14	3.23E+09	3.68E+05	1.77	0.30
30/06/2024	16/08/2024	47	3.50E+10	5.78E+06	14.66	1.42

Table S8.1 – Table of eruptive phases inferred from thermal radiance. Each phase is reported with its start and end dates, duration, thermal energy, involved magma volumes and its max and mean magma output rate.

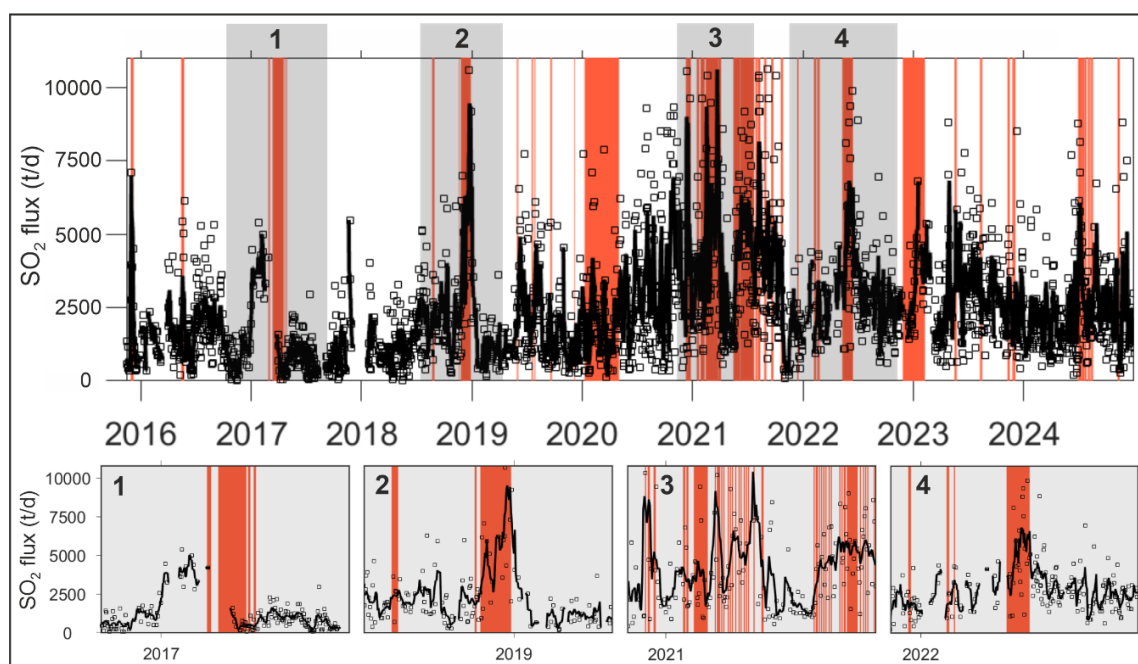


Figure S8.1 – Detailed view of daily-averaged SO₂ fluxes, showing the escalation before, during, and after some lava effusion phases (red bars).

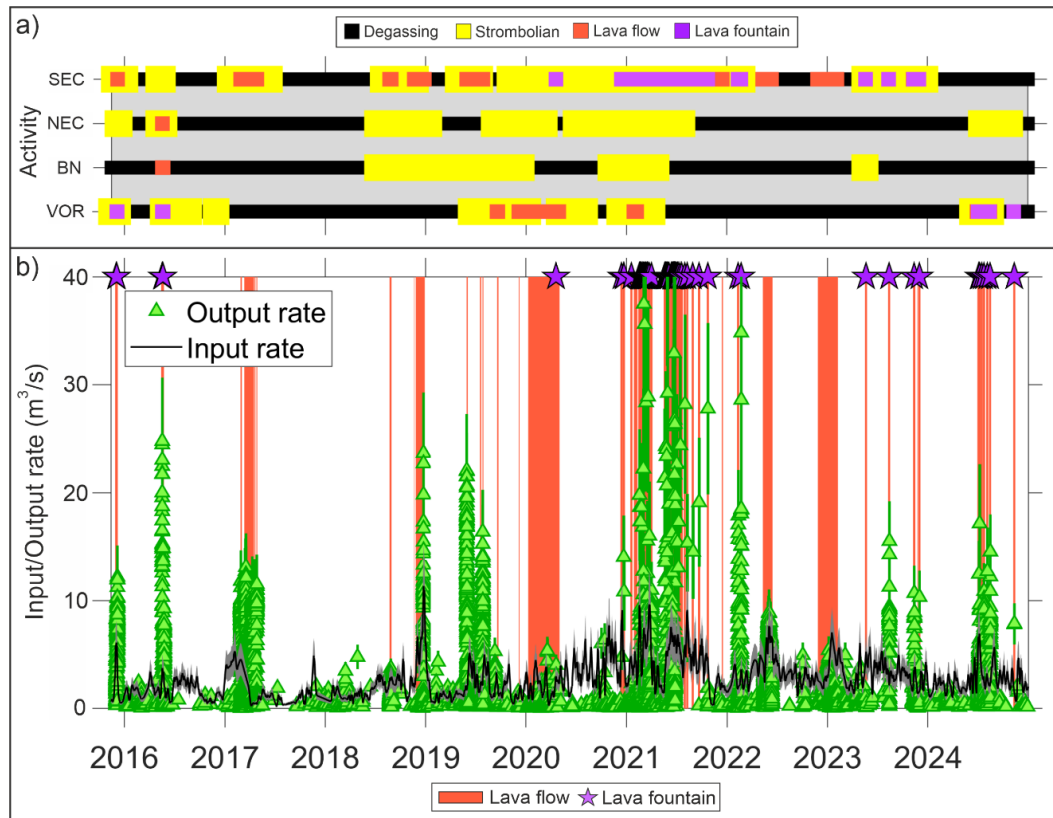


Figure S8.2 - Magma input and output rates derived from SO_2 flux measurements and thermal data. The intermediate scenario of the magma input rate (see Paragraph 8.2.3 in main text) is represented as a black solid line (7-day moving average), while the minimum and maximum scenarios are encompassed within the grey area. The triangle-marked green line shows the intermediate scenario of the magma output rate (see Paragraph 8.2.4 in main text), while the green vertical bars identify the minimum and maximum scenarios.

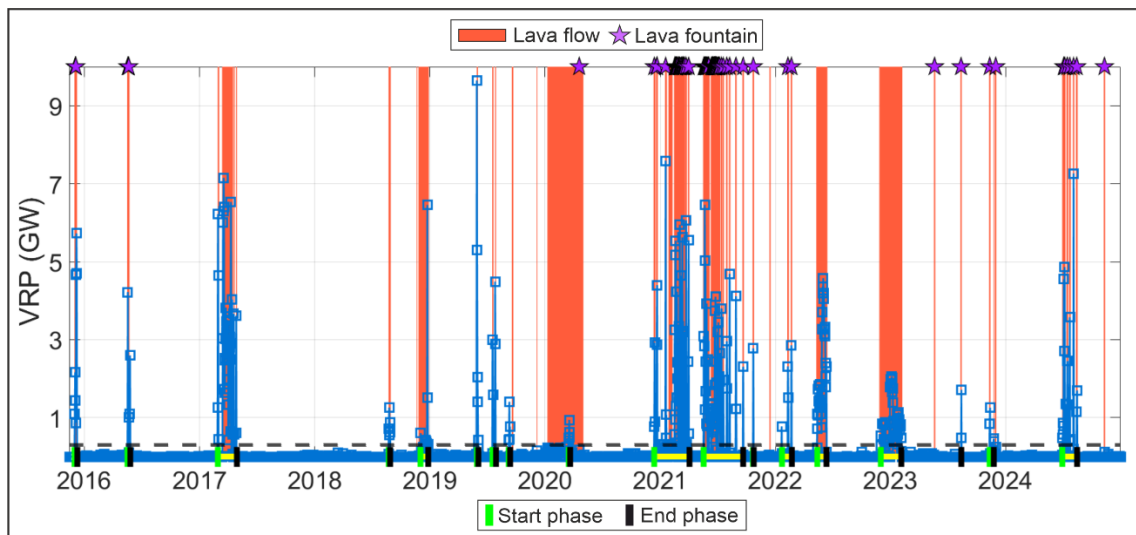


Figure S8.3 – Main eruptive phases identified taking as eruption all the daily-averaged VRP values above the threshold of 0.3 GW (black dashed line). The onset of an eruptive phase (green bars) is fixed at the first positive detection (VRP above the threshold). If others positive detections occur within the following 30 days, they are merged into a unique eruptive phase (yellow bars). The end of an eruptive phase (black bars) is fixed at the last day with VRP above the threshold, once no positive detections are encountered in the following 30 days. The 0.3 GW value can be considered as a minimum threshold, since the distribution of daily-averaged VRP shows eruptive peaks consistently above 1 GW.

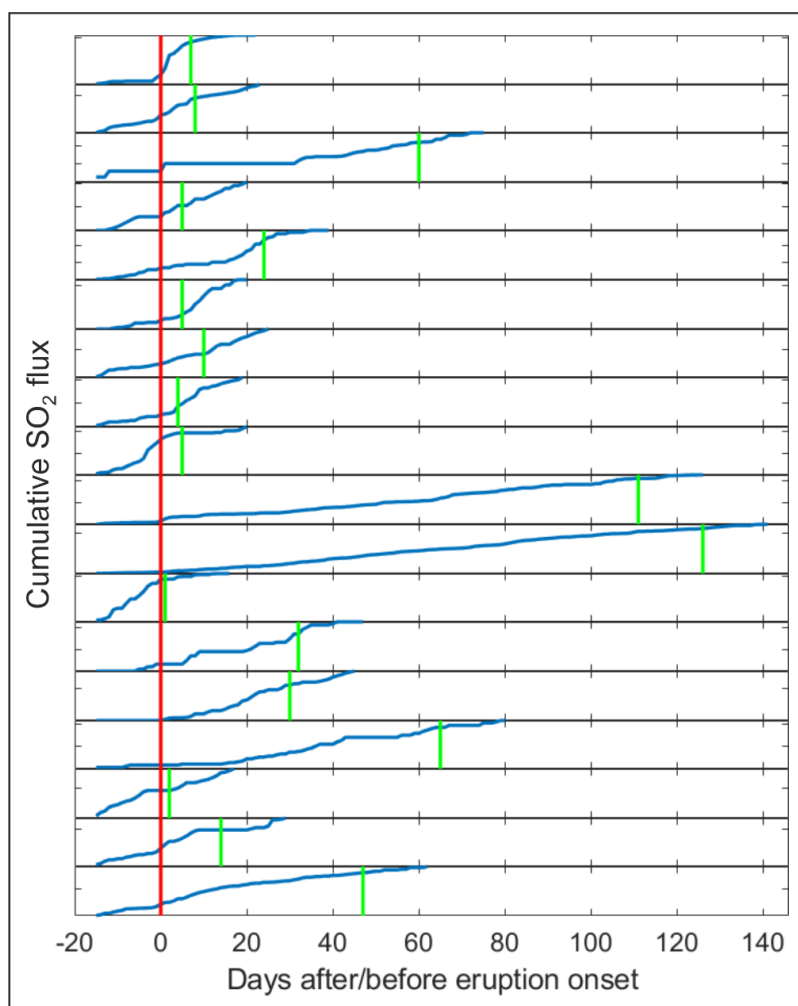


Figure S8.4 – Cumulative SO_2 degassing trends spanning 15 days before onset and 15 days after the end of all thermal-derived eruptive phases, aligned to the eruption onset (red bar at day 0). The green vertical bar marks the end of the eruptive phase and the transition from syn-eruptive to post-eruptive periods. It is visible a degassing signal accompanying almost every eruption, although its intensity is largely variable.

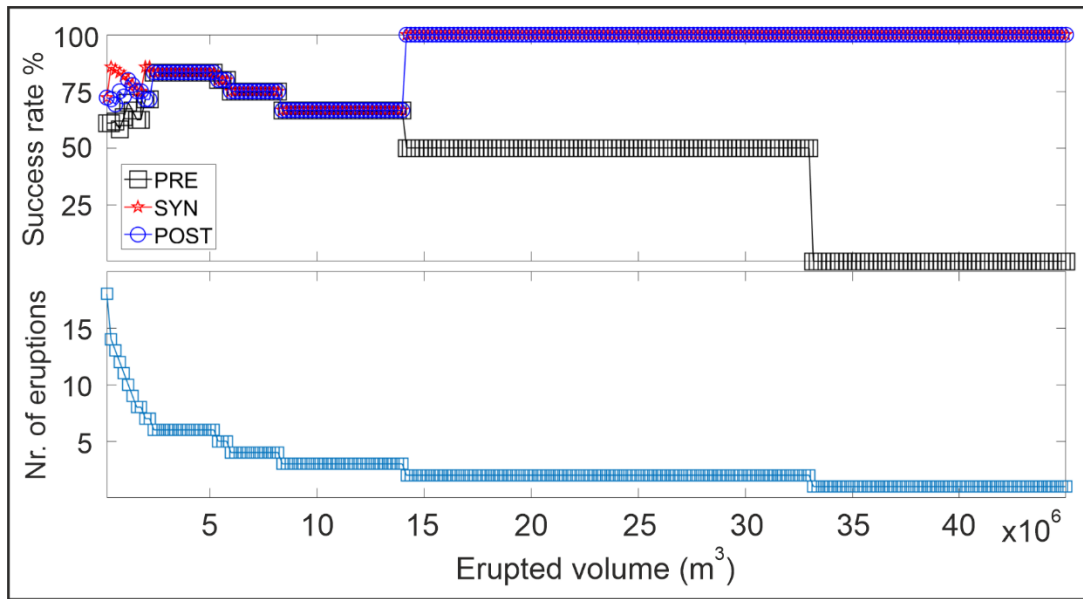


Figure S8.5 - Success rate in function of erupted volumes. The success rate increases progressively for increasing volumes up to 5 Mm^3 . Extending this analysis toward higher voluminous and less abundant eruptive phases we found that for eruptive volumes between 5 and 14 Mm^3 , the success rate gently declines but remains above 65%. For volumes exceeding 14 Mm^3 , only two eruptive phases are recorded (see also Table S8.1), which encompass the two large paroxysmal sequences of 2021. In these cases, the success rate reaches 100% for the syn- and post-eruptive periods, but drops during the pre-eruptive period. This (statistically low) decrease must be considered in the view of large eruptive events dynamics, whose degassing pre-eruptive patterns is visible on larger timescales (Aiuppa et al., 2023; Paonita et al., 2021; Allard et al., 2002) than our analysed window (15 days). Furthermore, these values are obviously influenced by the second paroxysmal sequence of 2021, which was preceded by a prolonged phase of quiescent degassing (Figure 8.2b, main text), with values falling below background level.

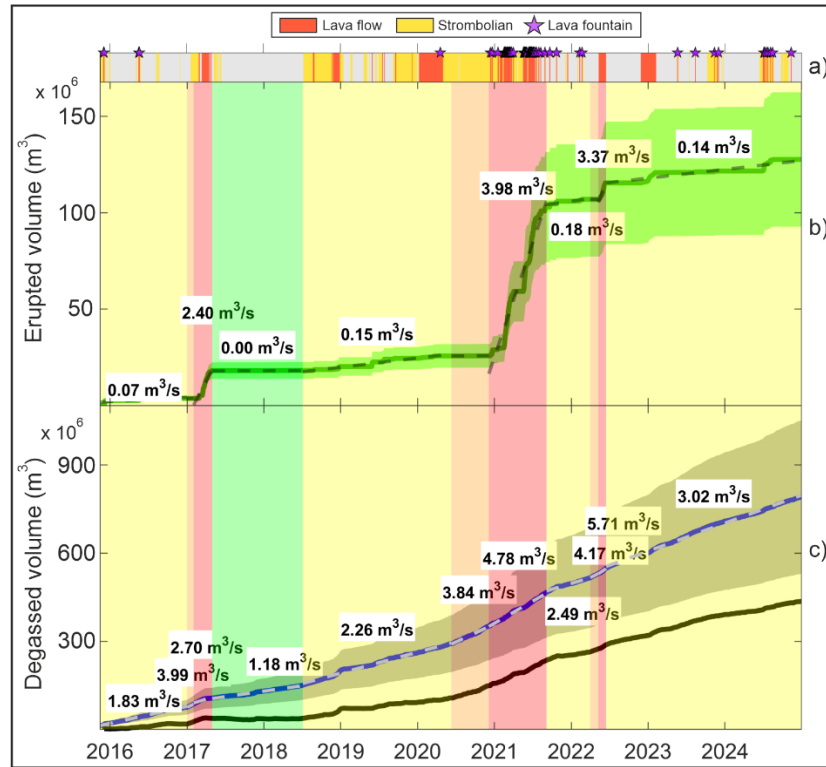


Figure S8.6 - Cumulative volume of erupted (output) and degassed (input) magmas during the analysed period, together with their time-averaged values during the main phases of volcanic activity inferred from thermal and SO_2 trends (see Paragraph 8.4.2 in main text); a) Eruptive activity at all summit craters derived from INGV-OE monitoring reports. b) Erupted magma volume derived by cumulating the magma output rates. c) Degassed magma volume (blue line) obtained by cumulating the magma input rates. The black solid line represents the “eruptible” magma volume (cfr. 8.4.4), obtained by subtracting the baseline magma input rate of $1.18 \text{ m}^3/\text{s}$ (green area).

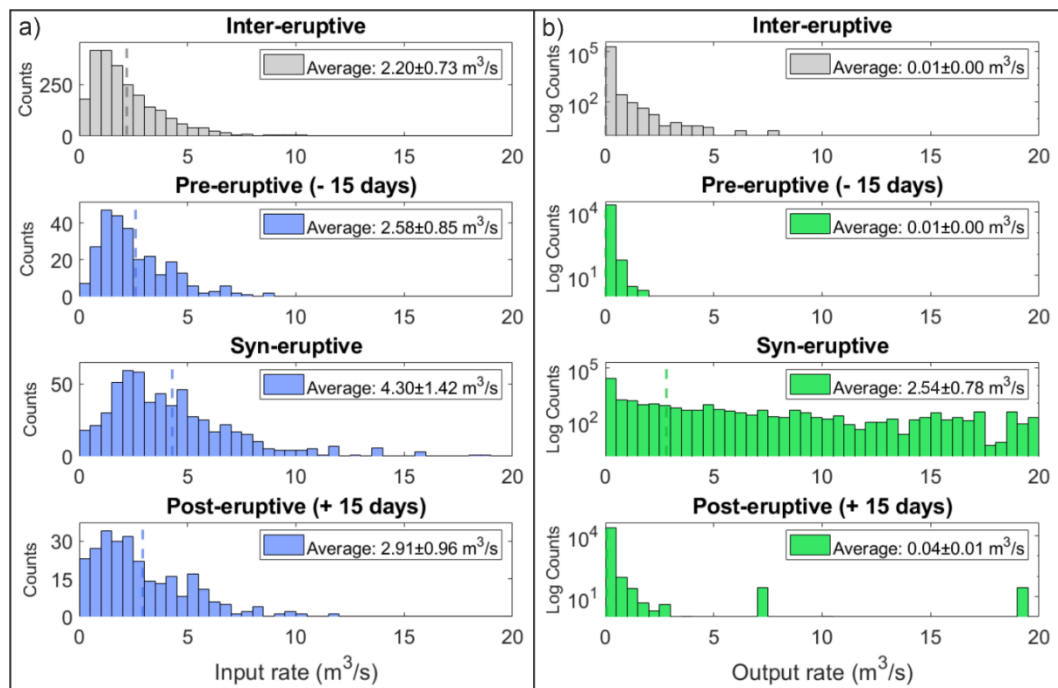


Figure S8.7 - Distribution of magma input (a) and output (b) rates during inter-event periods and pre-, syn-, and post-eruptive stages. The dashed lines show the mean value of the distribution.

CHAPTER 9

A SO₂ FLUX STUDY OF THE ETNA VOLCANO 2020-2021 PAROXYSMAL SEQUENCES

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Abstract

The persistent open-vent degassing of Mt. Etna is often punctuated by months-long paroxysmal sequences characterized by episodes of violent Strombolian to lava fountaining activity. Understanding these gas-fueled transitions from quiescence to eruption requires routine measurement of gas fluxes. Here, we report SO₂ flux measurements, obtained from a permanent UV Camera system, collected over a two-year-long period spanning two paroxysmal sequences of Etna's New South East Crater (NSEC) in December 2020/April 2021 and May/October 2021. In both cases, SO₂ flux increased from ≤ 3250 Mg/day during 'ordinary' activity to ≥ 4200 Mg/day. We interpret these distinct SO₂ degassing regimes in light of seismic and thermal observations and drawing on numerical simulations of sulfur degassing constrained by parental melt sulfur contents in Etna's hawaiites. We find that initiation of a paroxysmal sequence results from an approximate doubling of the time-averaged rate of magma supply (and degassing) above the sulfur exsolution level (~ 150 MPa pressure), to >4 m³/s. This corroborates recent models that argue for the triggering of paroxysmal sequences by escalating supply of volatile-rich magma to a reservoir $\sim 3\text{--}4$ km below the summit region. The non-stationary nature of magma flow and volcanic degassing we identify highlights the need for sustained surveillance to characterize long-term atmospheric budgets of volcanic volatiles.

9.1 Introduction

Magmatic degassing – the exsolution and separation of volatiles from melts – is a critical step in magmatic processes (Oppenheimer et al., 2014). The gas bubbles that form due to decompression/cooling upon crustal emplacement of magmas are the fuel of explosive volcanic eruptions (Gonnermann and Manga, 2003) and their release to the atmosphere can have local/regional (Mather, 2015) and global scale environmental impacts (e.g., during large-scale explosive eruptions; Robock, 2000).

This critical role gas plays in volcanic processes has motivated attempts to quantify gas emission rates from active volcanoes, with a focus on ground- and satellite-based techniques that target measurement of the more easily detectable volcanic SO₂ flux (Oppenheimer et al., 2011). However, as volcanic SO₂ flux catalogues become increasingly complete (Mori et al., 2013; Shinohara, 2013; Carn et al., 2017; Fischer et al., 2019; Arellano et al., 2021), the non-stationary nature of volcanic degassing emerges more and more clearly: even the so-called “persistent gas emitters” (Aiuppa et al., 2019) exhibit strong temporal fluctuations in emissivity, alternating periods of large SO₂ emissions with periods of reduced degassing (e.g., Miyakejima volcano: Kazahaya et al. (2004) vs. Mori et al. (2013); Bagana volcano: Carn et al. (2017) vs. McCormick Kilbride et al. (2019)), or even degassing shut-off (e.g., Ambrym volcano: Bani et al. (2009) and Allard et al. (2016)).

vs. Moussallam et al. (2021)). While this unsteady degassing behavior challenges attempts to derive robust volcanic gas output inventories (Werner et al., 2019), it also opens avenues for eruption forecast (Poland and Anderson, 2020), as periods of heightened degassing typically coincide with, and sometimes precede eruptions (e.g., Reath et al., 2019b). In any case, it is now well recognized that sustained observations are needed to characterize the fluctuating nature of volcanic degassing (Kern et al., 2022).

Mount Etna in Sicily has been the focus of some of the most sustained SO₂ flux monitoring from ground-based observations (Caltabiano et al., 2004; Salerno et al., 2009a). Operational monitoring of SO₂ flux dates back to 1987, with some intermittent measurements in 1975 to 1986 (Caltabiano et al., 2004). The long time series of measurements, supplemented by satellite observations (e.g., Queißer et al., 2019), have identified Etna as one of the strongest persistent volcanic gas emitters worldwide (Allard, 1997; Shinohara, 2013; Carn et al., 2017).

Etna's persistent degassing activity is typically sustained by quiescent (passive) gas discharge from shallow convecting magma (Allard, 1997), accompanied by mild (Strombolian) intra-crater explosive activity. This ordinary activity is frequently interrupted by effusive eruptions (Branca and Del Carlo, 2004, 2005; Andronico et al., 2005; Branca et al., 2011) and, periodically, by violent paroxysmal explosive activity (Andronico et al., 2021b). Since the 1980s, there has been an intensification of this latter explosive behavior, with periods of quiescent open-vent degassing at its summit craters being frequently interrupted by powerful paroxysmal sequences, typically lasting months, and consisting of clusters of violent, short-lived Strombolian to lava fountaining episodes (Andronico et al., 2021b and references therein). These paroxysmal events vary widely in style (from mild Strombolian to large-scale lava fountaining), intensity (with up to 14 km high eruptive columns; Corsaro et al., 2017), duration (from < 2 to >24 hours) and recurrence intervals (from hours to years). Most often, these events occur in clusters of more or less regularly spaced (~10 days repose interval on average) episodes (post-2000 sequences consisted of three to 64 episodes), now described as a paroxysmal sequence (Andronico et al., 2021b). Paroxysmal activity produces locally/regionally dispersed eruptive clouds (Poret et al., 2018) that cause tephra fallout on the inhabited flanks of the volcano. Tephra deposition results in a variety of hazards and impacts (Andronico et al., 2015; Osman et al., 2019), and in substantial economic losses (Barnard, 2004). This increasingly explosive nature of Etna's volcanism has been related to a more volatile-rich, potassic magma feeding its transcrustal plumbing system (Métrich et al., 2004; Corsaro et al., 2009).

Although these paroxysmal sequences have at times involved multiple craters simultaneously (e.g., Corsaro et al., 2017), most since 2011 have been focused at the New South East Crater (NSEC) (Figure 9.1; Behncke et al., 2014; De Beni et al., 2015). Because all the summit craters can degas

simultaneously, SO₂ sensing tools able to resolve emissions from distinct vents/craters are very useful for understanding the volcano's behavior. This capability is afforded by a recently deployed permanent UV Camera system (D'Aleo et al., 2016, 2019; Delle Donne et al., 2019) (Figure 9.1).

Here, we report on a UV Camera-based SO₂ flux time series spanning two years during capturing two paroxysmal sequences of Etna's NSEC in December 2020/April 2021 and May/October 2021 (Andronico et al., 2021b; INGV-OE, 2021). Our aims are to: (i) constrain the diverse SO₂ flux behavior of ordinary vs. paroxysmal eruptive periods, and (ii) use the results to improve understanding of the trigger(s) of Etna's paroxysms.

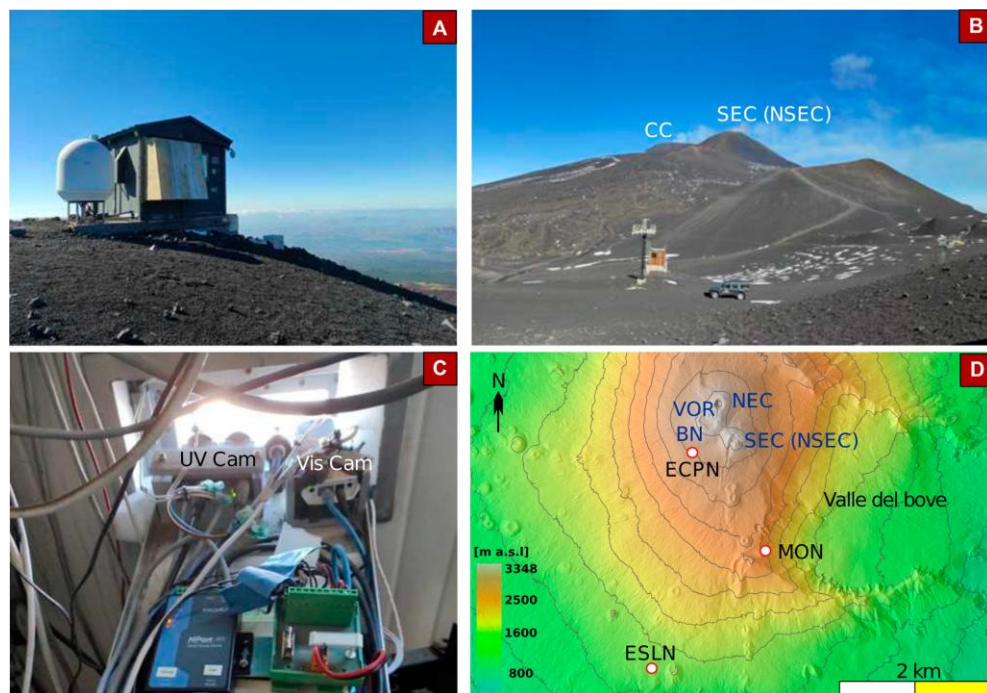


Figure 9.1 – (A) The INGV-OE Montagnola hut; (B) The summit crater terrace as seen from the Montagnola; (C) The UV Camera system inside the Montagnola (MON) hut: UV Cam: UV Camera; Vis Cam: Visible Camera; (D) Google Earth map of the Etna's summit region, showing the location of the seismic (ESLN and ECPN) and UV Camera (MON) instruments.

9.2 Materials and methods

9.2.1 UV Camera measurements

The SO₂ flux results we report on are derived from processing of images taken by the permanent “La Montagnola” (MON) UV Camera system (Figure 9.1). Hardware/software characteristics and the system's functionalities have been described (D'Aleo et al., 2016, 2019; Delle Donne et al., 2019). In brief, the system employs two JAI CM-140GE-UV cameras sensitive to UV-radiation, fitted with two different band-pass optical filters (Full Width at Half Maximum, FWHM, of 10 nm) with central wavelengths of 310 and 330 nm, respectively. The two co-aligned cameras are programmed to record daily sequences of images (520×676 pixel at 10-bit) at 0.5 Hz rate during optimal illumination

conditions (e.g., from 7.30 AM to 12.30 PM UTC time). Image acquisition is automated by a Fujitsu RX100 Workstation.

The daily images are post-processed by a standard protocol (Kantzas et al., 2010; Delle Donne et al., 2019), in which sets of simultaneously acquired images (from the two cameras) are combined into single absorbance images, applying the Lambert-Beer law. This allows quantification of the relative absorption of scattered UV sunlight by in-plume volcanic SO₂ over the cameras' field of view (FoV). Sequences of SO₂ absorption images are then converted into SO₂ column density images using the proportionality ratio between absorbance and SO₂ column density, derived from an Ocean Optics USB2000+ spectrometer pointing in a defined subregion of an absorbance image (Figure 9.1). Finally, SO₂ column densities are integrated along a profile stretching over the median portion of a rectangular integration sub-area, positioned over the crater terrace (Figure 9.1). The obtained SO₂ integrated column amounts ($\text{kg}\times\text{m}^{-1}$), once multiplied by plume speed ($\text{m}\times\text{s}^{-1}$; derived by applying an optical flow algorithm that tracks gas fronts in consecutive image frames; Delle Donne et al., 2019), yield a 0.5 Hz time step for SO₂ flux ($\text{kg}\times\text{s}^{-1}$) measurement, examples of which are shown in Figure 9.2a-c.

The entire operation sequence described above is completed using a MATLAB code that can deliver SO₂ flux time series in near real-time (Delle Donne et al., 2019). The results reported here (Figures 9.2, 9.3) are, however, based on post-hoc manual operator check and validation of these automatically processed data. In particular, the image sequences are visually inspected, and any frame of limited (or no) visibility (cloud coverage), or any frame (e.g., C4 and C6 in Figure 9.2 discussed below) with visible ash in the plume (as indicated by a co-exposed visible camera), are omitted from calculation of SO₂ flux daily averages (illustrated in Figure 9.3b). This is because the presence of ash in the plume is known to significantly interfere (suppress) the in-plume SO₂ signal (e.g., frames C3 and C5 in Figure 9.2 discussed below), resulting in artificially low SO₂ fluxes. The implication is that our observations severely underestimate syn-eruptive SO₂ emissions during a paroxysm's climax, which can be more fully characterized from spaceborne instruments (Corradini et al., 2021). However, as paroxysmal episodes are relatively short-lived (minutes to hours), the under-represented paroxysmal SO₂ contribution is relatively marginal (D'Aleo et al., 2019) if long-term (days to years) SO₂ degassing trends are of interest, as it is the case here.

Artificial suppression of the real SO₂ flux signal by ash is not the only disadvantage of UV Cameras compared with other more established measurement techniques, e.g., Differential Optical Absorption Spectroscopy (DOAS; Platt and Stutz, 2008; Galle et al., 2003, 2010). Targeting the proximal – and therefore often most optical opaque – portion of a plume (Figure 9.2), UV Cameras are more prone to errors caused by in-plume multiple scattering caused by aerosols and water droplets

(Kern et al., 2010a). We minimize this effect by selecting (from operator inspection of visible camera images) frames with visually transparent (uncondensed) plumes. More challenging is to account for the effect of radiation scattering (light dilution) in the line of sight between the volcanic plume and the UV Camera (Kern et al., 2010a, 2010b). This can attenuate the SO₂ signal, leading to significant underestimation of SO₂ column amounts, depending on the distance between the observation site and the plume. To estimate random and systematic errors in SO₂ column amount retrievals caused by light dilution, we apply the methodology of Campion et al. (2015) assuming an observation distance of 3 km (the distance between MON and Etna's NSEC crater). For the Etna's specific conditions, the methodology of Campion et al. (2015) allows derivation of a linear regression between measured and theoretical true scan column densities with a correlation coefficient (R^2) of 0.99, and a slope of ~0.68. From this regression, we estimate a potential systematic underestimation of the true SO₂ column densities of ~32%, and an average random error associated with each measurement of ~20%.

The UV Camera FoV (Figure 9.1) also limits the range of detectable gas emissions to those produced by the South East Crater (SEC) complex (that includes the NSEC) and from the southern portion (Bocca Nuova) of the Central Craters (D'Aleo et al., 2019) while leaving undetected the emissions of the North East Crater (NEC) and Voragine (the VOR emissions can only be measured during vigorous explosive activity that produce high, sustained plumes; D'Aleo et al., 2019). Therefore, our SO₂ flux measurements are unrepresentative of the bulk (total) plume emissions that, on Etna, are regularly monitored by more distant observations with scanning UV spectrometers (Salerno et al., 2009a).

The daily averaged SO₂ fluxes are listed in Supplementary Table S9.1. The chronology of volcanic activity (see below), and the complete SO₂ flux time series, are illustrated in Figures 9.3a and 9.3b.

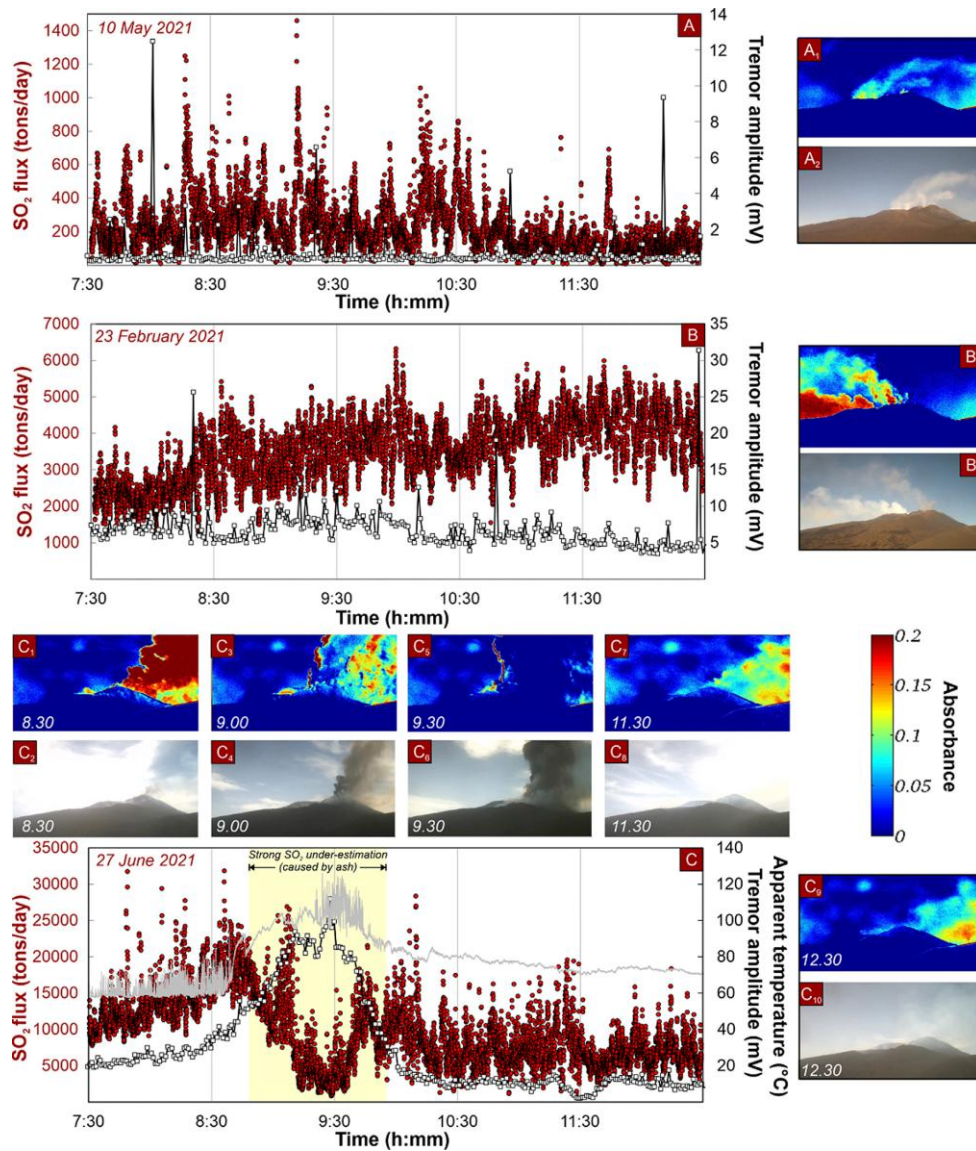


Figure 9.2 - SO_2 flux time series (red circles, 1 measurement every 2 seconds) obtained in three distinct measurement days (time as UTC time). (A) May 10, 2021, a measurement day characteristic of passive degassing during a quiescence period (QP1) (B) February 23, 2021, representative of passive degassing during a repose interval in-between two consecutive paroxysmal episodes of paroxysmal sequence PS1 (C) June 27, 2021, taken as example of measurements during a temporal interval encompassing a paroxysmal episode. In (A) to (C), seismic tremor RMS amplitude (open squares; data from INGV-OE ECPN broadband seismometer) is shown for comparison (as 1 minute average). Note different scales for both SO_2 flux and tremor in (A) to (C), demonstrating a progressive escalation in degassing and seismic activity. In (C), the apparent temperatures derived by processing (using the NewSaraterm software; Andò and Pecora, 2006) images captured by the ENT infrared camera are also shown (grey solid line). The SO_2 flux signal is abated by volcanic ash during peak of fountaining activity at circa 9.30 UTC time. In this phase (identified by yellow background), the low derived SO_2 fluxes are therefore a measurement artefact - this and other (two other measurement days in total) data subset affected by strong ash in the plume have been excluded from further processing (e.g., they do not concur to calculation of daily SO_2 flux averages illustrated in Figure and listed in Supplementary Table S9.1). Photos are absorbance and visible images of Etna's summit in the three specific days: May 10, 2021 (A1 and A2); February 23, 2021 (B1 and B2); June 27, 2021 (C1 to C10; taken at specific times as indicated). In the absorbance images, colour tones are proportion to in-plume SO_2 absorption of scattered UV sunlight (colour tone scale, from 0 to 0.2, shown in bottom right portion of the figure).

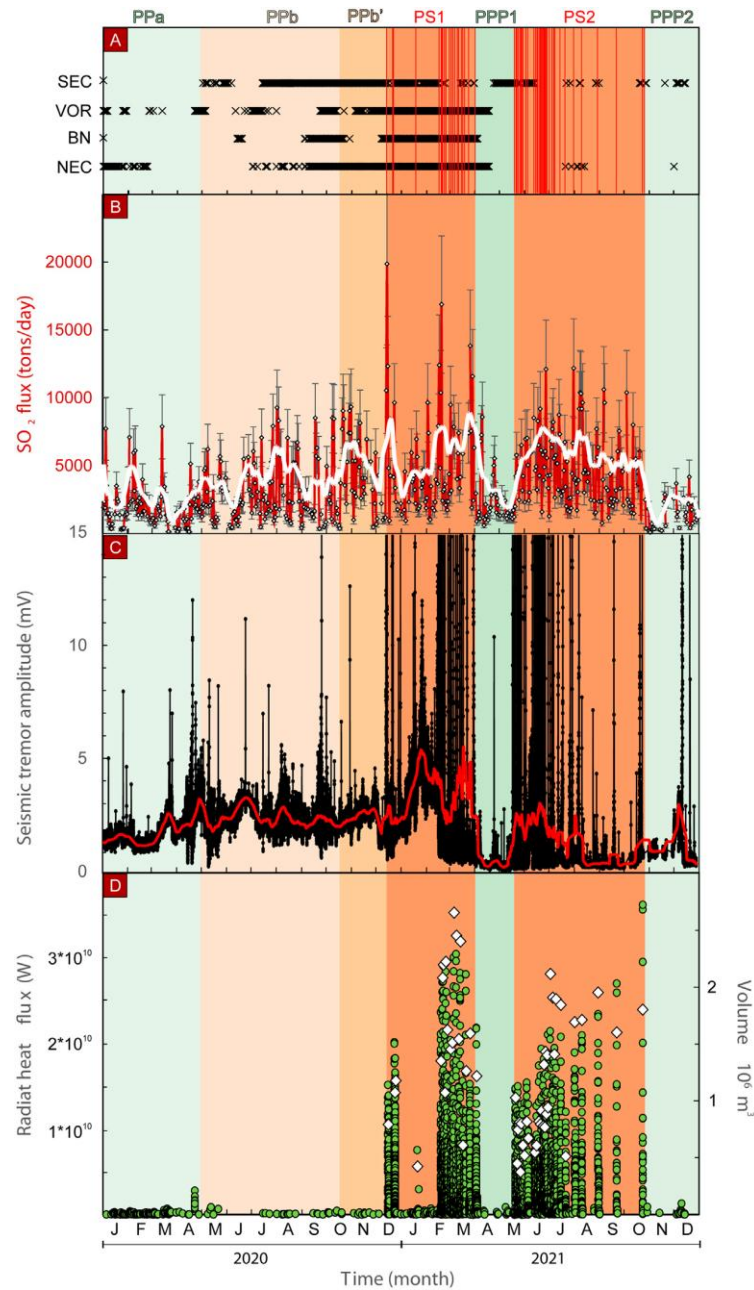


Figure 9.3 - (A) Chronogram of volcanic activity at Etna throughout 2020-2021. Crosses indicate mild explosive activity (either impulsive ash discharge events or intra-crateric Strombolian activity) at one of the summit craters (NEC: Northeast crater; VOR: Voragine crater; BN: Bocca Nuova crater; SEC: Southeast crater). Red vertical bars identify paroxysmal episodes at SEC (lava fountains during paroxysmal sequences PS1 and PS2; indicated by dark orange shading). During the precursory phase PPb, shading with different orange tones mark periods of progressive acceleration of magma transport (as evidenced by the derived magma input rates of Figure 9.4) and activity build-up at SEC. (B) Temporal record of daily averaged SO_2 fluxes (30% error bars are shown). The while solid line is a mobile average of the SO_2 flux maxima. (C) Minute-averaged seismic tremor RMS amplitude (black symbols and line), as measured at the INGV-OE ESLN broadband seismometer (a very similar temporal trend – yet shifted at higher values but with some temporal gaps in the winter season – is observed at ECPN seismometer; see Supplementary Figure S9.1). The red solid line is a 20-days mobile average drawn to illustrate the long-term (weeks/months) tremor fluctuations. (D) Time series of radiant heat flux (green circles), as derived from processing of MSG-SEVIRI multispectral images (one data every 15 minutes). The white diamonds indicate the erupted volumes (in 10^6 m^3) estimated from the radiant heat fluxes using the methodology of Ganci et al., (2012, 2013).

9.2.2 Volcanic tremor and radiant heat

To more fully characterize eruptive activity, the SO₂ flux record is complemented with seismic tremor data and thermal records from both satellite and ground-based instruments (Figures 9.2, 9.3).

Volcanic tremor is the most distinctive seismic signal recorded on active volcanoes. Generally associated with physical processes involving interactions between magmatic fluids and surrounding rocks inside the volcano (Chouet and Matoza, 2013), according to prevailing theory volcanic tremor originates from acoustic resonances of fluid-filled cracks triggered by pressure transients (Chouet, 1996). Volcanic tremor is often associated with the upward migration of magma and gases in the plumbing system (McNutt and Nishimura, 2008), and provides useful information on volcanic activity evolution (e.g., Almendros et al., 2001; Patané et al., 2013). At Mt. Etna, these seismovolcanic signals, and their variation over time, have turned out to be strictly related to eruptive activity (e.g., Patané et al., 2008; Viccaro et al., 2014), and have provided clues on pressurization or depressurization phenomena within the plumbing system (Cannata et al., 2015). Furthermore, the temporal evolution of volcanic tremor amplitude has been found to strongly correlate with the intensity of explosive activity (Andronico et al., 2021b) (compare Figures 9.2a, 9.2b, 9.2c). In fact, tremor amplitude gradually increases during the activity ramp-up that leads to a paroxysmal episode, peaking at the eruption climax, and then decreasing rapidly to return to a normal levels at the end of a paroxysmal episode (Alparone et al., 2003; Cannata et al., 2008) (Figure 9.2c). In order to track volcanic tremor amplitude changes, we calculate the Root-Mean-Square (RMS) of the seismic signal recorded by the vertical component of Serra La Nave seismic station (ESLN), located on the upper southern flank (Figure 9.1). This station is equipped with a broadband (40s cut-off period), 3-component Trillium Nanometrics™ seismometer, acquiring at a sampling rate of 100 Hz. The RMS time series is calculated on 1 min long time windows of seismic signal filtered in the band 0.5-5.5 Hz. This frequency band includes most of the volcanic tremor energy (e.g., Cannata et al., 2008). The entire RMS time series is illustrated in Figure 9.3c. The seismic station located at Cratere del Piano (ECPN), closer to the crater, exhibits a similar trend (Supplementary Figure S9.1).

Etna's paroxysmal episodes are also characterised by thermal observations from both ground and space. In order to derive high-rate information on thermal activity during a paroxysmal episode (data in Figure 9.2c), we use ground-based long-wave infrared (LWIR) records derived from a FLIR Thermovision A40M infrared camera located in Nicolosi, 15 km south of the summit craters (ENT). The camera is installed on the roof of a school in front of the INGV headquarters in Nicolosi. Thermal images are converted on board by the camera into a single output value that corresponds to the maximum temperature (Figure 9.2c) recorded in a region of interest, centred on the zone potentially

affected by eruptive activity. This temperature value is transmitted at 1 Hz, through a WiFi system, to the acquisition centre in Nicolosi, where thermal images are processed in real time by the NewSaraterm software (Andò and Pecora, 2006), developed with LabVIEW™ 8.0, to detect ash-rich eruptive columns, explosive and effusive activity. This software calculates, with a median filter, the daily thermal offset and extracts, for each measure, the difference between the detected value and the dynamic thermal offset. NewSaraterm also sends messages to the INGV-OE Control Room in Catania when a set of thresholds is exceeded, to alert on-duty staff for early warning procedures. The maximum apparent temperatures acquired by this camera are also published in real time on the INGV-OE Intranet page. The system has demonstrated its ability to recognize thermal anomalies over background. The temperatures displayed in the video frames are much lower than the true temperatures at the target owing to distance between the thermal camera site and the target (the summit craters), and the effect of environmental factors such as air humidity, pressure, air temperature, and the presence of protective lenses on the camera (Sawyer and Burton, 2006; Spampinato et al. 2011; Ball and Pinkerton, 2006). The temperature values illustrated in Figure 9.2c are thus apparent, not absolute, as they have not been corrected for the aforementioned factors. The chosen temperature range on the infrared camera is from -40°C to 120°C , and temperatures exceeding 130°C are not shown in Figure 9.2c.

Longer-term fluctuations in radiant heat (Figure 9.3d) are characterised using radiant heat flux derived from space-borne images acquired by the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) on board Meteosat Second Generation (MSG) satellite. MSG-SEVIRI multispectral images are retrieved via the CL-HOTSAT system, a new version of the HOTSAT system (Ganci et al., 2011; 2016, 2018b) implemented in the OpenCL standard to obtain near-real-time monitoring of volcano-related thermal activity. The SEVIRI sensor is especially useful to observe and track rapid eruptive events such as Etna's paroxysmal explosions (Calvari et al., 2021a; 2022a), since it allows the acquisition of images in the thermal and medium infrared region of the electromagnetic spectrum at very high temporal resolution (every 15 minutes; up to 5 minutes using the rapid scanning service at medium latitudes). The CL-HOTSAT system developed at INGV-OE allows for automatic detection of thermal anomalies and quantification of thermal activity through calculation of the radiant heat flux. The radiant heat flux time series in 2020-2021 is illustrated in Figure 9.3d.

In the case of effusive eruptions, the radiant heat flux can be converted into a time average discharge rate (TADR) (Harris et al., 1998) assuming a thermal balance between the heat within the erupted flow volume and that lost from it. This balance implies that (i) the volumetric flux is proportional to flow area, and (ii) lava surfaces cool exponentially with time and distance from the source (Harris and Baloga 2009). These assumptions cannot be verified in the case of transient events

such as Etnean paroxysmal events, during which steady-state, and hence thermal equilibrium, cannot be reached (Garel et al., 2013). However, for these short and transient eruptive events, a satellite-derived volume estimation can be derived from modelling the radiant heat flux curve measured during the cooling-dominated phase of activity (Ganci et al., 2012; 2013). Our satellite-derived radiant heat flux (Figure 9.3d) is converted into magma volumetric fluxes (Figure 9.3d) by modelling the time (t) required for lava flow to cool following equation:

$$t = \left\{ \left[\left(\frac{T_h - T_a}{\varepsilon \sigma (T_{surf}^4 - T_a^4) + h_c (T_{surf} - T_a)} - \frac{R_c R_r}{R_c + R_r} \right) k \right] 1000 \frac{1}{2\lambda} \right\}^2 \frac{1}{\kappa} \quad (\text{Eq. 9.1})$$

where T_{surf} is the surface temperature for a stagnant, stable, cooling lava surface, T_h is the initial temperature, T_a is ambient temperature, ε is emissivity, σ is the Stefan-Boltzmann constant, h_c is the convective heat transfer coefficient, k is thermal conductivity, λ is a dimensionless scaling value, κ is thermal diffusivity, and R_c and R_r are thermal resistance terms for convection and radiation, respectively.

Converting the surface temperature in radiant heat flux, hence including the radiating area, and fixing a minimum and maximum thickness expected for these short-lived events, an estimation of the emplaced volume can be obtained. Part of the thermal dataset used here is also discussed in Ganci et al., (2023).

9.3 Results

9.3.1 Volcanic activity

A chronology of volcanic activity during our observational interval (January 1 2020 to December 31 2021) is illustrated in Figure 9.3a, as derived from INGV-OE monitoring reports (INGV-OE, 2021). As evident, volcanic activity has taken a variety of forms during the study period, with the four summit craters being intermittently (and at time all concurrently) active. As our primary goal is to analyze degassing dynamics at NSEC (the most active crater, but also the one that can be most successfully observed from our UV Camera position, Figure 9.1), we classify volcanic activity based on the characteristics exhibited by activity at this specific crater. Based on this, we categorize our observational interval into seven categories (activity periods), as discussed below.

The Precursory Phase PPa (January-April 2020) is distinguished for the lack of any visible eruptive activity at the NSEC (Figure 9.3a). Volcanic activity is therefore confined at NEC and VOR that exhibit some relatively continuous intra-crateric explosive (ash expulsion and mild Strombolian events) and effusive (at VOR) activity.

The Precursory Phase (PPb) (May 2020 to December 12, 2020) marks resumption of volcanic activity at the NSEC. Activity progressively intensifies (becomes increasingly more frequent; Figure

9.3a) at the NSEC during PPb, but remains limited to mild Strombolian activity and intermittent ash expulsion events. No paroxysmal episode takes place in this phase. Sub-period PPb', the last 2 months of phase PPb (Figure 9.3a), is identified based on a visible acceleration in degassing activity at NSEC (based on the SO₂ flux record; see below).

Onset of paroxysmal activity on 13 December marks onset of PS1, the first of two especially powerful paroxysmal sequences (dark orange in Figure 9.3a) occurring in 2020-2021 (Andronico et al., 2021b; Bonaccorso et al., 2021; Calvari et al., 2021a; De Gori et al., 2021; INGV-OE, 2021; Marchese et al., 2021; Corsaro and Miraglia, 2022). PS1 is opened by 4 “transitional” (following the terminology of Andronico et al. (2021b): “strong Strombolian activity alternating with short periods of lava fountains”) eruptive episodes on 13, 21 and 22 December 2020, and on 18 January 2021 (Andronico et al., 2021b; Bonaccorso et al., 2021; Corsaro and Miraglia, 2022) (Figure 9.3a). Since 16 February 2021, paroxysmal episodes become more rhythmic and closely spaced (Figure 9.3a). In total, PS1 includes 21 paroxysms, 17 of which are powerful lava fountaining episodes between February 16 and April 1 2021 (Figure 9.3a). Although all episodes occur at NSEC, PS1 is notable for that all summit craters exhibit some milder intra-crateric explosive activity (Figure 9.3a).

The PS1 paroxysmal sequence closes abruptly with a final paroxysm on 1 April 2021, and is followed by circa 2 months of relative quiescence (no paroxysmal activity) in Post Paroxysmal Phase 1 (PPP1).

Paroxysmal activity resumes again at NSEC on May 19 2021, marking the onset of a second paroxysmal sequence (PS2) that terminates on October 23. PS2 includes 34 episodes in total (INGV-OE, 2021) (Figure 9.3a), and is followed by a period of relative quiescence (Post Paroxysmal Phase 2; PPP2) until the end of the year (Figure 9.3a).

9.3.2 SO₂ fluxes

Figure 9.2 shows examples of UV-Camera derived, 0.5 Hz SO₂ flux time series obtained in three distinct days, taken respectively as representative of (a) passive degassing during a quiescence period, (b) passive degassing during a paroxysmal sequence (e.g., during a repose interval in-between two consecutive paroxysmal episodes), and (c) a temporal interval encompassing a paroxysmal episode. These examples, although not exhaustive, illustrate the variety of Etna's degassing modes in 2020-2021, and clearly identify a progressive SO₂ flux increase from degassing regime (a) to (c). During quiescent periods (e.g., Figure 9.2a), the SO₂ flux exhibits the typical “pulsed” degassing behavior seen in previous work (Tamburello et al., 2013), in which nearly periodic SO₂ flux fluctuations repeat at periods of tens to hundreds of seconds. These are interpreted to reflect either atmospheric/dispersion processes, or the rhythmic release of sequences of concentrated SO₂ clouds

(or puffs) from the open vents (Tamburello et al., 2013). In this specific example, the daily averaged SO₂ flux is typically at <1000 Mg/day level, and seismic tremor amplitude is at low levels, too (Figure 9.2a, 9.3). Degassing regime (b) (passive degassing during repose intervals between two consecutive paroxysmal episodes) is similar to regime (a), but the SO₂ flux fluctuates at around higher (>>1000 Mg/day) time-averaged levels, and seismic tremor amplitude is also more intense (Figures 9.2b, 9.3). The SO₂ flux reaches its highest levels (>5000 Mg/day) in the run-up to a paroxysmal episode, during which seismic tremor and ground-based thermal activity also escalate (Figure 9.2c). Seismic tremor and thermal activity peak during the paroxysmal climax (Figure 9.2c), during which our SO₂ time series are abnormally low. This apparent sin-paroxysm SO₂ drop is a technical artifact due to the optically opaque nature of the ash-laden eruptive cloud (Figure 9.2c) that precludes quantitative SO₂ detection (D'Aleo et al., 2019).

The ash-free (cfr. 9.2.1) SO₂ flux results acquired during each observation day (examples in Figure 9.2) are averaged to derive the daily mean SO₂ fluxes illustrated in Figure 9.3b. The plot clearly highlights large temporal oscillations in the daily averaged SO₂. In the relatively calm PPa phase, the SO₂ flux averages at 1977 ± 1531 Mg/day (Figure 9.3b), but nearly doubles during the subsequent PPb phase of intensifying activity at the NSEC (mean, 3351 ± 2515 Mg/day) (here and below, we quote mean ± 1 standard deviation). Onset of PS1 on December 13 marks an abrupt SO₂ flux increase; the mean flux for this period is 4836 ± 3667 . A large inter-daily variability is observed (range, 880-19850 Mg/day) during PS1, and the peak SO₂ flux in our time series (19850 Mg/day) is observed on December 14 (a few hours after the first paroxysmal episode on December 13). End of PS1 on April 1 marks an abrupt SO₂ emission drop, and the SO₂ flux remains at low levels of ~2600 Mg/day (range, 580-8500 Mg/day) during the entire PPP1, to manifestly increase again during PS2 (mean, 4290 Mg/day; range 760-12160 Mg/day). The lowest fluxes are recorded in the PPP2 (mean, 1484 ± 1059 Mg/day).

9.3.3 Volcanic tremor and radiant heat flux

Tremor amplitude (Figure 9.3c) exhibits long-term (weeks- to months-long) fluctuations (as exemplified by the 20 days moving average; red curve), over which sharp and transient (hours) increases are superimposed that mark the repeated Strombolian and lava fountaining episodes in 2020-2021. The two paroxysmal sequences (PS1 and PS2) are therefore well evident in the seismic tremor record as clusters of repeated, abrupt tremor amplitude escalations during intense explosive intensity. The longer-term trend (red curve) identifies a progressive (though irregular), mild tremor amplitude increase from PPa to PPb. This tremor amplitude escalation culminates in phase PS1, during which seismic tremor amplitude remains sustainedly high (background tremor, red curve,

peaking on January 27). Seismic tremor amplitude then abruptly drops during phase PPP1, to resume again in phase PS2, though at reduced levels relative to phase PS1. Tremor amplitude is overall at low levels in phase PPP2, with some transient increases in December 2021 (Figure 9.3c).

Thermal anomalies are observed in SEVIRI images in correspondence to almost all the paroxysmal eruptions (Figure 9.3d). Images recorded during the 17 March, 27 May and 16 June paroxysmal episodes are affected by consistent cloud coverage that hampers derivation of erupted volumes. The paroxysmal episodes of the PS1 paroxysmal sequence are the most energetic, with heat flux peaks above 20 GW (Figure 9.3d). These convert into volumes of up to $\sim 2.5 \times 10^6 \text{ m}^3$ per episode (Figure 9.3d), providing an average output rate of about $7 \text{ m}^3/\text{s}$. The second paroxysmal sequence (PS2) is instead characterized by prevalently lower heat fluxes ($< 20 \text{ GW}$), and by an average output rate of $2.8 \text{ m}^3/\text{s}$. The median volume per episode is also lower ($< 1.6 \cdot 10^6 \text{ m}^3$), and $< 1 \cdot 10^6 \text{ m}^3$ until 22 June (Figure 9.3d). The last two episodes (21 September and 23 October) are exceptional for their heat flux ($> 20 \text{ GW}$) and estimated volumes (of $\sim 1.5 \cdot 10^6 \text{ m}^3$) (Figure 9.3d).

9.4 Discussion

Our results demonstrate that the SO_2 flux (Figure 9.3b) accelerates during the two 2020-2021 paroxysmal sequences. The cumulative SO_2 degassed mass, illustrated in Figure 9.4b, exhibits a steeper trend in phases PS1 and PS2, confirming escalating degassing activity. The two paroxysmal sequences are additionally characterised by repeated, sharp escalations in seismic tremor amplitude (Figure 9.3c) and radiant heat flux (Figure 9.3d).

That the SO_2 flux regime at Etna modulates in response to changing activity level is not a new finding itself, as there is plenty of evidence for an SO_2 flux escalation during both flank eruptions (Bruno et al., 2001; Caltabiano et al., 2004; Andronico et al., 2005) and summit crater paroxysmal sequences (Patanè et al., 2013; Spampinato et al., 2015; Corsaro et al., 2017; Coppola et al., 2019; Calvari et al., 2020b). The relationship between SO_2 , seismic tremor (Salerno et al., 2018), and heat flux (Coppola et al., 2019; Corradini et al., 2020) has also been investigated earlier. However, our 2020-2021 dataset is relevant because of the especially clear contrasting SO_2 flux regimes of paroxysmal vs. ordinary periods, and for the relatively abrupt SO_2 changes at the transitions to/from a paroxysmal sequence (compare Figures 9.3a, 9.3b). These contrasting SO_2 degassing regimes are further demonstrated by Figure 9.4, in which well-defined break-in-the-slopes in the cumulative SO_2 degassed mass trend (Figure 9.4b) mark each activity transition (Figure 9.4a). Our SO_2 record offers an opportunity to contributing to a better understanding of the mechanisms that govern the onset and duration of a paroxysmal sequence.

9.4.1 Sulfur as a tracer of magma feeding source processes

Recent advances in geophysical (Alparone et al., 2007; Aloisi et al., 2011, 2018; Patanè et al., 2013; Bonaccorso and Calvari, 2017; Cannata et al., 2018; De Gori et al., 2021) and geochemical (Allard et al., 2005; Aiuppa et al., 2010c; La Spina et al., 2015; Paonita et al., 2021) monitoring, combined with careful volcanological, petrological and remote sensing observations (Corsaro et al., 2017; Calvari et al., 2018), have led to enormous progress in understanding, interpreting and modelling the drivers of Etna's paroxysmal sequences. Consensus has been reached that the transition from ordinary to paroxysmal activity at NSEC is a response to escalating supply of volatile-rich, mafic magma into a vertically elongated, shallow (~2-4 km depth below the summit, bvs) magma storage zone (Figure 9.5a). Mixing of this mafic magma with more evolved, shallow resident magma (Kahl et al., 2015; Corsaro et al., 2017; Giuffrida and Viccaro, 2017) would cause gas-driven pressure build-up over timescales of days to months (Giuffrida and Viccaro, 2017), and ultimately eruption (Bonaccorso et al., 2013, 2021; Patanè et al., 2013; Corsaro et al., 2017; Cannata et al., 2018). For example, the PS1 sequence in late 2020 to early 2021 has recently been proposed to represent the culmination of a magma resupply event of the ~2-4 km deep magma storage zone, started in late 2020, as also indicated by a seismic swarm on December 19 2020 (Bonaccorso et al., 2021). The escalating tremor amplitude since December 13, as seen in our results (Figure 9.3c), is entirely consistent with this hypothesis.

Indirect evidence for recharge/over-pressuring of the shallow Etna's magma ponding zone comes from the petrology/geochemistry of erupted products (e.g., Pompilio et al., 2017), from modelling of seismic and geodetic signals (e.g., Bonaccorso et al., 2021), and from the chemistry of volcanic gases released prior to (Aiuppa et al., 2010c; Paonita et al., 2021) and during (Allard et al., 2005) the lava fountaining episodes. Inversion of geodetic data (Palano et al., 2008, 2017; Aloisi et al., 2011, 2018; Cannata et al., 2018), and petrological information (Métrich et al., 2004; Corsaro et al., 2017; Giuffrida and Viccaro, 2017; Cannata et al., 2018), have been used to suggest the mafic magma feeding paroxysmal sequences is sourced by an intermediate (at ~ 8-11 km depth below sea level, bsv) magma storage zone (Figure 9.5a), part of a vertically elongated, complex transcrustal magmatic feeding systems extending down to the Moho (Giuffrida and Viccaro, 2017) (Figure 9.5a). However, a fundamental challenge remains the identification of the magma input rate threshold (if any) at which the volcano behaviour transitions to a paroxysmal sequence.

SO₂ fluxes are especially useful in this regard as they can be converted into magma input (degassing) rates with the petrological method based on the equation:

$$Q_{in} = \frac{\phi_{SO_2}}{2\Delta X_S \cdot (1-x) \cdot \rho_m \cdot 86.4} \quad (\text{Eq. 9.2})$$

where Q_{in} is the magma input rate (in m^3/s) above the SO_2 exsolution level (the level at which effective sulphur (S) degassing from melt into a fluid phase starts), Φ_{SO_2} is the SO_2 flux (in Mg/day), ΔX_S is S loss upon degassing (as inferred from the difference between parental melt and residual - post-eruption - S contents in Etna's magmas; Figure 9.5b), and x and ρ_m are melt crystallinity and density (in kg/m^3), respectively.

In brief, the equation calculates the volumetric magma input rate required to justify the surface-measured SO_2 output, based on knowledge of initial (pre-degassing) and final (post surface emplacement) S content in melt, and has extensively been used at Etna to constrain degassing magma budgets over decadal (Allard, 1997; Allard et al., 2006), yearly (Coppola et al., 2019), or even daily (D'Aleo et al., 2019) timescales. Estimates point to a general decline of time-averaged Q_{in} values, from 5.4-8.9 m^3/s in the 1975-1995 period (Allard, 1997), to ~ 3 m^3/s in 1995-2005 (Allard et al., 2006), and ~ 1.6 m^3/s in 2005-2010 (Coppola et al., 2019), reflecting an SO_2 flux drop following the 2001 Etna's flank eruption. The common conclusion is, however, that such SO_2 -derived, time-averaged magma input rates are larger (1.8-4 times higher) than the mass eruption rate, implying long-term endogenous volcano growth (Allard, 1997) and deformation (Coppola et al., 2019).

Application of Equation 9.2 requires a model that suitably describes S behaviour during magmatic degassing of Etna's alkali basalts. This model, starting from knowledge of the initial S content in Etna's parental magmas, should be able to predict the fraction of S lost to vapor phase at each step of the magma decompression path, and up to surface pressure (0.1 MPa) (Allard, 1997). Deeply trapped, primitive melt inclusions are typically used to infer the initial (pre-degassing) parental melt S contents (Wallace and Edmonds, 2011). At Etna, melt inclusion results for the 2001 and 2002-2003 eruptions (Métrich et al., 2004; Spilliaert et al., 2006) indicate a parental melt S content of ~ 3000 ppm (0.3 wt. %), and a systematic decrease of dissolved S with pressure decrease (with S loss to vapour starting at pressures ≤ 150 MPa; Figure 9.5b). This 3000 ppm estimate, and 95% S extraction from melt upon degassing at ≤ 150 MPa (e.g., $\Delta S = 0.00285$ in Eq. 9.2), have been used in previous magma input rate estimates (Allard, 1997; Allard et al., 2006; Coppola et al., 2019). However, more recent work (Gennaro et al., 2019) has found more variable parental melt S contents in magmas, and less systematic (than found for the 2002-2003 melt inclusions; Spilliaert et al., 2006) pressure-dependence of melt S contents (Figure 9.5b). The authors suggest that variable redox conditions exert a strong control on dissolved S in Etna's magmas.

In view of the above, we use the model of Moretti et al (2003) to simulate S behaviour during de-compressional degassing of Etna's magmas, using two distinct scenarios for the initial S contents (2001 runs: 3000 ppm, same run conditions as in Aiuppa et al., 2007; FS runs: 2200 ppm, melt composition as in Gennaro et al., 2019). We also explore the role of magma redox by running

degassing model simulations over a range of redox conditions (from -2 to +2 log units below/above the Nickel-Nickel Oxide buffer, NNO) (see Table 9.1). Our model results (Figure 9.5b) output pressure-dependent model dissolved S contents matching the compositional range of Etna's melt inclusions (Figure 9.5b). Importantly our model calculations demonstrate that, independently on model input conditions, S loss to vapor is negligible for pressures ≥ 200 MPa, and only becomes significant for pressures ≤ 150 MPa (Figure 9.5c). Therefore, in spite the S degassing path is manifestly redox-dependent (with more reducing conditions - NNO-2 – leading to some larger “deeper/earlier” S degassing), it remains that S loss to vapor is minimal in the deep Etna's plumbing systems (e.g., at conditions deeper than the 8-11 km bsv storage zone of Figure 9.5a). This makes of the surface measured SO₂ flux a key tracer of the rates of magma supply, from the intermediate magma storage zone to the shallow portions of the Etna's plumbing system, where paroxysmal sequences are initiated (Figure 9.5).

We caution that our degassing models offer a simplified view of real degassing conditions of Etna's magma. For example, our models are run in isothermal conditions, and therefore do not consider the effect of crystallization upon cooling, and the consequent role crystals may play in favouring bubble nucleation (Woitischek et al., 2020). Likewise, the model assumes gas-melt equilibrium at each step of the decompression path, and therefore cannot account for any disequilibrium generated during gas bubble expansion within the melt (Oppenheimer et al., 2018). This latter effect, however, mostly impacts gas redox and sulphur speciation (e.g., the SO₂/H₂S ratio) rather than total S degassing behaviour. Tests made by initialising runs at different initial volatile conditions indicate that varying the parental melt H₂O content leads to some minor shift in the derived pressures of sulphur degassing initiation, but does not alter our overall conclusion of a shallow (≤ 150 MPa) condition of sulphur loss to the gas phase. Therefore, although simplified, our models offer a useful representation of the portion of the Etna's plumbing system in which sulphur degassing actively takes place.

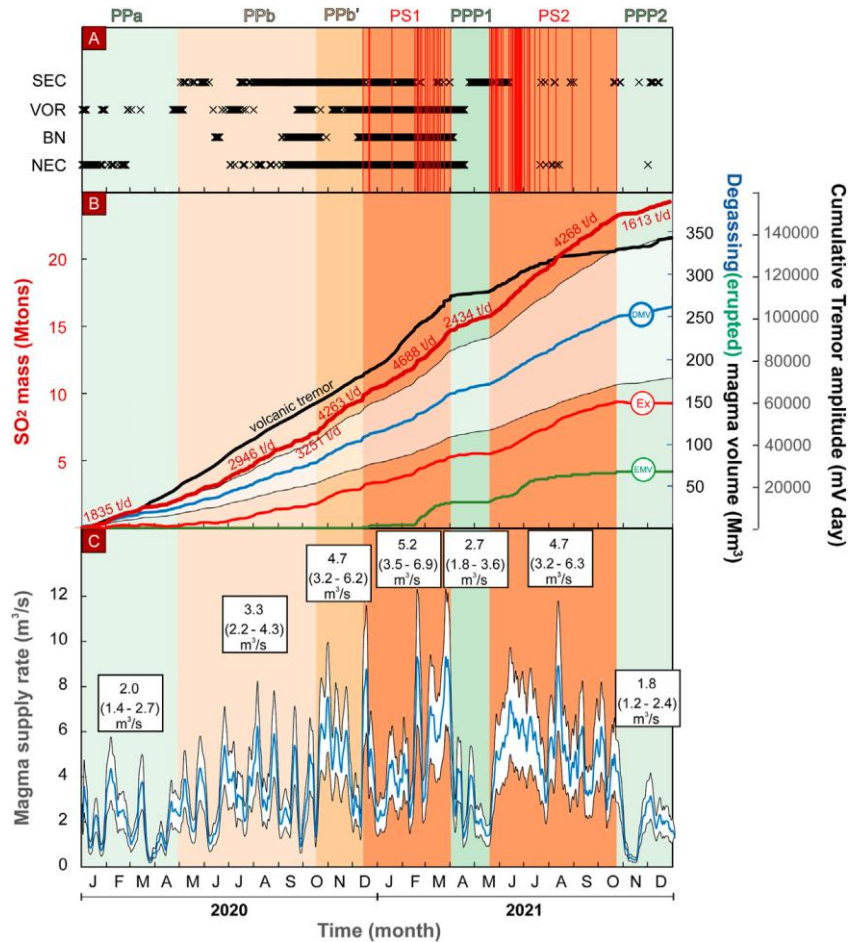


Figure 9.4 - (A) Chronogram of volcanic activity at Etna throughout 2020-2021 (same as Figure 9.3); (B) Cumulative discharged SO_2 mass (dark red solid line), derived from data in Figure 9.3 (data-gaps in the acquisition are filled by linear interpolation). Numbers in red represent time-averaged SO_2 fluxes, calculated from the slopes of the (interpolated) SO_2 mass trend. The light red line represents the Excess SO_2 mass (Ex), calculated from the cumulative discharged SO_2 mass by removing a background SO_2 flux of 1835 Mg/day (PPa average). Also shown are the cumulative Degassed Magma Volume (DMV, the volume of degassing magma required to sustain the 2021-2021 cumulative SO_2 release), obtained by summation of the volumetric magma input rates derived by applying Equation 9.2 to the interpolated SO_2 flux time series. We present an intermediate scenario (blue curve labelled "DMV") and upper/lower ranges (see white dashed area) (see text). The cumulative Erupted Magma Volume (EMV) is calculated by summing up eruptive volumes shown in Figure 9.3d; (C) Weekly mobile averages of magma input rates obtained by solving Equation 9.2. We present an intermediate scenario (blue curve) and upper/lower ranges (white area; see text). Temporal fluctuations of these weekly averaged magma input rates are used to identify distinct activity periods (as illustrated by the coloured bands; see text).

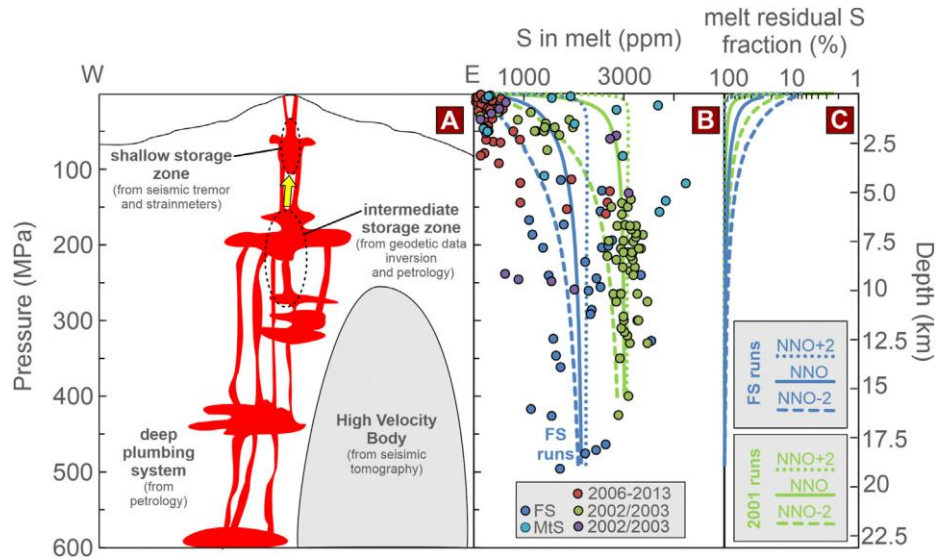


Figure 9.5 - (A) Schematic representation (E-W cross-section) of Etna's transcrustal plumbing system, as derived from synthesis of information arising from a variety of studies and disciplines: geodesy (Palano et al., 2008, 2017; Aloisi et al., 2011, 2018), seismic tremor (Alparone et al., 2007; Cannata et al., 2018), seismic tomography (Patanè et al., 2003, 2013; De Gori et al., 2021), strainmeters (Bonaccorso et al., 2013, 2021), volcanic gases (Allard et al., 2005; Aiuppa et al., 2010c; Paonita et al., 2021), and petrology (Métrich et al., 2004; Kahl et al., 2015; Corsaro et al., 2107; Giuffrida and Viccaro, 2017); (B) Pressure-dependence of S content in Etna's melt inclusions (data from Métrich et al., 2004; Spilliaert et al., 2006; Gennaro et al., 2019). Data for specific eruptions (FS, Monte Spagnolo (MtS), 2002/2003) and summit activity periods (2006-2013) are shown. Lines describe the model predicted (model of Moretti et al, 2003) evolutions of dissolved S contents in Etna's melt during simulated decompression paths. Green lines, 2001 model runs (initialised assuming a parental melt S content of 3000 ppm, and same run conditions as in Aiuppa et al., 2007); Blue lines, FS runs (initialised assuming a parental melt S content of 2200 ppm, and melt composition as in Gennaro et al., 2019). Three runs were carried for each scenario to explore a range of redox conditions (from -2 to +2 log units below/above the Nickel-Nickel Oxide buffer, NNO) (see Table 9.1). (C) Pressure-dependent, model predicted (same runs as (B)) fraction (in %) of S remaining dissolved in the melt during decompressional degassing.

	Initial pressure (MPa)	T (°K)	Redox (ANNO)	H ₂ O (wt%)	CO ₂ (wt%)	S (wt%)	SiO ₂ (wt%)	TiO ₂ (wt%)	Al ₂ O ₃ (wt%)	(FeO) T (wt%)	MnO (wt%)	MgO (wt%)	CaO (wt%)	Na ₂ O (wt%)	K ₂ O (wt%)
2001 model run [£]	400	1300	-2	3.0	2	0.3	43.7	1.7	15.7	11	0.21	6.8	10.8	3.1	1.96
"	"	"	0	"	"	"	"	"	"	"	"	"	"	"	"
"	"	"	2	"	"	"	"	"	"	"	"	"	"	"	"
FS run [§]	"	"	-2	"	"	0.22	48.6	0.22	10.2	8.5	0.17	9.5	14.2	1.7	0.94
"	"	"	0	"	"	"	"	"	"	"	"	"	"	"	"
"	"	"	2	"	"	"	"	"	"	"	"	"	"	"	"

[£]Major element composition of 2001 basaltic melt inclusions from Métrich et al., (2004) (same run conditions as in Aiuppa et al., 2007); [§]Major element composition of melt FS tephra (Gennaro et al., 2019).

Table 9.1 - Data input parameters of our degassing model runs. Parental melt composition (major and volatile elements) is represented by the primitive Etna's olivine-hosted melt inclusions. All runs simulate magma decompression under isothermal conditions (Temperature T = 1300 °K) from an initial pressure of 400 MPa down to atmospheric conditions (0.1 MPa). Redox conditions are kept constant in each model run, and vary from -2 to +2 log units below/above the Nickel-Nickel Oxide buffer in the different runs. See text for discussion.

9.4.2 SO₂-derived magma input rates

Stemming from the above, we use Equation 9.2 to convert our daily averaged SO₂ fluxes (ϕSO_2 ; data from Figure 9.3b) into daily averaged magma input rates (Q_{in}) by taking into account the possible full spectrum of ΔS and crystallinity x values in Etna's magmas (Figure 9.4b,c).

We illustrate the possible range of magma input rates Q_{in} as a white shaded area in Figure 9.4c (in the diagram, the Q_{in} values are expressed in the form of 7 days mobile averages). This area is limited by an upper range estimate (obtained using $\Delta S = 0.00205$ and crystallinity $x = 0.3$) and a lower range estimate (obtained fixing ΔS at 0.00285 and crystallinity x at 0). These upper and lower range scenarios correspond to initial S contents of 2200 and 3000 ppm respectively, and 95% S extraction from melt upon degassing (see above). Density is fixed at 2700 kg/m³ in all calculations. The blue solid line in Figure 9.4c represents an intermediate scenario, obtained by averaging results of the upper and lower range scenarios. The same upper/lower/intermediate range estimates are illustrated in Figure 9.4b in the form of cumulative degassing magma volume, e.g., the cumulative volume of degassing magma required to explain the surface SO₂ emissions. This cumulative volume is obtained from the summation of daily Q_{in} values.

Our results constrain the time-averaged magma input rate Q_{in} for the entire temporal interval (2020-2021) at 3.6 ± 2.7 m³/s (the mean values quoted here and below are based on the intermediate scenario; blue curve), within the range of previous estimates (Allard, 1997; Allard et al., 2006; Coppola et al., 2019). However, we observe large Q_{in} temporal fluctuations (Figure 9.4c), with a progressive increase from PPa (mean: 2.0 m³/s; lower-upper scenario averages: 1.4-2.7 m³/s) to PPb (mean: 3.3 m³/s; lower-upper scenario averages: 2.2-4.3 m³/s) and finally PS1 (mean: 4.7 m³/s; lower-upper scenario averages: 3.5-6.9 m³/s) (Figure 9.4c). Then, after a sharp drop in PPP1 (mean: 2.7 m³/s; lower-upper scenario averages: 1.8-3.6 m³/s), Q_{in} escalates again during PS2 (mean: 4.7 m³/s; lower-upper scenario averages: 3.2-6.3 m³/s). The lowest estimated Q_{in} values are for PPP2 (mean: 1.8 m³/s; lower-upper scenario averages: 1.2-2.4 m³/s) (Figure 9.4c).

The calculations indicate that the magma input rate is 2-3 higher during a paroxysmal sequence (PS1 and PS2) than during the ensuing Post Paroxysmal phases (PPP1 and PPP2). Also, paroxysmal sequences exhibit on average a factor 30-45% Q_{in} increase relative to the precursory Phase (PP). At this point, we focus our attention on the last two months of the precursory phase (PPb'; identified in dark orange in Figure 9.3 and 9.4), in which the time averaged SO₂ flux increases (to ~4260 Mg/day), and the corresponding Q_{in} accelerates to values (mean: 4.7 m³/s; lower-upper scenario averages: 3.2-6.2 m³/s) in all similar to those characteristic of PS1 and PS2. Manifestly, the Q_{in} rump-up pre-dates (by 1-2 months) the onset of PS1.

Ultimately, our results (Figure 9.4) suggest that onset and duration of a paroxysmal sequence are controlled by the rates of magma transport into the shallow magma plumbing system that feeds the NSEC (Figures 9.4, 9.5). The key aspect to consider is that our SO₂ flux measurements are unrepresentative of the bulk (total) plume, but rather only capture the SO₂ emissions from the South East Crater (SEC) complex (that also comprises the NSEC) and from the southern segment of the Central Craters (D'Aleo et al., 2019). Notably, our SO₂ fluxes for the 2020-2021 paroxysmal sequences (PS1-PS2; Figure 9.3b) are in the same range of the SO₂ flux time-averages derived by the INGV-OE network of UV scanners (INGV-OE, 2021; Salerno et al., 2022) that sense the distal, bulk (total) plume (Salerno et al., 2009a). This fact strongly suggests that the southern portion of the crater terrace (SEC-Bocca Nuova) becomes the Etna's dominant source of gas (of SO₂, at least) during a paroxysmal sequence. The corollary is that our inferred magma input rates (Figure 9.4c) are intended to represent supply rates to the SEC-Bocca Nuova feeding system – these can only approach the total magma input rates only when/if activity concentrates at NSEC (e.g., shortly prior, or during, a paroxysmal sequence).

9.4.3 SO₂ vs. radiant heat and seismic tremor

Additional clues can be derived from a comparative analysis of the SO₂ flux record (Figure 9.3b) with independent geophysical evidence (Figure 9.3c, 9.3d).

We infer that, in the two years covered by our study, ~15 Tg of SO₂ have cumulatively been released (Figure 9.4b). When the intermediate scenario is considered (blue curve labelled DMV), this cumulative SO₂ mass output requires degassing of $\sim 260 \cdot 10^6$ m³ of magma (from a minimum of $180 \cdot 10^6$ m³ in the lower range scenario to a maximum of $354 \cdot 10^6$ m³ in the upper range scenario). Of these, we infer that roughly half ($\sim 120 \cdot 10^6$ m³) have sustained degassing during the 266 days interval covering the two paroxysmal sequences (PS1-PS2). Our MSG-SEVIRI results (Figure 9.3d), owing to the relatively coarse (1-3 km) spatial resolution of the multispectral images used, capture only feeble and inconstant thermal anomalies during open-vent ordinary activity (PPa, PPb, PPP1 and PP2 phases) (Figure 9.3d). In contrast, very intense radiant heat fluxes are detected by SEVIRI associated to each of the PS1/PS2 paroxysmal episodes (Figure 9.3d). These radiant heat fluxes, once converted into erupted volumes (Figure 9.3d), constrain the total (cumulative) magma output (required to explain the satellite-measured thermal emissions) at $\sim 70 \cdot 10^6$ m³ (Figure 9.4b). These calculations imply that, during the 2020-2021 paroxysmal sequence, the output magma volume ($\sim 70 \cdot 10^6$ m³) corresponds to a relatively large (~58 %) proportion of the magma supplied to (and releasing SO₂ from) the shallow Etna's magma plumbing system. Such relatively large output/input fraction (0.58) is well above that typically observed during ordinary activity, when a large output vs. input unbalance

is typically observed (input/output ratios of ~ 0.1 , Allard et al., 2006). The implication is that, during a paroxysmal sequence, the transfer of magma to the surface is facilitated and, as previously found (Coppola et al., 2019), the output/input ratio approaches 1 (e.g., all magma supplied to the shallow magmatic system is erupted).

Comparison between SO_2 flux and seismic tremor amplitude is also informative (Figures 9.3, 9.4, 9.6). The seismic tremor record (Figure 9.3c) indicates clear high-frequency fluctuations (resulting from transient, intense seismic tremor increases during explosive activity) that are superimposed on a more continuous, longer-period trend. Focusing on this longer-term component (expressed by the 20 days moving average, exemplified by the red curve in Figure 9.3c), it is evident that seismic tremor is higher during a paroxysmal sequence (both PS1 and PS2) than during ordinary activity (see background seismicity in phases PPa, and PPP1 and PPP2). However, while the two paroxysmal sequences exhibit similarly elevated SO_2 degassing regimes (see Figures 9.3b, 9.4b), there is a marked contrast in their seismic tremor signatures, with PS2 being associated to less intense tremor (compare red trend in PS1 and PS2; Figure 9.3c). Also, while the SO_2 flux escalation manifestly starts in phase PPb', well before onset of paroxysmal activity (cfr. 9.4.2), the tremor increase in phases PPb and PPb' is modest, with a notable escalation being only observed during PS1 (background tremor peaks on January 27). This complex SO_2 flux vs. seismic tremor relationship is additionally indicated by Figure 9.6 that compares the time-averaged SO_2 flux vs. seismic tremor signatures for the 7 activity phases. This plot confirms that the precursory phase (PPb') is essentially manifested by temporally increasing SO_2 fluxes at nearly invariant tremor (relative to PPb), and that the PS1 and PS2 sequences have similar (within uncertainties) time-averaged SO_2 fluxes but diverse ($> \text{factor } 2$) seismic tremor amplitudes. Ordinary activity (phases PPa, PPP1 and PPP2) is characterised by consistently low SO_2 fluxes ($< 4000 \text{ Mg/day}$) and seismic tremor amplitude ($< 2 \text{ mV}$). The conclusion is that, although SO_2 flux and tremor are broadly correlated in the long-term (Figure 9.6; see also Salerno et al., 2018), their relationship is complicated by temporal swings in between the multiple tremor sources recognised on Etna (Cannata et al., 2008, 2015; Zuccarello et al., 2022).

Although precise seismic tremor source location is beyond the scope of this study, we use the ratio of the seismic tremor amplitudes measured at the ECPN and ESLN seismic stations (e.g., the ECPN/ESLN tremor amplitude ratio; Figure 9.7) to investigate the temporal stability vs. variability of the seismic tremor source in 2020-2021. This approach stands on the basis that temporal shifts in the ratio of tremor amplitudes recorded at summit (ECPN) vs. intermediate height (ESLN) seismic station (see location in inset of Figure 9.7b and in Figure 9.1d) are informative of spatio-temporal changes in seismic tremor source (Alparone et al., 2003, 2007; Cannata et al., 2008). We identify

large oscillations in the ECPN/ESLN seismic tremor ratio in 2020-2021 (Figure 9.7) that are clearly indicative of a non-stationary seismic tremor source. These ratio fluctuations, in combination with information on seismic tremor location (extracted from INGV-OE reports; see Figure 9.7), support distinct PS1 vs. PS2 tremor signatures; in which, during phase PS2, seismic tremor source is irradiated deeper (in May-June 2021; as indicated by low ECPN/ESLN ratio) and/or underneath BN/VOR crater sector (during July-October 2021), while seismic tremor source is more stably underneath the SEC in phase PS1 (Figure 9.7). A manifest upward migration (increasing ECPN/ESLN ratio) of the seismic tremor source, to cluster underneath the SEC, is also detectable in phases PPb and PPb' (Figure 9.7).

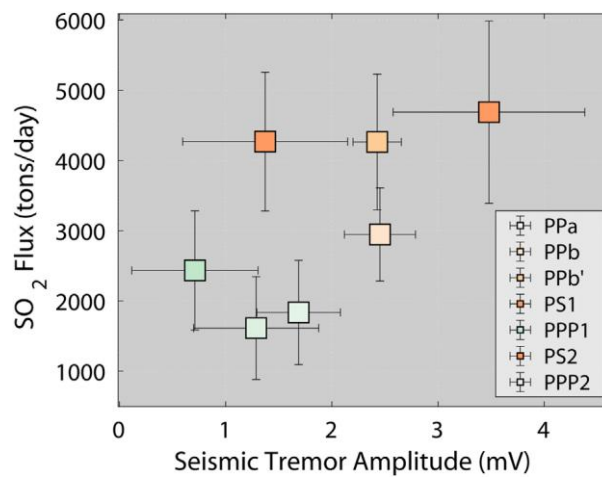


Figure 9.6 - Scatter plot of time averaged seismic tremor amplitude vs. SO_2 flux in the 7 activity phases discussed in the text. Data plot as mean $\pm$ 1 standard deviation. Ordinary activity (phases PPa, PPP1 and PPP2) are characterised by reduced tremor amplitude (< 2 mV) and SO_2 flux (< 3000 Mg/day). In the preparatory phase PPb, and especially PPb', the mean SO_2 flux increases to > 3000 Mg/day, and mean seismic tremor amplitude is between 2 and 3 mV. The two paroxysmal sequences exhibit similar mean SO_2 flux regimes but contrasting seismic tremor signature. See text for discussion.

9.4.4 Implications for the drivers of paroxysmal sequences

Drawing on all the evidence, we propose a mechanism in which, during ordinary activity (e.g., phase PPa), volcanic activity is concentrated at the Central Crater (VOR) and NEC (Figures 9.2a, 9.3a). The VOR/NEC vent area has long been recognised as the most stable (persistent) magma degassing pathway during ordinary activity, while the SEC is thought to represent the surface expression of a secondary lateral branching (connected to the VOR/NEC in a branched feeding conduit geometry) (Marchetti et al., 2009; Aiuppa et al., 2010c; Cannata et al., 2015). Our derived low SO_2 fluxes (< 2000 Mg/day) and magma input rates ($\sim 1\text{-}3$ m³/s) during the PPa phase (Figures 9.3, 9.4, 9.6) imply limited magma transport toward the south-eastern branching system (that feeds the SEC), and rather suggest preferential magma channelling in the “stable” central feeding conduit system. Seismic tremor locations clustering in the VOR/NEC area confirm this (INGV-OE, 2020;

Figure 9.7). The radiant heat flux is low (Figure 9.3d), implying limited surface magma emplacement. This is typical of ordinary activity phases (e.g., Coppola et al., 2019), when a high input/output magma unbalance is typically observed that implicates shallow magma circulating in the central conduits evolves in open-system degassing conditions (Aiuppa et al., 2010c; Edmonds et al., 2022). Open-system degassing implies efficient (low-pressure) separation of gas bubbles from melt to sustain quiescent open-vent degassing (and minor intra-crateric explosive activity) at VOR/NEC, and leads to recycling of dense/degassed magma deep into the plumbing system (Allard, 1997). Seismic tremor amplitude remains at low (background) levels (Figures 9.3, 9.6).

Increasing SO₂ fluxes (Figures 9.3, 9.4, 9.6) and magma input rates (Figure 9.4c) in phase PPb imply preferential magma migration toward the southern part of the crater terrace, and its channelling into the structurally/gravitationally unstable (Bonforte et al., 2017) Etna's south-eastern upper flank, where the SEC branching system insists (Marchetti et al., 2009; Aiuppa et al., 2010c; Patanè et al., 2013). This increased magma supply rate causes progressive re-awakening of the NSEC (Figure 9.3a), and some moderate seismic tremor increase (Figures 9.3c, 9.6). Notably, the seismic tremor source consistently migrates upward and toward the SEC in this period (Figure 9.7; INGV-OE, 2020). The process intensifies in phase PPb' when (at relatively invariant tremor amplitude; Figure 9.6) the SO₂ flux ramps up to > 4000 Mg/day, suggesting accelerating magma transport (and degassing) toward the SEC. We propose that, as the magma input rate into the shallow SEC magma plumbing system increases to > 4 m³/s (PPb' to PS1), such larger volumes of feeding mafic magma cannot be accommodated by quiescent (passive) open-system degassing, but rather lead to magma eruption in a paroxysmal sequence. We argue, based on previous work, that ascent of faster moving bubbly magma may lead to foam accumulation on-top of the SEC shallow magma reservoir (Allard et al., 2005), ultimately leading to pressure-build up (Bonaccorso et al., 2013, 2021), and finally eruption. Elevated seismic tremor and radiant heat at the PS1 onset mark explosive magma outburst in repeated paroxysmal episodes (Figures 9.3c, 9.3d). In this view, the sequence of paroxysmal episodes would perdure until fresh magma feeding into reservoir that feeds the NSEC vanishes down to the quiescent (persistent activity) values (in phase PPP1; Figure 9.4). Ultimately the process repeats again in May to December 2021, with phases PS1 and PPP2.

Our SO₂ flux record (Figure 9.3b) is evidence for the utility of spatially resolved UV Camera observations in detecting gas/magma migration prior to/during clustering of volcanic activity at one specific crater. In particular, the manifest SO₂ flux increase in phase PPb' (Figures 9.3, 9.6), well before onset of paroxysmal sequence PS1, suggests that UV Camera observations have the potential to trace increasing magma supply weeks to months in advance to resumption of paroxysmal activity. However, comparison between the two recorded paroxysmal sequences (Figures 9.3, 9.4, 9.6) clearly

shows that the transition from ordinary to paroxysmal activity can not only happen progressively (as in the case of PS1), but also suddenly (PS2). SO₂, tremor and thermal records all increase abruptly at the PS2 onset, and we see no detectable precursory SO₂ flux increase beforehand (Figure 9.3b). The diversity between PS1 and PS2 sequences is additionally demonstrated by the excess cumulative SO₂ mass, shown in Figure 9.4b (red curve labelled “Ex”). This is calculated by subtracting the average SO₂ flux for phase PPa (taken as representative of background degassing) from the cumulative SO₂ mass. This excess SO₂ mass progressively ramps up (during phases PPb to PPb') prior to PS1, implying a gradual, protracted (several months long) acceleration in the SO₂ degassing regime. In contrast, the excess SO₂ mass does not show any precursory increase prior to onset of PS2, and rises contemporaneously with the start of paroxysmal activity. Tremor amplitude also rises during PS2, but to a lesser extent than in PS1 (Figures 9.3, 9.6), implying distinct tremor source area (Figure 9.7). In particular, the prevalingly low ECPN/ESLN tremor amplitude ratios (Figure 9.7) suggest deepening of seismic source location in phase PPP1, and at the onset of phase PS2 (INGV-OE 2021). This comparison demonstrates the challenges in anticipating paroxysmal behaviour at Etna, especially when the driving processes are rapid (hours) and deeply initiated (e.g., PS2).

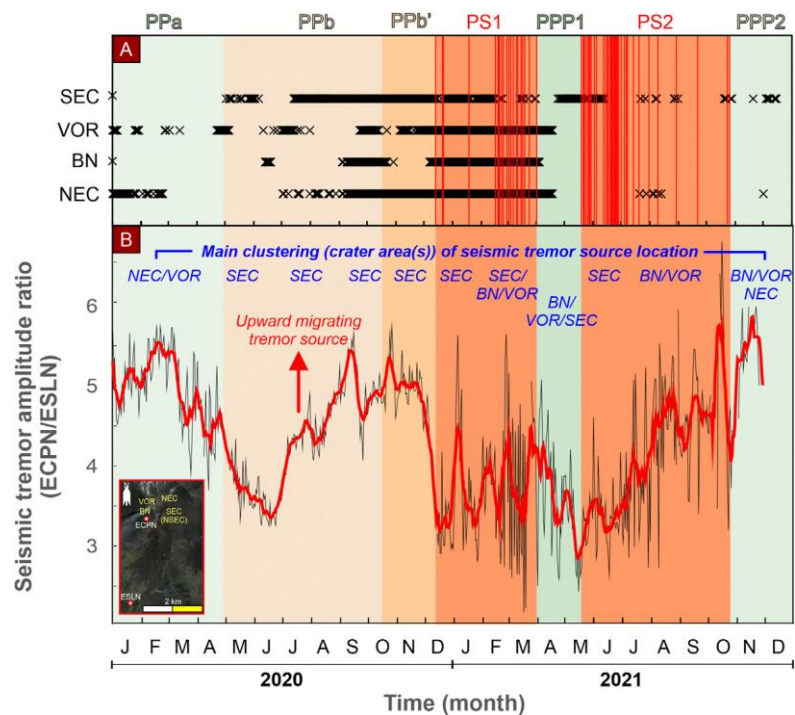


Figure 9.7 - (A) Chronogram of volcanic activity at Etna throughout 2020-2021 (same as Figure 9.3). (B) Temporal record (2020-2021) of the ratio of tremor amplitudes measured at the ECPN and ESLN seismic stations. The ratio is calculated by ratioing the daily averaged tremor amplitudes from the two stations (a 10 days mobile average is shown in red). The temporal variability of the ratio supports significant spatio-temporal changes in seismic tremor source (Alparone et al., 2003, 2007; Cannata et al., 2008). These temporal swings are additionally indicated by seismic tremor locations (in blue), extracted by INGV-OE reports (INGV-OE, 2020, 2021), that indicate the crater sector below which seismic tremor source locations are clustered.

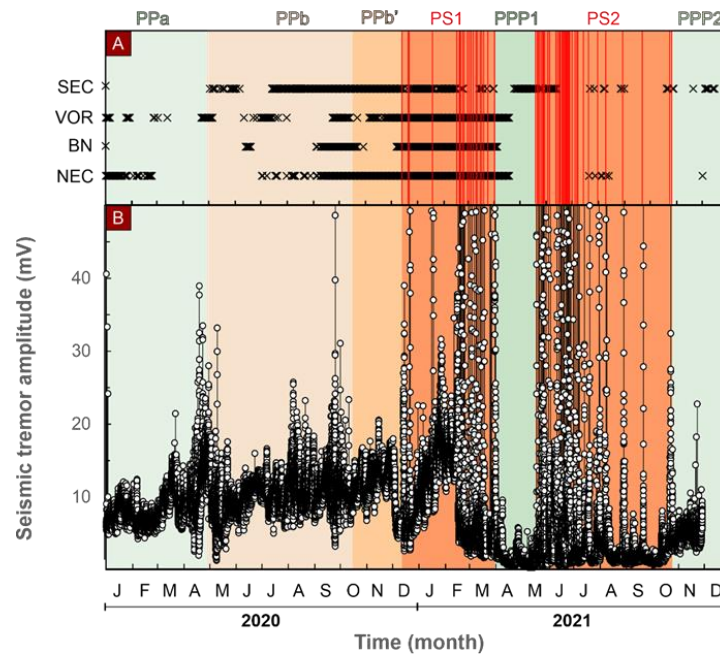
9.5 Conclusion

We have shown here that the Etna's SO₂ flux emissions ramp up, by a factor 2-3 or more (on average), during a paroxysmal sequence (relative to ordinary activity). During a paroxysmal sequence, peak SO₂ flux emissions as high as ~20 Gg/day can be reached, or ~10 times more than the background (ordinary activity) time-averaged levels. Our results, therefore, confirm the fluctuating, non-stationary nature of volcanic gas release at open-vent, persistently degassing volcanoes (Carn et al., 2017). The implication is that, at the many persistently degassing volcanoes worldwide, semi-continuous gas observations (with automated instrumental networks, e.g., Arellano et al., 2021) should be privileged over more infrequent (campaign-style) measurements.

Our results also confirm the benefits of using volcanic SO₂ sensing tools (such as UV Cameras) that can spatially resolve emissions from distinct vents/craters. At the many volcanoes worldwide having multiple degassing vents, UV Cameras, especially if used in tandem with UV scanning spectrometers that image the bulk (total) plume from more distal locations (e.g., Salerno et al., 2009a), can effectively allow tracking intensification of degassing at one specific vent (Etna's NSEC in our case) before paroxysmal activity commences there.

Interpretation of SO₂ flux time series requires a model is available for S evolution during magma ascent/degassing in the subsurface (e.g., Figure 9.5). Modeling S degassing behavior is still complicated by our still fragmentary understanding of S partitioning behavior between melt and fluid (e.g., Ding et al., 2023). It is anticipated that S studies at volcanoes should become even more interdisciplinary in the future, merging expertise from gas and melt inclusion geochemistry, experimental petrology, and thermodynamics/modelling.

SUPPLEMENTARY MATERIAL (FIGURES)



Supplementary Figure S9.1. (A) Chronogram of volcanic activity at Etna throughout 2020-2021. Crosses indicate mild explosive activity (either impulsive ash discharge events or intra-crateric strombolian activity) at one of the summit craters (NEC: Northeast crater; VOR: Voragine crater; BN: Bocca Nuova crater; SEC: Southeast crater). Red vertical bars identify paroxysmal episodes at SEC (lava fountains during paroxysmal sequences PS1 and PS2; indicated by dark orange shading). (B) Minute-averaged seismic tremor RMS amplitude, as measured at the INGV-OE ECPN broadband seismometer.

SUPPLEMENTARY TABLE:

Due to the large size, the Supplementary Table S9.1 for this article can be found online at:

<https://www.frontiersin.org/articles/10.3389/feart.2023.1115111/full#supplementary-material>

CHAPTER 10

CONCLUDING REMARKS

This thesis has addressed the role of volcanic gas monitoring in advancing both the understanding of magmatic processes and the capacity to forecast eruptive dynamics at persistently active open-vent volcanoes. By exploiting long-term, high-resolution SO₂ flux datasets acquired with permanent UV Camera systems at Mount Etna and Stromboli volcanoes, it has been possible to investigate degassing patterns across a wide spectrum of eruptive styles.

The research carried out highlights the diagnostic power of SO₂ and CO₂ fluxes in advancing the capacity to forecast eruptive events at persistently active open-vent volcanoes, while also emphasizing the importance of addressing methodological uncertainties and integrating multiple observational approaches. At Stromboli, the first long-term comparison between a near-vent UV Camera and distal scanning spectrometers (employing the DOAS technique) SO₂ flux measurements revealed a systematic offset. This discrepancy is primarily attributable to plume-atmosphere interactions affecting gas column density retrievals from scanning spectrometers, as well as radiative transfer and topographic effects influencing UV Camera observations. To overcome these limitations, an integrated SO₂ flux was proposed, combining the strengths of both techniques and providing measurements less affected by external factors (e.g., meteorological, illumination and volcano topography conditions) than using any of the two techniques alone, and hence a better proxy of volcano behaviour. Moreover, statistical analyses of multi-year SO₂ and CO₂ datasets demonstrated that Stromboli's major explosions are frequently preceded by a consistent pattern of reduced SO₂ flux and elevated CO₂/SO₂ ratios. These findings supported the development of a gas-based composite indicator capable of retrospectively forecasting a significant fraction (over 70%) of explosions on timescales of weeks. Furthermore, the analysis of gas plume composition and fluxes during the eruptive sequence of 4-11 July 2024 eruption, highlighted the complementary role of CO₂ and SO₂ fluxes, proposing a CO₂-driven, deep trigger for the paroxysmal explosion of 11 July. Comparison with previous degassing signals further indicates that excess pre-eruptive CO₂ is a systematic precursor of paroxysmal activity, reinforcing the role of CO₂ plume monitoring as a valuable tool for eruption forecasting at open-vent mafic volcanoes, albeit with intrinsic limitations in constraining the exact timing and evolution of eruptive sequences.

At Etna, ~10 years of continuous SO₂ flux measurements, integrated with satellite-derived thermal fluxes, enabled the characterization of degassing regimes across the main eruptive phases. Results demonstrate that, despite the challenges posed by variations in eruption source location,

intensity, and instrumental limitations, the employed UV Camera system was capable to successfully resolve a clear (above background) increase in eruptive SO₂ flux, achieving relatively high detection efficiencies for the pre-eruptive, syn-eruptive, and post-eruptive periods. Volumetric estimates derived from thermal and degassing data confirmed the large imbalance between degassed and erupted magma volumes at Etna, with only ~27% of the ascending magma being effectively discharged during eruptions, and the remaining magma fraction (~73%) remaining stored in the intrusive magmatic system. The extensive UV Camera-derived SO₂ flux dataset (~10 years) identifies the 2020-2021 paroxysmal sequences at Etna as the most intense eruptive phases of the decade. These sequences were triggered by rapid injections of volatile-rich magma into a shallow reservoir at ~3-4 km depth, which promoted sulphur exsolution and degassing, and were associated with an approximate doubling of the time-averaged magma supply rate.

Overall, obtained results demonstrates that UV Camera measurements provide critical insights into the degassing dynamics of open-vent systems, capturing both long and short-term anomalies that may herald eruptive activity. At the same time, these findings highlight the inherent limitations of relying on a single monitoring technique: while UV Camera observations offer valuable and high-frequency measurements, technical and environmental constraints can limit their effectiveness. These considerations underscore the necessity of integrating gas data with complementary geophysical, petrological, and satellite-based datasets to obtain a more comprehensive understanding of volcanic processes. Progress in these directions will be essential for refining monitoring strategies and improving the interpretation of eruptive precursors, enhancing eruption forecasting capabilities.

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