

# New insights into 2021 La Palma eruption degassing processes from direct-sun spectroscopic measurements

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**Abstract.** In a world increasingly impacted by climate change and natural hazards, atmospheric monitoring networks are essential for informed decision-making. During the 2021 La Palma eruption, we combined existing and rapidly deployed instruments to monitor volcanic gas emissions up to 140 km from the source. We used direct-sun measurements from low- (EM27/SUN) and high- (IFS-125HR) resolution Fourier Transform InfraRed (FTIR) spectrometers contributing to key atmospheric global networks. In La Palma, the EM27/SUN was combined with a Differential Optical Absorption Spectroscopy (DOAS) instrument. We present new FTIR retrieval methods to derive the  $\text{SO}_2$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{HF}$ ,  $\text{HCl}$  relative abundance in the plume from both low- and high- resolution solar absorption spectra. Using Sentinel-5P TROPOMI data, we derived  $\text{SO}_2$  fluxes and estimated total emissions of  $1.8 \pm 0.2$  Mt  $\text{SO}_2$ ,  $19.4 \pm 1.8$  Mt  $\text{CO}_2$ ,  $0.123 \pm 0.005$  Mt  $\text{CO}$ ,  $0.05 \pm 0.01$  Mt  $\text{HCl}$ , and  $0.013 \pm 0.002$  Mt  $\text{HF}$  over the course of the eruption. These results are consistent with mass balance derived from petrologic degassing estimates. This study demonstrates that high- and low-resolution FTIR and DOAS spectrometers, integrated within global monitoring networks, can provide quantitative constraints on volcanic gas composition and fluxes over large distances. Such capabilities are directly applicable to volcanic crisis monitoring, complement satellite observations, and support improved assessments of volcanic impacts on atmospheric composition at regional scales.

## 1. Introduction

Volcanic emissions of greenhouse gases and pollutants remain poorly constrained due to the limited number of volcanoes well monitored for gas emissions. The present knowledge relies on either short-term records at permanent stations or on discrete campaigns of measurements, mostly during eruptive crises. Characterizing volcanic degassing processes is essential to improve our understanding of the multi-species volcanic gas

45 emissions across various geodynamic settings and their long- and short-term impact on the atmospheric  
46 composition.

47 The abundance and composition of dissolved volatiles control the buoyancy and viscosity of magmas,  
48 making them a primary driver of eruptive dynamism and duration (Longpré et al., 2025). Water (H<sub>2</sub>O) and  
49 carbon dioxide (CO<sub>2</sub>) are the most abundant species in volcanic degassing, followed by sulfur dioxide (SO<sub>2</sub>) and  
50 halogen-derived species (mainly halides). They show different solubility in magma, which depends mainly on  
51 pressure, temperature and redox conditions (Gennaro et al., 2020; Cassidy et al., 2022). CO<sub>2</sub> and H<sub>2</sub>O are  
52 usually among the deepest exsolved gas species, followed by SO<sub>2</sub> and halogens in sub-surface. Therefore, the  
53 exploration of their pre- and co-eruptive relative abundance can reveal critical information on pressurisation of  
54 the magma plumbing system, as well as on ascent rates and volatile exsolution pathways (Voigt et al., 2014;  
55 Taquet et al., 2019). The temporal evolution of the  $\Delta\text{CO}_2/\text{SO}_2$  ratio and halogen-derived species-to-SO<sub>2</sub> ratios in  
56 volcanic plumes have often been used to infer the respective contribution of deep to shallow magmatic  
57 processes in the transitions in eruptive dynamism such as changes in the bubble contents in the magma chamber,  
58 replenishment, magma batches mixing or fractional crystallisation (Harris and Rose, 1996; Shinohara et al.,  
59 2003, 2008; Werner et al., 2012; La Spina et al., 2015). Volcanic plume compositions, when combined with  
60 seismic and structural data, help constrain volatile fluxes, magma ascent rates, and the architecture of the  
61 magmatic plumbing system. Integrating gas measurements with petrological constraints from matrix, melt  
62 inclusions (MI), and fluid inclusions (FI) enables reconstruction of pre-eruptive volatile contents and degassing  
63 pathways, which are key to modeling eruption dynamics (e.g.: Ubide et al., 2023; Longpré et al., 2025).

64 The 2021 Cumbre Vieja (La Palma) fissure eruption (from 19 September to 14 December 2021, VEI  
65 3), called Tajogaite, was the first subaerial eruption in 50 years in the Canary Islands archipelago and thus the  
66 first opportunity to directly assess the amount and composition of volcanic degassing during an eruption in  
67 Canary Islands (Burton et al., 2023). It was preceded by up to 12 low intensity seismic swarms between October  
68 2017 and September 2021, occurring at depths between 20 and 30 km, without evidence of surface deformation  
69 (Torres-Gonzalez et al., 2020; Mezcua and Rueda, 2023). Some of these seismic swarms were accompanied by  
70 changes in flux or composition of trace gases (CO<sub>2</sub>, He, Rn) in soil or at the Dos Aguas cold spring located in  
71 the Caldera de Taburiente to the north (Torres-Gonzalez et al., 2020; Padrón et al., 2022). These observations  
72 were interpreted as evidence of magma migration from a deeper upper mantle reservoir to a shallower sub-  
73 crustal reservoir (Padrón et al., 2022). On 11 September 2021, a new seismic swarm occurred at ~10 km depth  
74 and intensified over the following days, accompanied by ground inflation reaching 30 cm (De Luca et al., 2022).  
75 Subsequently, the seismicity migrated towards the surface and the Tajogaite eruption started on 19 September  
76 2021. Several craters opened and grew along a NW-SE eruptive fracture (Muñoz et al., 2022) on the western  
77 flank of the Cumbre Vieja Ridge (CVR). The eruption exhibited simultaneously multiple eruptive styles at  
78 various summital and flank vents, including more than 100 m-high hawaiian lava fountains, strombolian  
79 spattering activity, ash venting, vulcanian explosions and significant effusive activity. Over the 85 days of its  
80 activity, it produced a  $\sim 1.8 \times 10^8 \text{ m}^3$  lava flow field (Civico et al., 2022) covering an area of 12 km<sup>2</sup>, and a  
81 tephra blanket with a total estimated volume of  $\sim 2.3 \times 10^7 \text{ m}^3$  (Bonadonna et al., 2022), provoking the  
82 evacuation of several thousands of people and the destruction of ~3000 buildings (Copernicus EMSR546,  
83 PEVOLCA reports). During the course of the eruption, volcanic gases were injected between 1000 and 6000 m  
84 a.s.l. (Bonadonna et al., 2022; Milford et al., 2023; Hedelt et al., 2025) and were transported over North Africa  
85 and Europe, as well as across the Atlantic to the Caribbean on several occasions (Hedelt et al., 2025). Total SO<sub>2</sub>  
86 emissions were estimated to be about 1.84 Mt (Milford et al., 2023) using the daily mass estimates derived from  
87 the TROPOspheric Monitoring Instrument (TROPOMI) measurements and provided by MOUNTS-Project  
88 ([mounts](#), Valade et al., 2019).

89 Geophysical and geochemical co-eruptive observations revealed insights into the structure of the  
90 plumbing system (d'Auria et al., 2022; Dayton et al. 2023) and melt evolution during the eruption (Day et al.,  
91 2022; Ubide et al., 2023; Dayton et al., 2024; Longpré et al., 2025). Co-eruptive seismicity defines to clusters  
92 (d'Auria et al., 2022; Del Fresno et al., 2023), the shallowest one ranging between 5 and 15 km depth starting on  
93 26 September 2021 and remaining until the end of the eruptive period, and the deeper ranging between 20 and  
94 25 km depth occurring from 1 October to 13 December 2021. Additionally, a temporal progression in the melt

95 chemical composition was observed: the initial erupted magma exhibited a tephritic composition (MgO ~6 wt%  
96 and TiO<sub>2</sub> ~4 wt%) and was gradually (< day 20; Day et al. 2022) replaced with a basanitic magma (MgO ~8  
97 wt% and TiO<sub>2</sub> 3.7 wt%) for the rest of the eruption (Day et al., 2022; Ubide et al., 2023). This type of transition  
98 reflects a behavior similar to that previously documented for the 1949 and 1971 Cumbre Vieja eruptions (Klügel  
99 et al., 2000), and was interpreted as mixing between a resident mush and deep fresh basaltic magmas in the  
100 shallow reservoir. Such changes in magma composition could contribute to changes in eruptive dynamism and  
101 might be reflected in surface gas composition changes. In fact, variability in the eruptive dynamism was  
102 observed through seismic and deformation monitoring (Del Fresno et al., 2023; Charco et al., 2023), tephra  
103 analysis and geochemical lava and ash studies (Bonadonna et al., 2022; 2023; Birnbaum et al., 2023; Longpré et  
104 al., 2025). In the early phase of the Tajogaite eruption, rapid cone growth and vent openings were accompanied  
105 with explosive tephra ejections. On 25 September, a significant cone collapse was accompanied with increasing  
106 explosive activity with evidence of white xeno-pumice fragments in tephra (Day et al., 2022; Romero et al.,  
107 2022). Lava became more fluid after the transition from tephritic to basanitic composition. By late October-  
108 November, plume height stabilized at 2500–3500 m a.s.l. (Córdoba-Jabonero et al., 2023), with lower SO<sub>2</sub>  
109 emissions (Milford et al., 2023). The final weeks saw intense activity, collapses, structural changes and vents  
110 reconfiguration (Gonzalez, 2022; Walter et al., 2023).

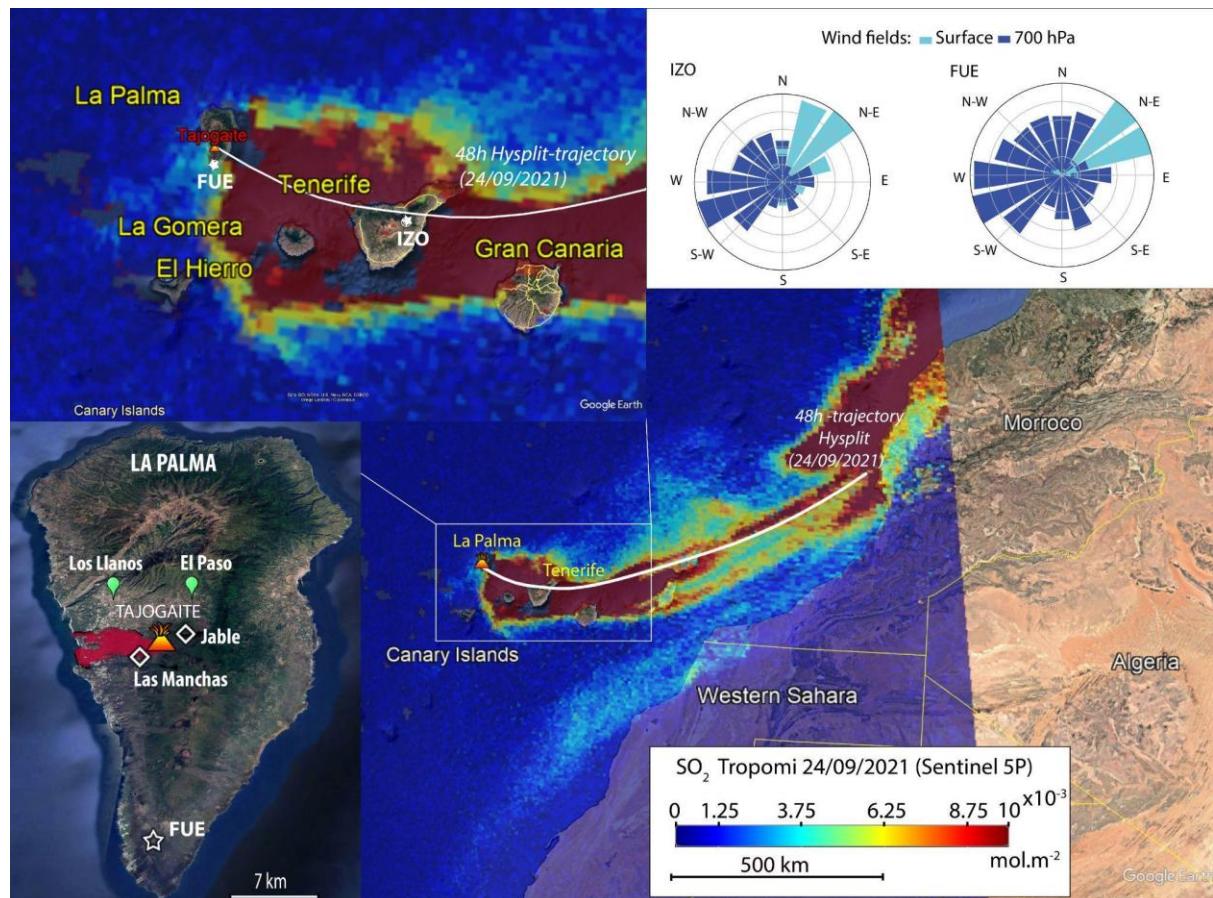
111 To date, only a few studies have reported the composition of the gas plume measured during the  
112 Tajogaite eruption, and none have provided a multi-species time series of estimated emission fluxes over the  
113 entire eruptive period. Erickson et al. (2024) derived CO<sub>2</sub> volcanic emission fluxes from drone-borne SBA-5  
114 infrared CO<sub>2</sub> sensors measurements and also measured  $\Delta\text{CO}_2/\text{SO}_2$  ratios using ground-based Multi-GAS  
115 instruments localized near the vent. Burton et al. (2023) reported the first time series of the  $\Delta\text{CO}_2/\text{SO}_2$  ratio of  
116 the gas plume, employing ground-based FTIR spectrometry techniques using incandescent ash plumes, lava  
117 fountaining and lava flow as thermal sources, and occasional solar absorption measurements. They also reported  
118 drone-borne and ground-based MultiGAS in-plume measurements. Recently, Asensio-Ramos et al. (2025)  
119 reported the first time series of  $\Delta\text{CO}_2/\text{SO}_2$ , SO<sub>2</sub>/HCl and  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios measured at the base of the eruptive  
120 column using open-path FTIR measurements with lava fountaining and lava flows as thermal source. Using the  
121 surface gas measurements, petrological data and estimates of lava emission rates, these authors reveal evidence  
122 of exceptional CO<sub>2</sub>-rich gas emission with respect to the emitted lava volume during the eruption. Recent  
123 studies showed the presence of particularly SO<sub>2</sub>- and CO<sub>2</sub>-rich compositions of deeply entrapped-MI in volcanic  
124 rocks from the Canary Islands (Longpré et al., 2017; Taracsak et al., 2019), which may be linked to mantle  
125 metasomatism (Hansteen et al., 1991; 1998).

126 This study presents a comprehensive time series of  $\Delta\text{CO}_2$ ,  $\Delta\text{CO}$ , HCl, and HF to SO<sub>2</sub> molar ratios  
127 measured in the Tajogaite volcanic plume between 21 September and 14 December 2021, spanning the full  
128 duration of the eruption. The measurements were conducted at distances of 15 km and 140 km from the vent  
129 using ground-based direct-sun FTIR and DOAS instruments, integrated into global atmospheric monitoring  
130 networks. Ground-based FTIR and UV direct-sun methods provide multi-species and time-resolved total column  
131 measurements of the main volcanic gases, regardless of the plume altitude, while ensuring operators and  
132 instruments safety (Butz et al., 2017; Taquet et al., 2023). They have the advantage of using the sun as a  
133 common and both homogeneous and constant-intensity source (at the timescale of a single measurement),  
134 providing solar spectra in a wide spectral range and with a high signal-to-noise ratio. We took advantage of the  
135 instrumentation installed at the Izaña Atmospheric Observatory (IZO) in Tenerife. Its high altitude and  
136 geographical location were ideal for repeatedly directly capturing the volcanic plume including in situ surface  
137 measurements, thereby enhancing the temporal density of our dataset. We estimated daily SO<sub>2</sub> volcanic  
138 emission fluxes from space-based TROPOMI/Sentinel-5P measurements, and used the measured species-to-SO<sub>2</sub>  
139 ratio to derive the emission fluxes of the other volcanic species and their total emissions. Our results are  
140 interpreted in the light of petrological (including new melt inclusions and matrix glass compositions presented in  
141 this study) and geophysical data taken from the literature.

## 142 2. Gas and particulate matter measurement sites and instrumentation

143 A comprehensive network for the monitoring of trace gases, aerosols and ash fallout was operative for  
 144 air quality monitoring and scientific research during the eruption. Monitoring efforts relied on a combination of  
 145 permanent stations which are part of international atmospheric research and air quality monitoring networks,  
 146 such as those in the Canary Islands Government Air Quality Monitoring Network (AQMN) and the facilities at  
 147 IZO, as well as additional equipments specifically installed for attending the volcanic emergency. In this  
 148 framework, the State Meteorological Agency of Spain (AEMET), through the Izaña Atmospheric Research  
 149 Center (IARC) and the Territorial Delegation of AEMET in the Canary Islands (DTCAN) and in collaboration  
 150 with the Spanish National Research Council (CSIC) and other institutions, deployed scientific instrumentation  
 151 on La Palma. The objectives of the deployment were: 1) real-time monitoring and characterization of the  
 152 vertical structure of the eruptive plume, carried out through the implementation of an aerosol profiling network  
 153 in the context of the European Aerosol, Clouds and Trace Gases Research Infrastructure (e.g.: ACTRIS, 2021;  
 154 Barreto et al., 2022; Álvarez et al., 2023); 2) complementing the air quality network observations managed by  
 155 the Government of the Canary Islands (Milford et al., 2023, and references therein); and 3) investigating the  
 156 physicochemical composition of the volcanic plume, its links with the evolution of the eruptive process and  
 157 studying the ash-gas-aerosol interactions (e.g.: Garcia et al., 2022; Cordoba-Jabonero et al. 2023; Cuevas et al.,  
 158 2024, and references therein).

159 We conducted remote sensing and surface gas and ash measurements during the entire eruptive period  
 160 at two stations localised in La Palma (FUE) and Tenerife (IZO) islands (Fig. 1, upper panel) to assess the co-  
 161 and post-eruptive compositional variability of the Tajogaite volcanic plume. In addition, aerosol and surface  
 162 SO<sub>2</sub> measurements were conducted at two locations on La Palma (Los Llanos, El Paso) and Tenerife (IZO).  
 163 Mobile in-situ plume measurements in La Palma using MultiGas were also performed during episodes of plume  
 164 grounding driven by favorable meteorological conditions. Figure 1 displays a map of the FUE and IZO stations,  
 165 as well as the MultiGas, aerosols and SO<sub>2</sub> measurement sites in La Palma concurrently with a typical SO<sub>2</sub> plume  
 166 as detected by space-based TROPOMI/Sentinel-5P sensor. The instruments at each site and the measurement  
 167 periods are summarised in Table 1 and detailed below.



169 **Figure 1: Location of our measurement stations in Canary Islands during the 2021 La Palma eruption (FUE and IZO**  
 170 **represent the Fuencaliente and Izaña stations, respectively, marked by white stars). SO<sub>2</sub> data from**  
 171 **TROPOMI/Sentinel-5P sensor are shown in the map for 24 September 2021, illustrating the typical plume dispersion**  
 172 **over hundreds of kilometres. The instruments implemented at FUE and IZO stations are summarised in Table 1.**  
 173 **Wind rose diagrams for surface and 700 hPa levels (corresponding to the average of the plume altitude during the**  
 174 **eruption) are also presented for the IZO and FUE stations (upper right panel), considering the entire eruptive period**  
 175 **and the ECMWF Reanalysis v5 (ERA5) data (ECMWF: <https://www.ecmwf.int/en/forecasts/dataset/ecmwf-reanalysis-v5>).** The base layer was sourced from Google Earth (© Google), while the SO<sub>2</sub> distribution map was  
 176 **derived from TROPOMI data accessed through the Sentinel Hub platform. The upper left panel presents a zoom on**  
 177 **La Palma Island including all of the surface (white hollow diamonds for MultiGAS), aerosols (green) and columns**  
 178 **gas (white stars) measurement sites from which data is used for this study. The Tajogaite eruption lava flow field (red**  
 179 **shaded area) was taken from the European Environment Agency Copernicus Emergency Management Service**  
 180 **(<https://emergency.copernicus.eu/mapping/list-of-components/EMSR546>).**

182 **Table 1: Solar FTIR - DOAS and surface gas and particulate matter in situ measurements conducted at the FUE,**  
 183 **IZO and La Palma stations from 21/09/2021 to 21/01/2022. Details on aerosols in situ measurements are given in**  
 184 **Rodriguez et al. (submitted).**

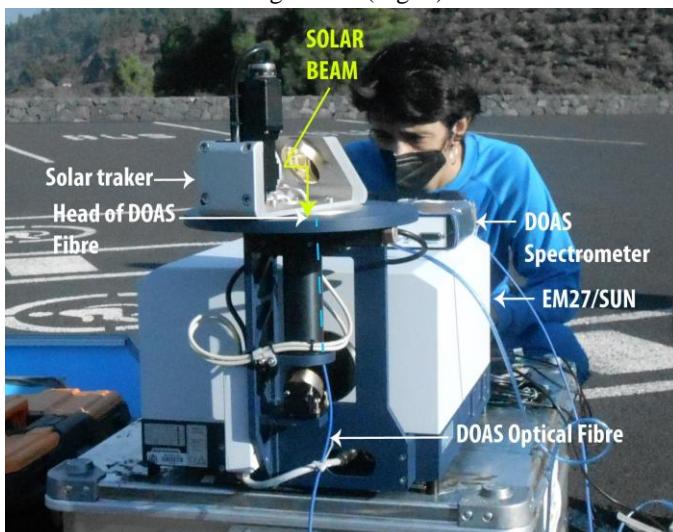
Station (Island) (geographical coordinates) altitude distance from the eruptive fissure	Instrument (Networks)	Measurement period	Fraction of measurement days capturing the volcanic plume (or post eruptive diffuse emissions)
FUE (La Palma) (28.49°N, 17.85°W) 630 m a.s.l. ~ 15 km	EM27/SUN#SN143 (COCCON)	25/09/2021 - 21/01/2022	21/59 (co-eruptive) 1/11 (post-eruptive)
	Combined EM27/SUN#SN143-DOAS	10/10/2021 - 10/12/2021	14/32 (co-eruptive)
IZO (Tenerife) (28.31°N, 16.50°W) 2373 m a.s.l. ~ 140 km	EM27/SUN#SN085 (COCCON)	20/09/2021 - 31/01/2022	4/38 (co-eruptive) 0/9 (post-eruptive)
	IFS-125HR (NDACC)	19/09/2021 - 31/01/2022	11/48 (co-eruptive) 0/13 (post eruptive)
	In situ UV fluorescence analyzers (SO <sub>2</sub> ) (GAW WMO network)	21/09/2021 - 31/12/2021	26/83 (co-eruptive) 1/16 (post eruptive)
	In situ Picarro (CO <sub>2</sub> , CO) (GAW WMO network)	19/09/2021 - 31/12/2021	26/85 (co-eruptive) 1/16 (post eruptive)
	Aerosol samplers	19/09/2021 - 31/12/2021	26/85 (co-eruptive)
El Paso (La Palma) (28.6590°N, 17.8481°W) 860 m.a.s.l.	Aerosol samplers	27/09/2021- 19/10/2021	18/22 (co-eruptive)
Los Llanos (La Palma) (28.6586°N, 17.913100°W 343 m.a.s.l.	Aerosol samplers	20/10/2021- 07/01/2022	52/55 (co-eruptive)

## 185 186 **2.1. The Fuencaliente station (FUE, La Palma Island)**

187 In the context of AEMET responsibilities, as a State Agency, for continuous monitoring of the  
 188 meteorological and climatic conditions and of atmospheric composition, a specific instrumental deployment has  
 189 been set up in La Palma. In particular, a new station for gas and particle monitoring was implemented at the San  
 190 Antonio Volcano visitors center of Fuencaliente, at the southern tip of La Palma Island, ~15 km from the  
 191 eruptive fissure of the Tajogaite volcano (Fig. 1). The FUE station included a wide range of instruments such as  
 192 a sun-lunar Cimel CE318T photometer, contributing to the Aerosol Robotic Network (AERONET), for aerosol

193 column measurements, a Lufft CHM15k ceilometer for aerosol and cloud vertical profiling and an all-sky  
194 camera for weather monitoring (Román et al., 2021) and a tephra trap.

195 A few days after the beginning of the eruption (on 25 September 2021), we deployed an EM27/SUN  
196 spectrometer (developed by the Karlsruhe Institute of Technology (KIT), in collaboration with Bruker Optics,  
197 Germany), which is the standard instrument of the Collaborative Carbon Column Observing Network  
198 (COCCON, Frey et al., 2019) dedicated to the measurement of greenhouse gases. This portable Fourier  
199 Transform Infrared (FTIR) spectrometer, equipped with a Quartz beamsplitter and two InGaAs photodetectors,  
200 provides low-spectral resolution ( $0.5 \text{ cm}^{-1}$ ) solar absorption spectra in the Near-Infrared (NIR) range (from 4000  
201 to  $11000 \text{ cm}^{-1}$ ), allowing the analysis of COCCON standard species ( $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{H}_2\text{O}$ ,  $\text{CH}_4$ ). It records double-  
202 sided forward-backward interferograms with a scanner velocity of 10 kHz and typically averages ten scans, so  
203 that a spectrum is acquired approximately every minute. The spectral range of this instrument also allows  
204 obtaining other gas species of interest for volcanology and air quality studies, such as halogen halides (HCl, HF)  
205 (Butz et al., 2017). From 10 October 2021 to 10 December 2021, following Butz et al. (2017) approach, we  
206 combined the EM27/SUN with a UV-Vis DOAS spectrometer (model Avantes ULS2048). The DOAS  
207 instrument had a  $50 \mu\text{m}$  wide slit entrance, and allows recording spectra in the 270-425 nm spectral range with a  
208 spectral resolution of 0.4 nm. We used a  $200 \mu\text{m}$  wide quartz-made optical fibre. Both instruments shared the  
209 incident sun radiation from the EM27/SUN solar tracker to add simultaneous measurements of  $\text{SO}_2$  with the  
210 same measurement configuration (Fig. 2).



211  
212 **Figure 2: Photograph of the combined EM27/SUN-DOAS direct-sun measurements set-up implemented at the FUE**  
213 **station during the Tajogaite eruption (La Palma Island). The DOAS optical fibre is introduced and attached in**  
214 **the FTIR sunlight collection tube, pointing towards the solar tracker mirrors. The yellow lines schematize the incident**  
215 **sunlight optical path. Photograph taken by R. Campion.**

216 The DOAS fibre was inserted and attached coaxially into the tube directing the light from the solar tracker  
217 toward the EM27/SUN spectrometer entrance (Fig. 2). By this way, it allows collecting the maximum light  
218 intensity with a minimal disturbance of the solar beam transmitted to the EM27/SUN. The fibre was connected  
219 to the DOAS spectrometer, installed in a protective case sheltered from solar radiation near the EM27/SUN  
220 instrument. DOAS direct-sun absorption spectra were routinely recorded using the MobileDOAS software  
221 (unpublished acquisition program developed for mobile DOAS measurements by C. Fayt and A. Merlaud from  
222 the BIRA-IASB institute) with an integration time of about 30 seconds, with on average 20 scans. The details of  
223 the spectral DOAS and EM27/SUN spectral analysis and retrievals are given in section 3.

## 224 **2.2. The Izaña Atmospheric Observatory (IZO, Tenerife Island)**

225 The proximity of the island of Tenerife to La Palma and the location of the IZO station in the free  
226 troposphere (2373 m.a.s.l) resulted in this international reference observatory to be affected several times by the  
227 Tajogaite volcanic plume. This allowed a more in-depth study of various aspects of the volcanic eruption from a  
228 multi-instrumental perspective. Given its strategic location and its excellent atmospheric conditions, IZO indeed

229 has a comprehensive state of the art program for atmospheric composition measurements. Uninterrupted  
230 meteorological and climatological observations started in 1916 and, since 1984, IZO has contributed to the  
231 GAW-WMO (Global Atmosphere Watch, World Meteorological Organization) program and to multiple  
232 international networks and databases (WDCGG, WOUDC, NDACC, TCCON, COCCON, AERONET, BSRN,  
233 MPLNET, E-GVAP, NOAA/ESRL/GMD CCGG, etc.; Cuevas et al., 2024, and references therein). Within  
234 IZO's atmospheric research activities, the station is equipped with high-resolution IFS-125HR and low-  
235 resolution EM27/SUN FTIR spectrometers, which provide ongoing long-term solar absorption measurements  
236 since 1999 and 2018, respectively. The EM27/SUN spectrometer is the same instrument model as that  
237 implemented at the FUE station allowing the analysis of CO<sub>2</sub>, CO, HF and HCl species, as previously described.

238 The IZO FTIR spectrometers routinely contribute to the Network for the Detection of Atmospheric  
239 Composition Change (NDACC, <https://ndacc.larc.nasa.gov>, last access: March 2025), Total Carbon Column  
240 Observing Network (TCCON, <https://tccon-wiki.caltech.edu>, last access: March 2025), and COCCON  
241 (<https://www.imk-asf.kit.edu/english/COCCON.php>, last access: March 2025) (Schneider et al., 2005; García et  
242 al., 2021). As part of NDACC activities, direct solar mid-infrared (MIR) absorption spectra are measured in the  
243 range of 700 to 4500 cm<sup>-1</sup>, with a spectral resolution of 0.005 cm<sup>-1</sup>. NDACC operations involve co-adding  
244 several scans to increase the signal-to-noise ratio, resulting in each spectrum acquisition taking several minutes.  
245 García et al. (2021) provide further details about the IZO FTIR program. The IZO IFS-125HR MIR solar spectra  
246 were used to analyse the SO<sub>2</sub> species alongside HCl and HF, which were also measured from the EM27/SUN  
247 spectra (unlike SO<sub>2</sub>). This approach further allowed us to evaluate the uncertainties associated with our new  
248 retrieval methods for the HF and HCl species (see section 3 and Appendix A). The details of the spectral  
249 analysis and retrievals are given in section 3.

250 Moreover, as part of the GAW-WMO program, continuous surface measurements of CO<sub>2</sub> (since 1984),  
251 SO<sub>2</sub> (since 2006), and CO (since 2008) are performed at IZO. Different in situ analyzers and measurement  
252 techniques have been used for measuring these gases: CO<sub>2</sub> with non-dispersive infrared (NDIR) gas Licor  
253 analyzers, CO with gas chromatography (GC) Trace Analytical RGA-3 instruments and SO<sub>2</sub> with ultraviolet  
254 (UV) fluorescence analyzers (Thermo 43C-Trace Level). Since 2015, CO<sub>2</sub> and CO have also been monitored  
255 using a cavity ringdown spectroscopy (CRDS)-based Picarro G2401 instrument. These observations are carried  
256 out following the strict GAW-WMO measurement protocols and their quality is periodically assessed by  
257 external audits by the World Calibration Center for surface Ozone, CO, Methane and CO<sub>2</sub> (WCC-Empa). The  
258 bias for the CO<sub>2</sub> and CO measurements in the frame of the GAW-WMO network is ± 0.1 ppm and ± 2 ppb,  
259 respectively (WMO, 2018). For SO<sub>2</sub>, the uncertainties are expected to be around ± 0.2 ppb (manufacturer  
260 specifications; see also Cuevas et al., 2024 and references therein). This continuous gas monitoring captured the  
261 Tajogaite plume composition on several occasions, when meteorological conditions allowed rapid and direct  
262 transport to the IZO station.

### 263        2.3. Retrieval of SO<sub>2</sub> volcanic emission fluxes from TROPOMI data

264        The SO<sub>2</sub> flux was retrieved by processing the images of the TROPOMI hyperspectral UV-SWIR sensor  
265 on-board the Sentinel-5P satellite. The images were processed by the traverse method, initially developed for the  
266 coarser resolution TOMS satellite images by Bluth et al. (1994) and later adapted to more recent sensors such as  
267 OMI and TROPOMI. The traverses are drawn across the plume semi automatically and the SO<sub>2</sub> flux is  
268 calculated using the equation:

$$269 \quad F = \sum X_i * L_i * \sin(\theta) * v$$

270 where X<sub>i</sub> is the SO<sub>2</sub> Vertical Column Density (VCD), L<sub>i</sub> is the length of the pixel, θ is the angle between the  
271 pixel row and the wind direction, and v is the plume transport speed. The SO<sub>2</sub> VCD was interpolated at plume  
272 height between the SO<sub>2</sub>\_1km and the SO<sub>2</sub>\_7km subproducts of the version 3 of the TROPOMI SO<sub>2</sub> product,  
273 described in Theys et al. (2021). The plume speed was obtained from the Global Data Assimilation System  
274 model of the NOAA, through the READY Archived Meteorology portal  
275 (<https://www.ready.noaa.gov/index.php>). For the flux calculation, we used the average wind speed at the plume  
276 altitude over the analysed plume portion. The plume altitude was estimated from visual observations such as  
277 photographs, distal webcam images (from Roque de los Muchachos) and HYSPLIT trajectory simulations,

278 picking the injection altitude that best reproduces the general plume direction observed on the TROPOMI  
279 image, and confirmed with the AEMET/IGN estimates for the coincident days. The SO<sub>2</sub> fluxes were finally  
280 estimated using the average of several traverses (usually a few tens and, in some occasions, up to two hundreds,  
281 depending on the coherence of the plume and the wind field that transports it). The traverse method does not  
282 work in cases of plume stagnation in a low wind environment and when the plume is split into several directions  
283 due wind shear. These situations happened during about 30% of the time of the eruption, causing some gaps in  
284 the SO<sub>2</sub> flux time series. We also excluded images where the plume was only partially captured.

## 285 **2.4 Mobile MultiGAS measurements**

286 During the eruptive period, mobile surface MultiGAS measurements (SO<sub>2</sub>, CO<sub>2</sub>, H<sub>2</sub>O, H<sub>2</sub>S) were  
287 carried out into the volcanic plume, between 28 September and 10 October 2021, when meteorological  
288 conditions allowed it to be sampled at ground level at a high concentration. The instrument comprises an  
289 MSR145 datalogger, an Edinburgh Gascard NG for CO<sub>2</sub> (0–1000 ppm) with a pump, a City Technology T3ST/F  
290 electrochemical sensor for SO<sub>2</sub> (0–50 ppm) and a City Technology T3H electrochemical sensor for H<sub>2</sub>S (0–20  
291 ppm). SO<sub>2</sub> concentrations up to 7 ppm were measured at distances of about 2 km, East and West of the vent  
292 (Fig. 1). Time series of concentrations of the different gas species were cross-correlated by adjusting the time-  
293 lag (usually between 5 and 9 seconds) and smoothing parameter until the best R-squared correlation coefficient  
294 was obtained. The measurements presented here have R-squared higher than 0.75.

## 295 **2.5 Sulfates aerosol measurements**

296 Samples of aerosols, or particulate matter (PM), smaller than 10  $\mu\text{m}$  (PM<sub>10</sub>) were collected at two sites  
297 in La Palma, at El Paso and at Los Llanos de Aridane and at IZO in Tenerife island. We used high volume  
298 samplers (30  $\text{m}^3 \cdot \text{h}^{-1}$ ) and quartz microfiber filters (150 mm diameter). Sulfate concentrations were determined  
299 by ion chromatography (Metrohm™ 930 Compact IC FLEX), after a leaching extraction in deionized milli-Q  
300 grade water of the sample by methods described in Rodríguez et al. (2012).

## 301 **2.6 Volcanic glass S, Cl and F contents and sulfide droplets composition**

302 We report 14 new compositions of MIs hosted in olivine, clinopyroxene and amphibole (kaersutite)  
303 crystals (Appendix B3; Supplementary Table). We also report Cl, F, and S contents in tephra glasses that were  
304 measured alongside major elements during the analytical session described in Gonzalez-Garcia et al. (2023),  
305 although only the major element data were published in that study. The volatiles were analysed using a Cameca  
306 SX-100 electron microprobe (EPMA) at the Department of Geosciences of the University of Bremen  
307 (Germany), with an acceleration voltage of 15 kV, beam current of 40 nA and defocused beam of 10  $\mu\text{m}$ ,  
308 following the methods described in Gonzalez-Garcia et al. (2023). The instrument was calibrated with a natural  
309 fluorite for F, pyrite for S, and Smithsonian scapolite for Cl. Counting times on peak were 120 s for F and 60 s  
310 for S and Cl. The analyses of F used the PHA (pulse height analysis) setting after Zhang et al. (2016); the  
311 interference of the FeL $\alpha$  line on the FK $\alpha$  peak was corrected using the overlay function of the Cameca software.  
312 The Smithsonian reference materials VG-2 glass, VG-A99 glass and Kakanui hornblende (Jarosewich et al.,  
313 1980) were analyzed along with the samples for precision and accuracy control. Accuracy is better than 6% for  
314 S and Cl and >20% for F; reproducibility is typically better than 10%. In addition, the composition of two  
315 sulfide droplets was semiquantitatively estimated by EDX (energy-dispersive X-ray) spectroscopy.

316 A Scanning Electron Microscope (SEM) was used to obtain high-resolution back-scattered electron  
317 (BSE) images of two sulfide droplets found in the tephra sample LM-2309 (Las Manchas, 23 September). The  
318 BSE images were acquired using a JEOL JSM-7610F gun emission scanning electron microscope installed at  
319 the Institute of Earth System Sciences, Leibniz Universität Hannover, Germany, using an accelerating voltage of  
320 15kV and a working distance of 15 mm. Bruker ESPRIT software was used for image acquisition.

## 321 **3. FTIR and DOAS analysis: Specific SO<sub>2</sub>, HCl, HF, CO<sub>2</sub> and CO retrievals**

### 322 **3.1. Spectral analysis from the combined EM27/SUN-DOAS system**

323                    **3.1.1 EM27/SUN retrievals (CO<sub>2</sub>, CO, HCl, HF)**

324                    The processing of EM27/SUN measurements was performed using the open-source PROFFAST  
325                    pylev1.2 packages developed by the KIT and used by the COCCON community. The COCCON standard  
326                    retrieval procedure used for the analysis of atmospheric CO<sub>2</sub>, CO, CH<sub>4</sub> and H<sub>2</sub>O species is fully described in  
327                    Frey et al. (2019), Alberti et al. (2022), Herkomm (2024a,b) and Feld et al. (2024). Here, we provide details  
328                    only on the specific retrieval strategies that we developed for volcanological applications. The PROFFAST  
329                    package includes a preprocess code generating the required spectra by a Fast Fourier Transform. The processing  
330                    incorporates various quality checks, as a signal threshold, intensity variations during recording, requirement of  
331                    proper spectral abscissa scaling, and generates spectra only from raw measurements passing all checks (the  
332                    remaining ones being flagged). We used the Instrumental Line Shape (ILS) parameters reported in Alberti et al.  
333                    (2022) following the COCCON standard recommendations. Calibrated spectra are then analyzed using the  
334                    PROFFAST radiative transfer and inversion models to derive the total columns by scaling the a priori Volume  
335                    Mixing Ratio (VMR) profiles iteratively until adjusting the simulated spectra to the measured spectra. Surface  
336                    pressures are derived from the in situ high precision sensor measurements (PCE-THB-40 at FUE and SETRA-  
337                    470 at IZO). All the EM27/SUN retrievals presented in this study were performed using the HITRAN 2020  
338                    spectroscopic linelists (Gordon et al., 2022). We used meteorological data and a priori VMR profiles based on  
339                    the sub-daily available GGG2020 TCCON meteorological data (MAP files downloaded from the Caltech server  
340                    and based on National Centers for Environmental Prediction (NCEP) reanalysis). We adapted the a priori VMR  
341                    profiles for the target species depending on whether the gas is purely volcanic (low atmospheric abundance) or  
342                    also has an atmospheric background. The spectral windows and retrieval strategies used for each species are  
343                    presented in Table 2 and detailed below.

344                    For the analysis of HCl and HF species, we utilized a priori VMR profiles with high concentrations  
345                    ( $1 \times 10^{-4}$  ppm) up to the altitude of the volcanic plume (~6 km a.s.l., based on IGN/AEMET; Milford et al.,  
346                    2023), and VMR concentrations for the upper levels derived from the Whole Atmosphere Community Climate  
347                    Model (WACCM v.6, <https://www2.acom.ucar.edu/gcm/waccm>, last access: february 2025) average profiles  
348                    provided by the National Center for Atmospheric Research (NCAR; James Hannigan, personal communication,  
349                    2014), which are commonly used by the NDACC community. In this case, we adapted the PROFFAST retrieval  
350                    inputs so that only the tropospheric portion (up to the altitude of the volcanic plume) was scaled, keeping the  
351                    stratospheric part as constant. This approach was previously employed to measure volcanic emissions of HCl  
352                    and HF from Mt. Etna, also relying on low-resolution EM27/SUN spectra (Butz et al., 2017), but utilizing the  
353                    PROFFIT package for the retrieval. We used new specifically optimised spectral windows (Table 2, HCl\_v2 and  
354                    HF\_v2) for the analysis of these two species to be able to detect even very low concentrations, as those detected  
355                    at the IZO station, 140 km from the eruptive fissure. The analysis was also conducted using the same spectral  
356                    ranges as Butz et al. (2017) (HCl\_v1 and HF\_v1 in Table 2) to evaluate the consistency and improvements  
357                    introduced by the new strategies for our application. Appendix A gives a full comparison between the results  
358                    obtained using the new and Butz et al. (2017) retrievals, as well as with those from the high-resolution spectra  
359                    analysis (see section 3.2) for side-by-side measurements.

360                    For the retrieval of volcanic CO and CO<sub>2</sub>, due to their high atmospheric abundance and variability, we  
361                    used the COCCON standard retrievals (scaling of the whole profile and use of the COCCON spectral windows  
362                    and TCCON priori VMRs) and then removed the atmospheric background to derive the volcanic contribution.  
363                    The column-averaged dry-air mole fraction of CO<sub>2</sub> and CO (XCO<sub>2</sub> and XCO) were estimated using the O<sub>2</sub> total  
364                    columns according to Wunch et al. (2011) ( $X_{gas} = 0.2095 \times Col\ gas \div Col\ O_2$ ) after applying air mass  
365                    independent and dependent correction factors (AICF and ADCF). We have slightly modified the standard  
366                    procedure for performing the O<sub>2</sub> retrieval by adding HF as species to be retrieved, using a specific a priori VMR  
367                    profile based on the WACCM v.6 climatology. However, the HF profile was adjusted to have a constant and  
368                    significantly higher concentration ( $1 \times 10^{-4}$  ppm) up to the maximum plume altitude. For the other interfering  
369                    gases, we used the a priori VMRs derived from the TCCON GGG2020 MAP files.

370                    To remove the background atmospheric concentrations of XCO<sub>2</sub> and XCO, we used the daily-averaged  
371                    IZO X<sub>gas</sub> time series to model the long-term natural variability with a third-degree polynomial, which was then  
372                    interpolated and subtracted from the FUE XCO<sub>2</sub> and XCO time series. Examples of XCO<sub>2</sub> and XCO background

373 fits are given in Fig. A3 and A4, respectively. For CO<sub>2</sub>, an additional intraday variability had to be taken into  
 374 account. It was simulated by averaging and fitting some intraday IZO XCO<sub>2</sub> time series which were not affected  
 375 by the volcanic plume. Intraday simulations were performed for each day, using the average fit and adjusting the  
 376 offset. The accuracy of the method was assessed by comparing the simulated XCO<sub>2</sub> background at the station  
 377 impacted by the volcanic plume with the measured XCO<sub>2</sub> background at the other station when it was not  
 378 affected by the plume (Fig. A3). The average and maximum absolute difference arising from this procedure  
 379 were found to be 0.1 and 0.8 ppm in extreme cases. Finally, the  $\Delta$ CO<sub>2</sub> and  $\Delta$ CO volcanic enhancements were  
 380 determined from the X<sub>gas</sub> enhancements by multiplying them by the dry air columns derived from the surface  
 381 pressure measurements and H<sub>2</sub>O total columns (Wunch et al., 2011).

382 **Table 2: Retrieval parameters used for the EM27/SUN and DOAS spectral analysis. “Sim” corresponds to the**  
 383 **interfering species only considered for the forward simulations. “\*\*” refers to similar spectral windows as Butz et al.**  
 384 **(2017).**

Gas	Instrument	Spectral Window (cm <sup>-1</sup> )	Interfering Gases	Strategy
HCl_v1 HCl_v2	EM27/SUN	5684.0 - 5795.0* 5703.5 - 5779.0	H <sub>2</sub> O, HDO, CH <sub>4</sub> H <sub>2</sub> O, HDO, CH <sub>4</sub>	High ( $1 \times 10^{-4}$ ppm) a priori HCl VMR between 0 - 5.8 km beyond: WACCM v.6
HF_v1 HF_v2	EM27/SUN	7765.0 - 8005.0* 3995.0 - 4043.0	H <sub>2</sub> O, CO <sub>2</sub> (Sim), O <sub>2</sub> H <sub>2</sub> O, HDO, CH <sub>4</sub>	High ( $1 \times 10^{-4}$ ppm) a priori HF VMR between 0 - 5.8 km beyond: WACCM v.6
CO <sub>2</sub>	EM27/SUN	6173.0 - 6390.0	H <sub>2</sub> O, CH <sub>4</sub> (Sim)	COCCON + post-process background correction
CO	EM27/SUN	4208.7 - 4318.8	H <sub>2</sub> O, HDO, CH <sub>4</sub> , N <sub>2</sub> O (Sim), HF (Sim)	COCCON + post-process background correction
O <sub>2</sub>	EM27/SUN	7765.0 - 8005.0	H <sub>2</sub> O, CO <sub>2</sub> (Sim), HF	High ( $1 \times 10^{-4}$ ppm) a priori HF VMR between 0 - 5.8 km beyond: WACCM v.6
SO <sub>2</sub>	direct-Sun DOAS	312.0 nm – 326.8 nm	O <sub>3</sub>	Levenberg-Marquardt (LM) algorithm

385  
 386

### 3.1.2 DOAS retrievals (SO<sub>2</sub>)

387 Solar DOAS spectra were processed using the QDOAS v2.111 software (Dankaert et al., 2014),  
 388 applying a Levenberg-Marquardt (LM) algorithm to retrieve the Slant Column Densities. We used the same  
 389 analysis strategy as described in Taquet et al. (2023), with the key parameters summarized in Table 2.  
 390 Wavelength calibration and slit function were determined by laboratory close-path measurement using a low-  
 391 density mercury lamp, and further adjusted based on the position and widening of the Fraunhofer lines during  
 392 the QDOAS processing. SO<sub>2</sub> was retrieved in the 312.0–326.8 nm spectral window according to Butz et al.  
 393 (2017). The high resolution solar spectrum from Chance and Kurucz (2010) was used as the reference spectrum.  
 394 We used the cross-section at 298 K from Vandaele et al. (2009) for SO<sub>2</sub> and the cross-section at 221 K from  
 395 Burrows et al. (1999) for the interfering gas O<sub>3</sub>. A third-order polynomial function was included in the fitting  
 396 routine to remove the broadband extinction. The I0 effect, due to the limited resolution of the spectrometers  
 397 (Platt et Stutz, 2008), was corrected using the QDOAS I0-correction algorithm applied for six fixed SO<sub>2</sub> slant  
 398 column values of 0.0,  $1.0 \times 10^{18}$ ,  $2.0 \times 10^{18}$ ,  $3.0 \times 10^{18}$ ,  $4.0 \times 10^{18}$ ,  $5.0 \times 10^{18}$  molec/cm<sup>2</sup> (the latter is close to the  
 399 maximum uncorrected slant column). Then, each corrected value is determined by interpolating the corrected

400 slant columns values. Unlike radiance scattered light measurements, the direct-sun configuration remains  
 401 unaffected by the Ring effect (Herman et al., 2009), which therefore was not considered in the retrieval. Finally,  
 402  $\text{SO}_2$  slant columns were converted into vertical columns by dividing them by the SZA-dependent air mass factor  
 403 (1/cos (SZA)) to be combined with the FTIR data.

#### 404 3.2 IFS-125HR analysis (HCl, HF and $\text{SO}_2$ )

405 The HCl and HF retrieval strategy from the IFS-125HR spectra is based on the NDACC-IRWG  
 406 recommendations (Infrared Working Group, IRWG, 2014), and on the adapted retrievals for volcanological  
 407 applications reported in Taquet et al. (2019) and Stremme et al. (2023). However, they have been optimised here  
 408 to properly capture tropospheric volcanic contributions up to 140 km from the eruptive fissure. Consistently  
 409 with the NDACC approach, both species were retrieved using the non-linear least-squares fitting algorithm  
 410 PROFFIT (Profile Fit, Hase et al., 2004), and considering the specified spectral regions and interfering gases  
 411 given in Table 3. The inversion procedure is solved using a first-order Tikhonov–Phillips regularization (L1,  
 412 Rodgers, 2000) on a logarithmic scale, where the VMR a priori profiles for the interfering gases are taken from  
 413 WACCM v.6 climatological profiles. The NCEP 12:00 UTC daily temperature and pressure profiles are  
 414 employed for the radiative transfer simulations.

415 The most significant changes with respect to NDACC involved the a priori VMR profiles considered  
 416 for the target gases, vertical L1 regularization, and the spectroscopic database. Similarly to the EM27/SUN  
 417 analysis, we adopt modified HF and HCl a priori VMR profiles with high concentrations ( $1 \times 10^{-4}$  ppm) up to the  
 418 maximum plume altitude (~6 km a.s.l.), which are completed for the IFS-125HR using WACCMv.6 information  
 419 beyond this altitude. In addition, the 2020 HITRAN spectroscopic linelists were utilized for all gases. Finally, in  
 420 contrast to the NDACC approach, where the lowermost and uppermost altitude levels are fixed to the a priori to  
 421 ensure stability in the retrieval, in this study, the first level is left unconstrained to provide flexibility in the  
 422 retrieval process in the lower troposphere.

423 In the case of  $\text{SO}_2$ , a harmonized and standardized FTIR strategy is not available within NDACC.  
 424 Therefore, in this work, we employ the strategy developed by García et al. (2022), which has been successfully  
 425 applied to various NDACC FTIR sites affected by volcanic  $\text{SO}_2$  emissions (Smale et al., 2023; García et al.,  
 426 2025). This approach is based on the study by Taquet et al. (2019), which presents  $\text{SO}_2$  total column amounts  
 427 from the measured solar absorption spectra in the  $2500 \text{ cm}^{-1}$  region using a scaling retrieval and the inversion  
 428 code PROFFIT. Similarly to HF and HCl volcanic products, the  $\text{SO}_2$  a priori VMR profiles are adapted in the  
 429 lower troposphere, while climatological WACCMv.6 profiles are considered for all interfering gases (Table 3).  
 430 Appendix A provides a summary of the comparison between the standard NDACC HCl and HF products and  
 431 those developed in this study, the new IFS-125HR  $\text{SO}_2$  retrievals, as well as the comparison between all the IFS-  
 432 125HR and EM27/SUN products.

433 **Table 3: Retrieval parameters used for the IFS-125HR analysis. “Sim” corresponds to the interfering species only**  
 434 **considered for the forward simulations. The spectral windows are acquired using the NDACC filter SC (S3) for HCl,**  
 435 **with the NDACC filter SA (S1) for HF, and with the NDACC filter SF (S6) for  $\text{SO}_2$ . Therefore, they are almost**  
 436 **coincident, but not simultaneous observations.**

Gas	Spectral Window ( $\text{cm}^{-1}$ )	Interfering Gases	Strategy
HCl	2727.73-2727.83 2775.60-2775.90 2821.40-2821.75 2925.75-2926.10	$\text{H}_2\text{O}$ (Sim), $\text{HDO}$ (Sim), $\text{O}_3$ , $\text{CH}_4$ (Sim), $\text{OCS}$ , $\text{NO}_2$ , $\text{N}_2\text{O}$ (Sim)	High ( $1 \times 10^{-4}$ ppm) HCl a priori VMR between 0 - 5.6 km, above: WACCM v.6
HF	4000.90-4001.05 4038.85-4039.08	$\text{H}_2\text{O}$ , $\text{O}_3$ (Sim), $\text{CH}_4$ (Sim)	High HF ( $1 \times 10^{-4}$ ppm) a priori VMR between 0 - 5.6 km, above: WACCM v.6
$\text{SO}_2$	2480.00-2520.00	$\text{H}_2\text{O}$ , $\text{CO}_2$ , $\text{O}_3$ , $\text{CH}_4$ , $\text{N}_2\text{O}$	High $\text{SO}_2$ ( $1 \times 10^{-2}$ ppm) a priori VMR between 0 - 5.6 km, above: WACCM v.6

437

## 4. Results

438

### 4.1. Evolution of the volcanic plume composition during the Tajogaite eruption

439

The temporal variability of the Tajogaite plume composition is examined through the time series of the ratios, some of them involving species with contrasting exsolution depths. Daily  $\Delta\text{CO}_2/\text{SO}_2$ ,  $\text{HCl}/\text{SO}_2$ ,  $\text{HF}/\text{SO}_2$ ,  $\text{HCl}/\Delta\text{CO}_2$ ,  $\text{HF}/\Delta\text{CO}_2$ ,  $\Delta\text{CO}/\text{SO}_2$  and  $\Delta\text{CO}/\Delta\text{CO}_2$  molecular ratios were estimated from the daily correlation plots of the total column time series, following the methodology as detailed in Taquet et al. (2019, 2023) and are reported in Fig. 3. The same method used for column-averaged ratios was applied to calculate the surface concentration ratios from GAW and MultiGAS measurements (also presented in Fig. 3). The background contribution of atmospheric species ( $\text{CO}_2$  and  $\text{CO}$ ) to these measurements was removed using daily polynomial curves fitted from the surface measurements without contribution of volcanic emissions (i.e.  $\text{SO}_2 < 0.05$  ppm). Additionally, we reported in the same figure our MultiGAS  $\Delta\text{CO}_2/\text{SO}_2$  measurements, obtained on 29 September, 2 and 7 October from Las Manchas (~500 m a.s.l., SW from the eruptive fissure, Fig. 1) and from the El Jable viewpoint (2100 m a.s.l., E of the eruptive fissure, Fig. 1), ranging between 1.7 and 14.3. The scarcity of FTIR measurements from early November until the end of the eruption, across all measurement techniques, is mainly due to poor or unsuitable weather conditions.

452

Our column-averaged  $\Delta\text{CO}_2/\text{SO}_2$  molecular ratios range between  $9 \pm 6$  and  $63 \pm 28$  (9-24 at IZO and 14-63 at FUE) during the eruption. These values are consistent with the surface measurements at IZO (ratios from  $5.6 \pm 0.1$  to  $18.3 \pm 0.7$ ) and with our MultiGAS measurements at La Palma (1.7 to 14.3). These values are also consistent with the proximal measurements reported in the literature including Open Path FTIR (Burton et al., 2023 and Asensio-Ramos et al., 2025) and MultiGAS (Burton et al., 2023; Erickson et al., 2024) measurements, ranging between 2 and 52 (shaded area in Fig. 3). All the measured  $\Delta\text{CO}_2/\text{SO}_2$  ratios define an increasing trend, at least until 2 November 2021 and show more scatter after this date (Fig. 3).

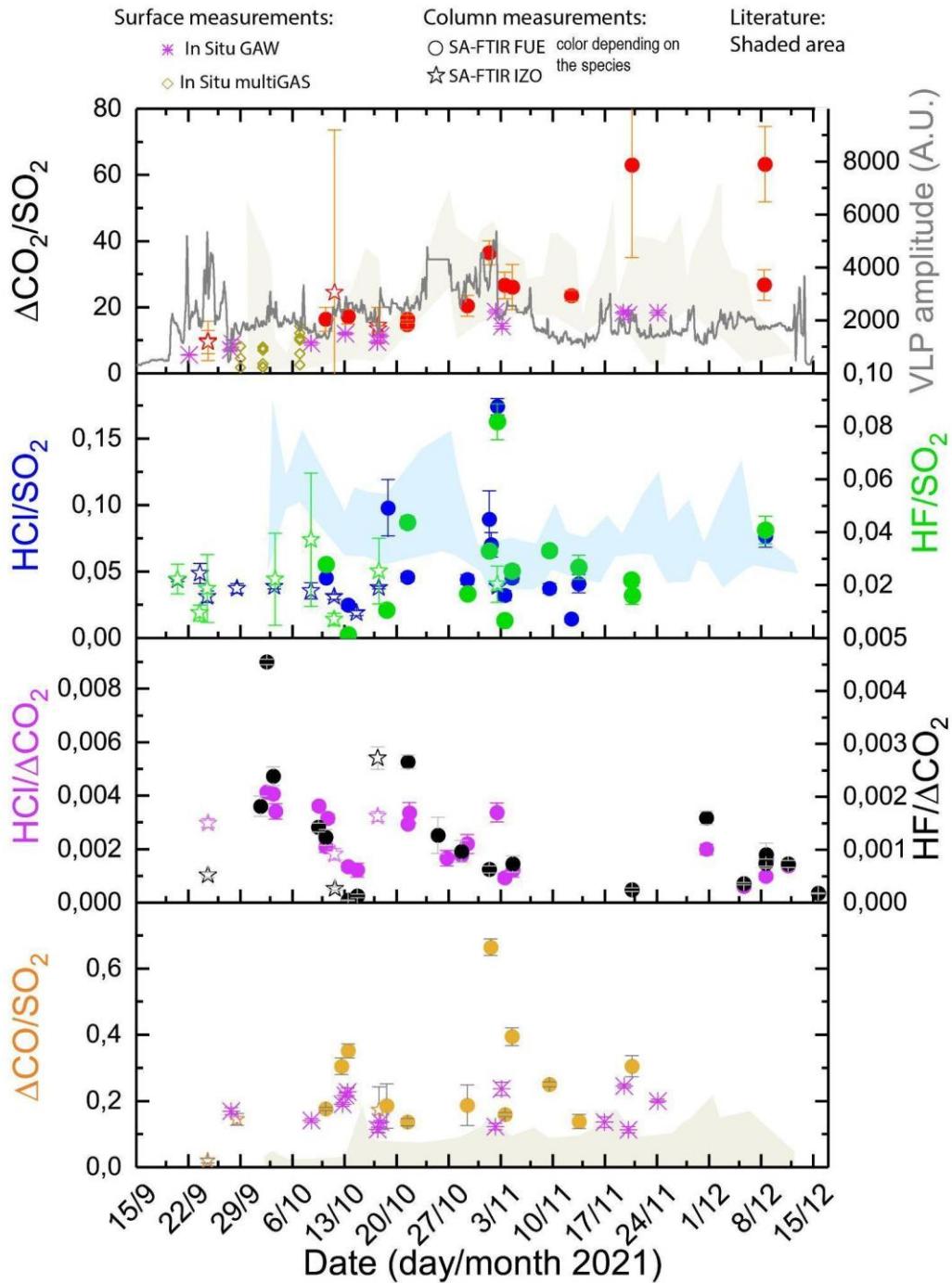
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$\text{HCl}/\text{SO}_2$  molecular ratios range between  $0.02 \pm 0.002$  and  $0.17 \pm 0.01$  (from 0.02 to 0.05 at IZO and from 0.02 to 0.17 at FUE) and show short-term variations around a nearly constant daily average of ( $0.05 \pm 0.03$ ) throughout the entire eruptive period. These ratios are consistent with the values of  $\text{SO}_2/\text{HCl}$  of 16.8 and 8 ( $\text{HCl}/\text{SO}_2 = 0.06$  and 0.12, respectively) reported in Burton et al. (2023), which corresponds to a lava fountaining plume and spattering event (Fig. 3). It is also consistent with the more recently published ratios ranging between 0.04 and 0.2 (Asensio-Ramos et al., 2025; Fig. 3).  $\text{HF}/\text{SO}_2$  molecular ratios vary between  $0.0012 \pm 0.0002$  and  $0.081 \pm 0.007$  (from  $0.001 \pm 0.001$  to  $0.082 \pm 0.007$  at FUE and from  $0.007 \pm 0.002$  to  $0.037 \pm 0.025$  at IZO) and show a similar day-to-day variability to that observed for the  $\text{HCl}/\text{SO}_2$  ratios through the eruptive period.  $\text{HCl}/\Delta\text{CO}_2$  molecular ratios exhibit values from  $(6 \pm 1) \times 10^{-4}$  and  $(4.1 \pm 0.1) \times 10^{-3}$  at FUE and from  $(2 \pm 1) \times 10^{-3}$  to  $(3 \pm 1) \times 10^{-3}$  at IZO, while the  $\text{HF}/\Delta\text{CO}_2$  ratios range from  $(0.5 \pm 0.1) \times 10^{-4}$  to  $(4.5 \pm 0.1) \times 10^{-3}$  at FUE and from  $(2.6 \pm 0.3) \times 10^{-4}$  to  $(2.7 \pm 0.2) \times 10^{-3}$  at IZO. Like  $\text{HCl}/\text{SO}_2$  and  $\text{HF}/\text{SO}_2$ , the  $\text{HCl}/\Delta\text{CO}_2$  and  $\text{HF}/\Delta\text{CO}_2$  ratios exhibit similar day-to-day variability. Their fluctuations include short-term decreasing trends, as observed between 2 and 14 October 2021 and between 21 October and 4 November 2021. The  $\Delta\text{CO}/\text{SO}_2$  FTIR ratios span from  $0.13 \pm 0.01$  to  $0.66 \pm 0.03$  at FUE and from  $0.02 \pm 0.01$  to  $0.17 \pm 0.07$  at IZO, and are relatively stable around the average of 0.24 with one extreme event, observed between 1 and 4 November 2021.

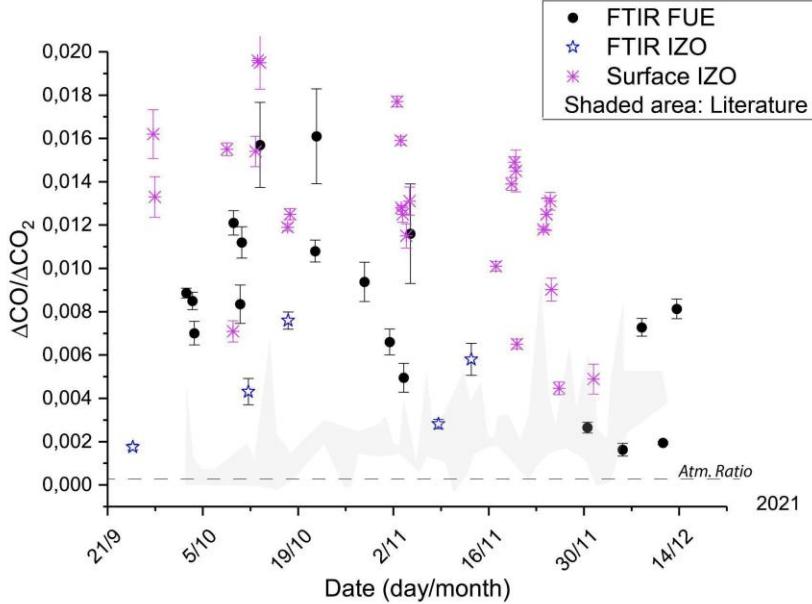
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During the initial phase of the eruption, prior to the eruptive pause on 27 September 2021, our ratios were comparable to those observed throughout the rest of the eruptive period, with  $\Delta\text{CO}_2/\text{SO}_2$  ranging between  $5.6 \pm 0.1$  and  $9 \pm 1.1$ ,  $\text{HCl}/\text{SO}_2$  between  $0.031 \pm 0.005$  and  $0.049 \pm 0.007$ , and  $\text{HF}/\text{SO}_2$  between  $0.009 \pm 0.003$  and  $0.022 \pm 0.006$ . A significant and abrupt increase in all species-to- $\text{SO}_2$  ratios is observed on 2-3 November 2021, which also coincides with a minor peak in the  $\text{HCl}$  and  $\text{HF}$ -to- $\Delta\text{CO}_2$  ratios. This event represents a notable and enduring change in gas ratio variability involving  $\text{CO}_2$ , (i.e.  $\Delta\text{CO}_2/\text{SO}_2$  and  $\text{HCl}/\Delta\text{CO}_2$ ) and coincides with a sudden decrease in the amplitude of seismic tremor (VLP and LP, Fig. 3 and Bonadonna et al., 2022). Prior to this date, the variability in the  $\Delta\text{CO}_2/\text{SO}_2$  ratio closely followed the increasing trend of VLP tremor amplitude, while afterwards it declined and exhibited a noticeable short-term variability until the end of the eruption. This noticeable change depicts two periods in our dataset (here after phase I and II), whose relationship with the previously described events and timeframes of the eruption (Bonadonna et al., 2022; Ubide et al., 2023; Milford et al., 2023) will be discussed in section 5. For  $\text{HCl}/\Delta\text{CO}_2$  and  $\text{HF}/\Delta\text{CO}_2$ , the ratios are

486 significantly lower during phase II (average of  $0.0012 \pm 0.0005$  and  $0.0007 \pm 0.0004$ , respectively) than during  
 487 phase I (average of  $0.0027 \pm 0.0009$  and  $0.0014 \pm 0.001$ , respectively). For other species, only a brief spike is  
 488 noted at this time, with ratios returning to Phase I levels at the onset of Phase II.



489  
 490 Figure 3: Variability of the Tajogaite volcanic plume composition during the eruption. Daily molecular ratios are  
 491 calculated from the daily species-to-SO<sub>2</sub> or species-to-CO<sub>2</sub> correlation plots of the total columns (SA: solar absorption  
 492 FTIR and DOAS measurements) and surface (GAW and MultiGAS analysis) time series. Only the ratios with a  
 493  $R^2 > 0.6$  in the correlation plots are reported here to exclude those with poor reliability. Data from literature is  
 494 presented as shaded areas, including the ratios reported by Burton et al. (2023), Erickson et al. (2024) and Asensio-  
 495 Ramos et al. (2025). The latter were derived from MultiGAS and Open-Path FTIR measurements. Very Long Period  
 496 (VLP; 0.4-0.6Hz) tremor amplitude (upper panel, gray line) is taken from Bonadonna et al. (2022).



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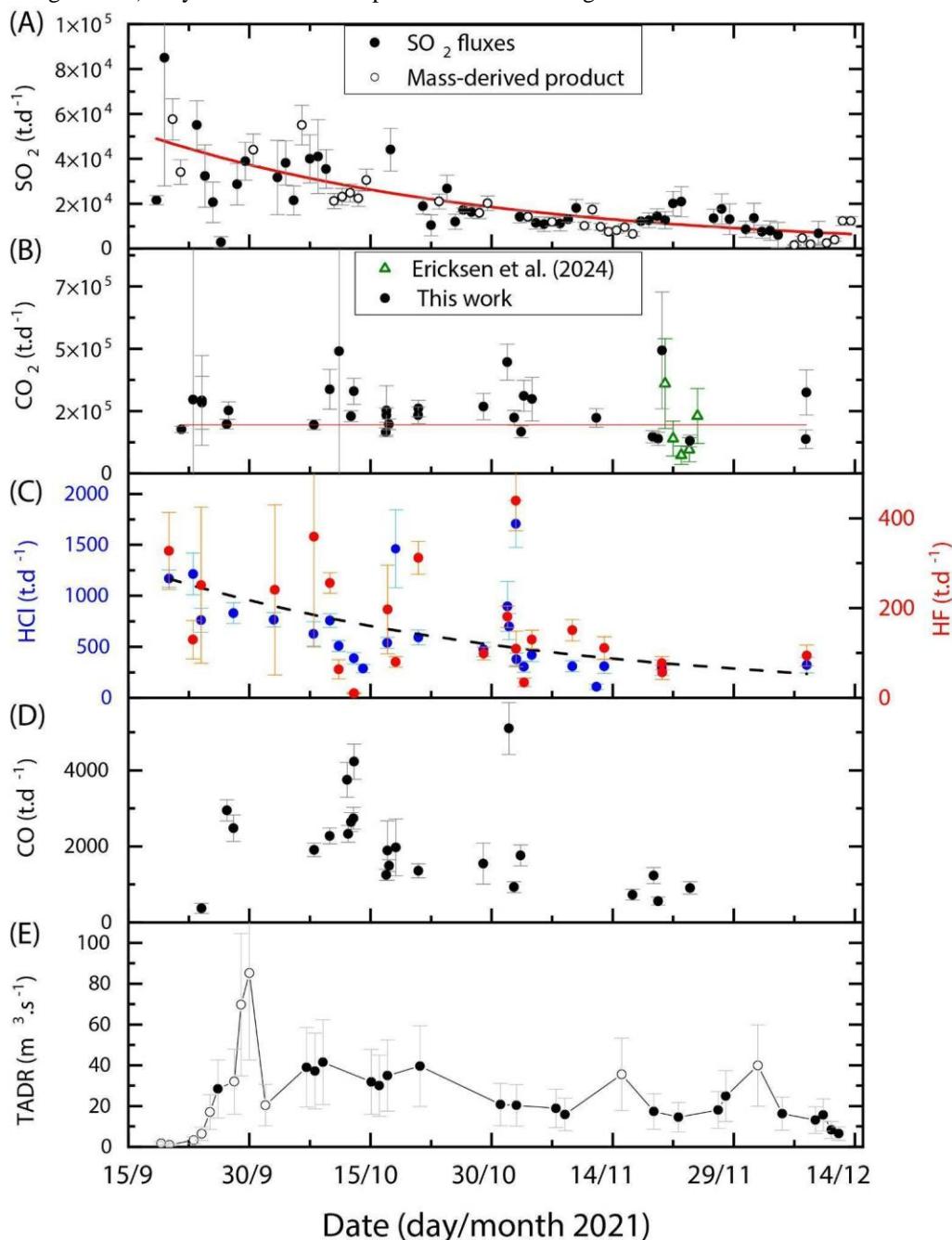
Figure 4: Time series of the  $\Delta\text{CO}/\Delta\text{CO}_2$  ratio at both FUE and IZO stations. The ratios at IZO presented here are derived from in-situ (purple) and FTIR (blue stars) measurements. Shaded areas present the data from the literature including Álvarez et al. (2023) measured by solar absorption FTIR and Asensio-Ramos et al. (2025) derived from OP-FTIR measurements. The dashed black line represents the long term atmospheric ratio (Atm. Ratio) measured at IZO (derived from Garcia et al., 2022).

503 Figure 4 presents the time series of  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios derived from FTIR solar absorption  
504 measurements at the FUE and IZO stations throughout the eruption, alongside with in situ surface measurements  
505 at IZO (GAW data). The  $\Delta\text{CO}/\Delta\text{CO}_2$  values observed at both sites and using both techniques are of the same  
506 order of magnitude, and exceed by more than one order of magnitude the average atmospheric background ratio  
507 at IZO ( $\sim 0.0002$ ). At FUE, the FTIR-derived ratios show a progressive increase from 0.0016 to 0.016 during the  
508 first 30 days of the eruption, followed by a decrease to lower values before mid-November. The surface  
509  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios at IZO fall within a similar range to those derived from FTIR at FUE, with some coinciding  
510 values in very good agreement. On average, the surface ratios at IZO are higher than the FTIR-derived ones at  
511 the same site. This discrepancy may be explained not only by the strong short-term variability in the  $\Delta\text{CO}/\Delta\text{CO}_2$   
512 ratios (only a few data points are coincident), but also by the fact that, although all these points coincide with the  
513 presence of  $\text{SO}_2$  (indicating the presence of volcanic plume), the correlation between  $\Delta\text{CO}$  and  $\text{SO}_2$  is relatively  
514 weak ( $R^2 < 0.6$ ), suggesting additional sources contributing to the CO enhancements. Furthermore, satellite  
515 imagery suggests that, on these days, the line of sight of the IZO FTIR instrument may have intersected aged  
516 volcanic plumes, potentially altering the retrieved  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios due to both geometric and compositional  
517 effects. The difference between the surface  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios observed at FUE and IZO and those (shaded area)  
518 reported by Asensio-Ramos et al. (2025) is discussed in Section 5.

## 519 4.2. $\text{SO}_2$ , $\text{CO}_2$ and halogen-derived volcanic emission fluxes and total emissions

520  $\text{SO}_2$  volcanic emission fluxes were estimated whenever the weather conditions made it possible  
521 following the method described in section 2.3 and reported in Fig. 5. The  $\text{SO}_2$  volcanic emission fluxes retrieved  
522 during this eruption exhibited a remarkably strong correlation ( $R^2=0.92$ , Fig. D3) with the daily  $\text{SO}_2$  masses  
523 (taken from MOUNTS website; Valade et al., 2019). To fill the long-term gaps in our  $\text{SO}_2$  fluxes time-series, a  
524 less reliable mass-derived product was included, derived from the linear relation between the  $\text{SO}_2$  volcanic  
525 emission fluxes and daily mass (Fig. 5A, empty circles). This was only applied to days with minimal  
526 accumulation. The  $\text{SO}_2$  volcanic emission fluxes time series exhibit a decreasing exponential trend (red curve),  
527 with an equation of the form  $y = a \times e^{-bx}$  and a coefficient of determination  $R^2= 0.63$ . Most mass-derived  
528 products were found to closely follow the overall trend (Fig. 5A, red curve), indicating that, despite inherent  
529 uncertainties, these estimates are likely robust enough to assess long-term variability in this case study. This also

530 suggests that short-term variations in wind direction or partial plume coverage in satellite images (initial  
 531 filtering criteria) may have a limited impact on the observed global trend.



532  
 533 **Figure 5:** (A) to (D) Emission fluxes of  $\text{SO}_2$ ,  $\text{CO}_2$ ,  $\text{HCl}$ ,  $\text{HF}$  and  $\text{CO}$  and (E) corrected TADR, following Plank et al.  
 534 (2023), during the eruption. The thick red line in (A) is the exponential fit to the  $\text{SO}_2$  emission fluxes time series. The  
 535 red line in (B) is the linear regression for the dataset. The black dashed line in (C) is the exponential fit to the  $\text{HCl}$   
 536 time series. Black points in (E) are part of the TADR- $\text{SO}_2$  emission flux correlation.

537 Volcanic emission fluxes for the other species were estimated by using daily species-to- $\text{SO}_2$  ratios and  
 538 either (i) interpolating the exponentially decreasing fit of  $\text{SO}_2$  fluxes or (ii) performing a linear interpolation of  
 539 the  $\text{SO}_2$  emission fluxes time series. The  $\text{HCl}$ ,  $\text{HF}$ ,  $\text{CO}_2$  and  $\text{CO}$  volcanic emission fluxes are shown in Fig. 5 B-  
 540 D, concurrently with the Time-Averaged Discharge Rate (TADR, Fig. 5E) time series of Plank et al. (2023),  
 541 multiplied by a factor of 2, as suggested by the authors to take the underestimation of the lava volume into  
 542 account.

543 A significant observation is the long-term decrease in the volcanic emission fluxes of  $\text{SO}_2$ ,  $\text{HCl}$ ,  $\text{HF}$ ,  
 544 and  $\text{CO}$ , which aligns with the TADR trend throughout the eruption, in contrast to the nearly stable trend of

545 CO<sub>2</sub>. The similarity of the trends of the daily average SO<sub>2</sub> emission fluxes and the TADR is further supported by  
 546 an excellent correlation (Pearson coefficient R=0.94; see Fig. E1), defining a slope of 14.1±1.2 kg of SO<sub>2</sub> per  
 547 thermal m<sup>3</sup> of discharged lava (lava volumes estimated using the radiant flux). This relationship includes 21/27  
 548 of the available TADR-fluxes pairs) and is mainly valid from 7 October 2021 onwards (Fig. E2, full circles).  
 549 The points corresponding to the onset of the eruption (outliers represented as hollow circles in Fig. E2) have  
 550 either higher SO<sub>2</sub> fluxes for a given TADR until the 25/09 or higher discharge rates after the 27/09 eruptive  
 551 break and until 30/09 at least (the next pair is that of 07/10, belonging to the correlation).

552 Another important observation is that the SO<sub>2</sub> flux peak recorded during the first week of the eruption,  
 553 accounting for approximately 20% of the total SO<sub>2</sub> emissions, occurs during a period of apparently low TADR  
 554 and around ten days prior to the first peak with maximum values of TADR for the eruption. The relationship  
 555 between the SO<sub>2</sub> volcanic emission fluxes and the TADR is examined in the light of the petrological data in  
 556 section 5.

557 Furthermore, the early November peaks in the HF, HCl, and CO emission fluxes time series, which align with  
 558 those observed in several ratios time series (Fig. 3), correspond to the inflection point in the overall flux decline,  
 559 occurring near the end of Phase I, as defined by Milford et al. (2023). Since the CO<sub>2</sub> volcanic emission fluxes  
 560 appear to be nearly constant throughout the entire eruptive period, we can interpret the lower HCl and HF-to-  
 561 CO<sub>2</sub> ratios of phase II as the result of globally lower fluxes during this period, in line with the pressure decrease  
 562 in the reservoir (Charco et al., 2024).

563 Table 4 presents the average volcanic emission fluxes for each species over the entire eruption distinguishing  
 564 between the results from the two previously described methods. Total emissions were estimated by combining a  
 565 Monte Carlo approach to account for uncertainties with trapezoidal integration to compute the area under the  
 566 curve, and are also reported in Table 4. The average fluxes over the entire eruptive period and the estimated total  
 567 emissions of SO<sub>2</sub>, HCl, HF, and CO<sub>2</sub> (Table 4) provide insight into the scale of the emissions of this eruption  
 568 with respect to other emission sources.

569 **Table 4: Estimate of total emissions during the eruption from gas to SO<sub>2</sub> ratios and SO<sub>2</sub> emission fluxes. The emission**  
 570 **fluxes estimates were performed using (1) an exponential fit for the SO<sub>2</sub> emission fluxes interpolation and (2) using**  
 571 **direct linear interpolation of daily SO<sub>2</sub> emission fluxes estimates (results between brackets). Total emissions to the**  
 572 **atmosphere are then derived combining the Monte Carlo and trapezoid integration methods.**

Species	Average specie to SO <sub>2</sub> mass ratios	Average volcanic emission fluxes (kg.s <sup>-1</sup> )	Total emissions (Mt)
SO <sub>2</sub>	1.0	300 ± 230	1.81 ± 0.18 (1.86 ± 0.09)
CO <sub>2</sub>	All studies: 12 ± 10 This study: 14 ± 9	2981 ± 1105	19.4 ± 1.8 (20.5 ± 1.9)
HCl	0.03 ± 0.02	7 ± 4	0.05 ± 0.01 (0.043 ± 0.003)
HF	0.0074 ± 0.0053	1.9 ± 1.3	0.013 ± 0.002 (0.013 ± 0.002)
CO	0.09 ± 0.05	23 ± 14	0.123 ± 0.005 (0.138 ± 0.009)

573 The total SO<sub>2</sub> emissions of 1.81 ± 0.18 Mt, derived from our exponentially decreasing fit, is similar to that  
 574 reported in Milford et al. (2023) using the daily SO<sub>2</sub> volcanic emissions derived from TROPOMI data (credit:  
 575 ESA, MOUNTS). These total SO<sub>2</sub> emissions are comparable to the emissions of the submarine 2011 Tagoro  
 576 eruption at El Hierro, that released between 1.8 and 2.9 Mt SO<sub>2</sub> into the ocean (estimated using the petrologic  
 577 method; see Longpré et al., 2017).

578 During the Tajogaite eruption, the highest  $\text{SO}_2$  emission fluxes occurred during the first ten days of the eruption  
579 (median of 37 kt/day during this period), and then had a lower median of about 20 kt/day. These  $\text{SO}_2$  emission  
580 rates are the same order of magnitude as the most recent basaltic eruptions such as for instance Piton de La  
581 Fournaise in 2020 (average: 0.9 kt/day; max: 25 kt/day, Hayer et al., 2023) in La Reunion island, Bárðarbunga  
582 in 2014-2015 (average of 50 kt/day over 6 months, Pfeffer et al., 2018) in Iceland, and lower than that found at  
583 Kilauea in 2018 (average of 200 kt/day; Kern et al., 2020), but the latter two exhibiting much higher eruptive  
584 TADR. For Tajogaite eruption, the high  $\text{SO}_2$  fluxes result from the high sulfur content of parental magma, as  
585 reflected by the average content of 3360 ppm in our MIs (Supplementary data), similar to the value of 3500 ppm  
586 reported in Burton et al. (2023) and Dayton et al. (2024).

587 For  $\text{CO}_2$ , we obtained a steady average emission flux of  $260 \pm 24$  kt/day, and total emissions of  $19 \pm 2$  Mt over  
588 the course of the eruption. This result aligns closely with the estimates of  $28 \pm 14$  Mt reported by Burton et al.  
589 (2023). These emissions represent 15% of global subaerial volcanic and tectonic annual emissions (Fischer and  
590 Aiuppa, 2020) or the equivalent of the annual  $\text{CO}_2$  budget of Ocean Island Basalt (OIB) volcanism, as estimated  
591 by LoForte et al. (2024). The high  $\text{CO}_2$  emissions with respect to the low extruded magma volume during  
592 Tajogaite eruption, compared to other effusive eruptions, are explained by the extraordinarily carbon-rich  
593 magma, as it is reflected in both fluid and melt inclusions (up to 2 wt%  $\text{CO}_2$  in MIs; Dayton et al., 2024). This is  
594 a characteristic of Macaronesian magmas and possibly of global OIB (Burton et al., 2023; LoForte et al., 2024;  
595 Van Gerve et al., 2024).

596 Daily total CO emissions emitted during the eruption, averaging 2 kt/day, were exceptionally high, with a  
597 cumulative total of  $0.12 \pm 0.01$  Mt. Only few volcanic CO emissions are reported in the literature, such as 0.15  
598 kt/day at Erebus volcano (Wardell et al., 2004), 0.007 kt/day at Oldoinyo Lengai (Oppenheimer et al., 2002),  
599 0.16 to 0.27 kt/day at Nyiragongo volcano (Sawyer et al., 2008a), 0.0007 kt/day at Erta Ale (Sawyer et al.,  
600 2008b) and are about one order of magnitude lower than our estimates during the Tajogaite eruption.

601 Finally, our estimated HCl and HF total emissions are about  $50 \pm 10$  kt and  $13 \pm 2$  kt, respectively, with an  
602 average of  $604 \pm 340$  t/day and  $173 \pm 86$  t/day. These emissions are in the same order of magnitude as that  
603 observed for other basaltic volcanoes, such as Etna (300-1300 t/day of HCl during the 2008-2009 eruption  
604 reported in Spina et al., 2023; 800 t/day of HCl and 200 t/day of HF in 1997 reported by Oppenheimer et al.,  
605 1998), Bárðarbunga volcano (500 t/day and 280 t/day for HCl and HF, respectively, reported in Galeczka et al.,  
606 2018). HCl and HF emissions from Tajogaite eruption are more than an order of magnitude higher than those  
607 observed at Kilauea volcano, which reported 12-22 t/day of HCl and 6-9 t/day of HF in 2008 and 2009 (Mather  
608 et al., 2012).

## 609 5. Discussion

### 610 5.1. Comparison of $\text{CO}_2$ , CO, HCl and HF to $\text{SO}_2$ ratios from different measurement 611 methods and sites

612 One of our key results is the remarkably strong consistency between the measured volcanic gas  
613 species-to- $\text{SO}_2$  ratios, whatever the measurement site, the technique and the instrument used (Fig. 3). The  
614 measurements conducted at the IZO station gave the excellent opportunity to assess the robustness of our  
615 estimated ratios, using both EM27/SUN and IFS-125HR instruments and their consistency with surface  
616 measurements. We found an excellent agreement between the HCl and HF total columns (with volcanic plume  
617 contribution) derived from the IFS-125HR and EM27/SUN products (see Appendix A for details).

618 We found a good comparability for the available  $\Delta\text{CO}_2/\text{SO}_2$  and  $\Delta\text{CO}/\text{SO}_2$  between surface and column  
619 measurements, reflecting an efficient vertical mixing. This also suggests that when the volcanic plume is  
620 detected by the surface measurements at the IZO station, the ground level concentrations are representative of  
621 the average volcanic plume composition. Since the IZO station is often located above the base height of the  
622 trade wind inversion (TWI) layer (Milford et al., 2023), volcanic plumes detected at IZO were typically  
623 transported rapidly through the low free troposphere. The progressive decrease in plume injection height  
624 throughout the eruption, combined with seasonal changes in the vertical stratification of the atmosphere (TWI  
625 height), resulted in sparse detections of the plume at the IZO station after mid-November 2021 (Milford et al.,  
626 2023). This led to a reduction of the coincident surface and total column observations.

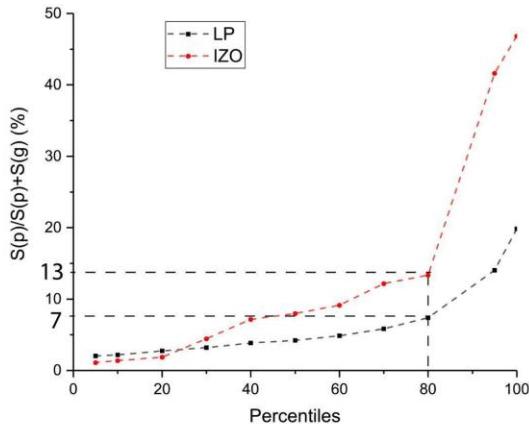
627 Moreover, the comparison of ratios at different distances from the eruptive vents (i) at IZO (140 km)  
 628 and (ii) near the active vent measured by OP-FTIR or MultiGAS (this work; Burton et al., 2023; Erickson et al.,  
 629 2024) allows qualitative assessment of the impact of in-plume reactions on our measurements. The ratios taken  
 630 from Burton et al. (2023) were derived from either in situ ground-based or drone-borne MultiGAS  
 631 measurements within the plume close to the volcanic vents, or, after 02/10/2021, from Open-Path FTIR  
 632 measurements pointing to the eruptive column and using the lava fountain as a source. Those reported by  
 633 Erickson et al. (2024) are limited to ground-based MultiGAS measurements. In any case, the gas measured by  
 634 these authors corresponds to the plume less than 1 km from the volcanic vents. Since  $\text{CO}_2$  is a non-reactive  
 635 species, a significant conversion of  $\text{SO}_2$  into sulfate aerosols ( $\text{H}_2\text{SO}_4$ ) during the transport between La Palma  
 636 and IZO should increase the  $\Delta\text{CO}_2/\text{SO}_2$  ratio. Hence, if significant conversion of  $\text{SO}_2$  to sulfates occurred during  
 637 the transport, the IZO ratios should be higher than those measured closer to the volcano. To examine this aspect,  
 638 we estimated the plume age for each recorded event using the Hysplit transport model, in both retro-trajectories  
 639 and forward simulation configuration mode. For meteorological data, we utilized 72-hour extended files  
 640 containing high-resolution meteorological information derived from the WRF-ARW model as input. This model  
 641 runs twice a day, using initial and boundary conditions from ECMWF's HRES-IFS data, with a resolution of  
 642  $0.09^\circ \times 0.09^\circ$  (for further details, refer to Appendix C). Table 5 shows the coinciding values of the  $\Delta\text{CO}_2/\text{SO}_2$   
 643 ratios measured at less than 1 km from the eruptive fissure (Burton et al., 2023) and at IZO (this work) and an  
 644 estimate of plume age for each event. Despite the limited number of coincident events at the two sites, no clear  
 645 dependence of this ratio on distance was observed for plumes with an age of 12 hours or less. Certain similarity  
 646 was found, at least until the beginning of November, even in cases of relatively old plumes (~12h), suggesting a  
 647 swift transport between La Palma and Tenerife islands and negligible in-plume reactions, at least  
 648 indistinguishable within the uncertainties of the ratios. In the troposphere, the  $\text{SO}_2$  to  $\text{SO}_4^{=}$  oxidation rates vary  
 649 significantly, from a few percent per hour by in-cloud droplet processes (driven by aqueous phase oxidation e.g.  
 650  $\text{H}_2\text{O}_2$ ) to a few percent per day (in dry air, driven by OH radicals) (Seinfeld and Pandis, 1998). Our results  
 651 suggest that this latter (slow dry oxidation) process may be the prevailing one during the transport in the dry  
 652 free-troposphere, from La Palma to IZO. This interpretation is supported by the sulfate aerosols measured in situ  
 653 in La Palma (Rodríguez et al., submitted) and at IZO, when the volcanic plume reaches the station, plotted in  
 654 Fig. 6. Figure 6A reports the statistical distribution of the ratio (in percent, %) of particulate sulfur (S(p), i.e.  
 655 sulfate  $\text{SO}_4^{=}$ ) over total sulfur (i.e. gas sulfur as sulfur dioxide (S(g)) plus S(p)) measured in the aerosols smaller  
 656 than 10 microns ( $\text{PM}_{10}$ ) at IZO and at La Palma during the eruption. Figure 6B shows the correlation plot of  
 657 S(g) as a function of S(g)+S(p). We observe a higher maximum conversion rate at IZO (45%) than in La Palma  
 658 (20%), as expected. However, 80% of the dataset (Fig. 6A and B) presents a conversion rate of their sulfur  
 659 content to  $\text{SO}_4$  below 15% and 7% at IZO and La Palma, respectively.

660 **Table 5: Comparison of the  $\Delta\text{CO}_2/\text{SO}_2$  ratio values at two different distances from the Tajogaite eruptive center and**  
 661 **estimate of plume age at IZO station. FTIR ratios are given between brackets to distinguish them from surface ratios.**

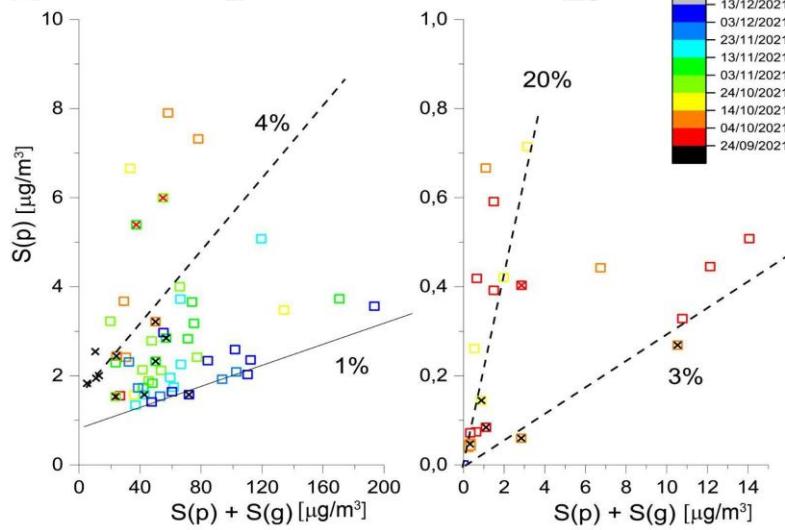
Date	Burton et al. (2023) Crater	IZO (140 km) Surface ratios (FTIR ratios)	Plume age at IZO (hour)
27/09/2021	6.8	$7.0 \pm 0.5$	~8h
07/10/2021 - 08/10/2021	13	$9.0 \pm 0.5$	~3h
13/10/2021	9;11	$12.0 \pm 0.3$	~12h
16/10/2021 - 17/10/2021	29	$10 \pm 0.5$ ( $13 \pm 1$ )	~12h
23/11/2021 - 24/11/2021	38.3	$18.3 \pm 0.7$	~12h

662

(A)



(B)



663

664 **Figure 6:** Statistical distribution of the  $S(p)$  over  $S(p)+S(g)$  ratio (in percentage, p and g refers to particle and gas  
665 respectively) measured at IZO (Tenerife) and at El Paso (La Palma) during the Tajogaite eruption. (A) shows the  
666 statistical distribution of the conversion rate estimated from the La Palma and IZO aerosols measurements.  
667 (B) reports  $S(p)$  as a function of  $S(p)+S(g)$  from the IZO and La Palma PM<sub>10</sub> analysis with the time as color scale. Crosses  
668 inside the square denote data points coinciding with FTIR measurements.

669 Furthermore, the time distribution of the  $S(p)/(S(p)+S(g))$  ratio (Fig. 6) suggests a higher conversion rate of SO<sub>2</sub>  
670 to sulfate during the first part of the eruption (until the beginning of November) compared to the second period.  
671 This trend appears closely tied to the volcanic plume's altitude relative to the Trade Wind Inversion (TWI), as  
672 described by Milford et al. (2023). During the first period of the eruption (until early November), plumes from  
673 explosive activity and fountaining vents often rose above the TWI, and surface measurements at La Palma likely  
674 captured older, dilute, more oxidized emissions from effusive vents trapped into the TWI. Conversely, from the  
675 beginning of November, the entire plume, comprising both explosive and effusive components, was more  
676 frequently trapped below the TWI, leading to the detection of younger, more concentrated, and less oxidized  
677 emissions at ground level. In any case, the plumes reaching IZO are most likely dominated by explosive  
678 emissions which, despite substantial transport times, exhibit oxidation rates below 15%. Such low conversion  
679 rates would not produce resolvable differences in our gas-to-SO<sub>2</sub> ratios. The last two events in Table 5 present  
680 some difference between both sites. On 16 October 2021, the FTIR and surface ratios at IZO are comparable,  
681 highlighting their robustness, however, they are a factor of 2-3 lower than those reported by Burton et al. (2023).  
682 We remark that for these days, the measurement target reported by these authors mention the base of the lava  
683 fountain instead of the spattering vents or passive degassing, as for the other three dates, implying different  
684 conditions and processes.

Finally, the  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios measured at FUE station (Fig. 4) and those recorded at IZO from surface measurements are on average higher and with higher variability than that recently reported in Asensio-Ramos et al. (2025) from open-path measurements (Fig. 4). This difference is likely due to the different measurement methods (solar absorption vs. open-path measurements using hot lava as source), implying different loci of measurements and gas contribution along their respective line of sight. Tajogaite volcano presented notable differences in eruptive behaviour between the different vents along the volcanic fissure, the higher elevated ones being more explosive than the lower ones. Recent studies suggest that eruptive dynamics may affect the abundance of redox-sensitive species (e.g.: Oppenheimer et al., 2018, Moussallam et al., 2019). Furthermore, we note that most of the Asensio-Ramos et al. (2025) measurement sites until the beginning of November (i.e: when our highest  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios were recorded) were located at the NNW from the eruptive fissure. With winds dominantly blowing towards the S and SW during this period, this configuration avoided a significant contribution of biomass and building burning plume to their measurements. It is not the case for the FUE measurements that were more likely to be affected by this contribution provoked by the advance of the lava flows. This hypothesis is also supported by the similarity of the  $\Delta\text{CO}/\Delta\text{CO}_2$  time series at FUE with the time series of the areas covered daily by the advancing lava flows (Appendix D), reflecting the extent of burnt vegetation. The typical values reported in the literature for the wildfires (Yokelson et al., 2007; Akagi et al., 2014; Vasileva et al., 2017; Álvarez et al., 2023) are generally higher than our values, by at least a factor of 5 likely explained by the different contributors of the measured plume, i.e. a mixing of volcanic plume and vegetation/infrastructures burning in the case of the 2021 La Palma eruption.

## 5.2 New insight into the eruption dynamics

The ratios and emission flux time series as well as total emission estimates presented here provide some information about the degassing processes during the Tajogaite eruption. Our time series of  $\text{SO}_2$  volcanic emission fluxes confirms the decreasing trend observed from the  $\text{SO}_2$  daily mass time series from Mounts (<http://www.mounts-project.com>, Valade et al., 2019) and reported in Milford et al. (2023). The concurrent decrease of  $\text{SO}_2$  emissions together with that of decreasing tephra accumulation rates and decreasing plume height was suggested to reflect the decrease of the pressure in the plumbing system (Milford et al., 2023). This was confirmed by the co-eruptive deflation trend observed and inverted by Charco et al. (2024), possibly related to the pressure drop due to drainage of the reservoir. The relatively good fit of the  $\text{SO}_2$  fluxes data obtained using an exponential function further supports this interpretation. We found a good correlation between the  $\text{SO}_2$  volcanic emission flux time series and the TADR (slope:  $14.1 \pm 1.2 \text{ kg of SO}_2 \text{ m}^{-3}$  of lava and  $R=0.94$ ). A similar correlation between  $\text{SO}_2$  emissions and effusive volumes has previously been observed during the 2021 Fagradalsfjall eruption (Pfeffer et al., 2024). The few outliers to this correlation (empty circles, Fig. E2) occurred during three distinct periods: (1) the initial days of the eruption, coinciding with the peak in  $\text{SO}_2$  emissions (2) just after the 27/09 eruptive pause, at the onset of sharp increase in effusion rates and (3) following the opening of the late November vents, north of the main vent alignment. These outliers correspond to abrupt changes in the output rate, likely associated with transient perturbations of the surface thermal structure-conditions known to affect the reliability of TADR estimations based on radiant density models (Coppola et al., 2016). Interestingly, applying the TADR values derived from the Pleiades-based volume estimates of Belart and Pinel (2022), which are averaged over 6-7 days, would bring at least three of these outliers back in line with the main trend. This suggests that apparent short-term imbalances between  $\text{SO}_2$  emissions and effusion rates may be rapidly compensated, resulting in a coherent degassing-effusion relationship over multi-day timescales. This is particularly evident at the beginning of the eruption, where the Belart and Pinel (2022) estimates yield significantly higher TADR values than those of Plank et al. (2023) (Fig. E2). Beyond these transient deviations, the correlation between  $\text{SO}_2$  flux and TADR remains remarkably consistent throughout the eruption, suggesting that the emitted  $\text{SO}_2$  predominantly reflects syn-eruptive magma degassing. This coherence, maintained over nearly three months of activity, indicates that the degassing regime remained stable once the eruption was fully underway. The early deviation from this trend, characterized by an apparent excess of  $\text{SO}_2$  emissions relative to effusion, may reflect the release of sulfur that had already exsolved in the shallow system prior to the eruption and its rapid release, followed, after the eruptive pause, by the evacuation

735 of the partly degassed magma. While this interpretation is consistent with the observed trends (Fig. E2), it  
736 remains tentative, given the absence of composition data for the earliest days of the eruption.

737 This correlation confirms that the emitted SO<sub>2</sub> only proceeds from the ascending magma. We observed a similar  
738 behavior for HCl, HF and CO emission fluxes, which contrasts with the almost constant CO<sub>2</sub> flux throughout the  
739 85 days of the eruption. This observation is fully consistent with the degassing model proposed by Burton et al.  
740 (2023), which suggests a decoupling between CO<sub>2</sub> and SO<sub>2</sub> degassing processes. According to this model, a  
741 CO<sub>2</sub>-rich volatile phase, already exsolved in the upper mantle reservoir, could account for a large fraction of the  
742 emitted CO<sub>2</sub> (up to ~80% according to Dayton et al., 2024), sustaining nearly constant CO<sub>2</sub> fluxes through the  
743 system. This difference is partially reflected in the time series of the ΔCO<sub>2</sub>/SO<sub>2</sub> ratio that steadily increases from  
744 the beginning of the eruption to the end of phase I, mimicking the trend of the VLP tremor amplitude. Such co-  
745 evolution abruptly ends at the beginning of November, from when the ratio becomes more variable. The CO<sub>2</sub>  
746 volcanic emission fluxes being constant within uncertainties during the whole eruption and the SO<sub>2</sub> volcanic  
747 emission fluxes being mainly controlled by the magma discharge rate, the steady increase of the C/S ratio during  
748 the first part of the eruption thus reflects the progressive decrease of the proportion of shallow (discharge)  
749 component relatively to the deep reservoir CO<sub>2</sub>-rich fluids. In the frame of overall lower SO<sub>2</sub> fluxes due to  
750 waning activity, the variability of the ratios of the phase II reflect the control of low SO<sub>2</sub> contents in the plume  
751 and short-term variability of the SO<sub>2</sub> emissions.

752 The early November transition between phase I and phase II follows the apparition of new vents at the end of  
753 October (Muñoz et al., 2022), interpreted as further propagation/opening of the underlying dike intrusion. This  
754 transition shortly anticipates an abrupt and enduring drop in tremor amplitude (both VLP and LP frequency  
755 bands; Bonadonna et al., 2022), geochemical changes (Ubide et al., 2023; Dayton et al., 2024) and hydrologic  
756 and hydrochemical changes in the aquifer. The latter comprises e.g. an influx of pure (most likely endogenous)  
757 CO<sub>2</sub> (Jimenez et al., 2024) that drastically increased the groundwater HCO<sub>3</sub><sup>-</sup> content at several sampling points  
758 from 27 October 2021 (Amonte et al., 2022; Garcia-Gil et al., 2023b) or the establishment of a direct  
759 relationship between the level in several groundwater wells and the tremor amplitude around 7 November 2021  
760 (Garcia-Gil et al., 2023a). VLP tremor amplitudes are especially sensitive to variations in magma ascent  
761 dynamics and conduit geometry (D'Auria et Martini, 2009; Bonadonna et al., 2022). Similar drops in VLP  
762 tremor amplitude were observed at other volcanoes, such as at Piton de la Fournaise (Duputel et al., 2023) where  
763 it was interpreted in terms of reduction of dyke dimension, heralding the end of the eruption. All these  
764 observations suggest that these events at the beginning of November constitute a turning point in the eruption  
765 implying significant structural changes in the plumbing system.

766 This turning point is particularly evident with the split described in the time series in the Sr isotopic  
767 compositions of the matrix, and interpreted as the consequence of a deep-origin melt injection replenishing the  
768 feeder system (Ubide et al., 2023). This interpretation further relies on this compositional change occurring in  
769 close time relationship with an increase in the magnitude of seismicity, VLP tremor amplitude and a short-term  
770 (5 days) rebound in the time series of daily SO<sub>2</sub> masses. We emphasize that the short-term increase in daily SO<sub>2</sub>  
771 masses observed between 28 October and 2 November 2021 should be interpreted with caution. First of all, at  
772 the depth of injection, SO<sub>2</sub> being mostly soluble in magma until a few hundred meters depth (Burton et al.,  
773 2023), any increase in SO<sub>2</sub> emissions would be due to an increase in lava discharge rate at the surface. Then, this  
774 apparent peak coincides with a period of low wind speeds and a reversal in wind direction at 700 hPa (ERA5  
775 data), which likely caused plume stagnation and gas accumulation. These meteorological conditions can lead to  
776 an overestimation of SO<sub>2</sub> masses derived from satellite data. Therefore, we do not interpret this increase as a  
777 definitive sign of enhanced volcanic degassing. The deep-origin melt injection at this period is further not  
778 supported by the absence of corresponding signals in the GPS baseline time series (Charco et al., 2024), TADR  
779 data (Plank et al., 2023), and our CO<sub>2</sub> fluxes and CO<sub>2</sub>/SO<sub>2</sub> ratios.

780 The observed multiparametric transition in the eruption dynamics at the beginning of November could be  
781 alternatively explained by a significant alteration of the magma pathway between the surface and the top of the  
782 magma chamber. With the waning of the eruption, the ascent rate decreased and the conduit became more  
783 unstable (Muñoz et al., 2022), with the opening of new vents from mid November (Gonzalez, 2022; Walter et  
784 al., 2023), resulting in interaction with the aquifer, changes in the tremor amplitude, mixing ratio and/or  
785 composition of endmembers and the return of radiogenic signatures.

786

### 5.3 Volatile mass balances and implications

787 Once released from the magma, volcanic gases suffer a number of processes such as oxidation,  
 788 scavenging and dissolution in aqueous fluids that can alter their original composition before their detection.  
 789 Integrating petrological constraints helps understanding volcanic degassing processes linking deep degassing to  
 790 atmospheric observations and refining our understanding of element cycling and the environmental impact of  
 791 volcanic plumes. We report here such an exercise estimating expected emissions estimated from petrological  
 792 data, and compare them with our estimates derived from atmospheric measurements.

793 **5.3.1 “Effective S degassing” and SO<sub>2</sub> mass balance**

794 Combining our SO<sub>2</sub> volcanic emission fluxes and new petrological data, complementing literature,  
 795 allow us to estimate a S degassing balance for the Tajogaite eruption. We used a similar Monte Carlo approach  
 796 as proposed in Dayton et al. (2024), but refining the degassing balance as follows. We use an erupted lava  
 797 volume of  $(177 \pm 5.8) \times 10^6 \text{ Mm}^3$  from Civico et al. (2022), a distal tephra volume of  $(22.8 \pm 1.8) \times 10^6 \text{ Mm}^3$  from  
 798 Bonadonna et al. (2023) and a cone volume of  $(36.5 \pm 0.3) \times 10^6 \text{ Mm}^3$  from Civico et al. (2022). The total  
 799 erupted mass is obtained applying a similar approach to Dayton et al. (2024), using densities of  $2403 \pm 170$   
 800  $\text{kg} \cdot \text{m}^{-3}$  for lava flows, based on an average percentage of vesicles for the erupted lava,  $1800 \text{ kg} \cdot \text{m}^{-3}$  for the cone  
 801 and  $1200 \pm 120 \text{ kg} \cdot \text{m}^{-3}$  for the tephra blanket (Bonadonna et al., 2022), resulting in a total erupted mass of  $5.2 \times 10^8$  tons. S degassing from the magma is usually estimated from petrological data (difference between MI and  
 802 matrix glass S contents), as in the mass balance of Burton et al. (2023) and Dayton et al. (2024) for the Tajogaite  
 803 eruption. The observed correlation between the TADR and our SO<sub>2</sub> volcanic emission fluxes allows us to  
 804 directly relate the degassed volume and the emitted S mass, with  $14.1 \pm 1.2 \text{ kg SO}_2$  emitted per “thermal” cubic  
 805 meter of lava (lava volumes estimated using the radiant flux). We corrected this thermal volume for the tephra  
 806 volume (blanket and cone) representing  $\sim 33\%$  of the total emitted volume, because this does not participate  
 807 significantly in the radiant flux. This resulted in  $9.4 \pm 0.8 \text{ kg degassed SO}_2$  per cubic meter of emitted lava,  
 808 which converts into  $2611 \pm 285 \text{ ppm effective S degassing}$ , considering above density and a correction of the  
 809 crystal mass fraction (25% following Dayton et al., 2024). This value is very similar to that obtained by Dayton  
 810 et al. (2024) using the difference between the S content of inclusions ( $3062 \pm 500 \text{ ppm}$ ) and matrix glasses ( $345 \pm 53 \text{ ppm}$ ). Note that the matrix S contents we present (average 534 ppm; N=52;  $\sigma=130 \text{ ppm}$ ; Supplementary  
 811 Table S1) are consistent with previously published datasets for the eruption (average of 403 ppm; N=438;  $\sigma=10 \text{ ppm}$ ;  
 812 Burton et al., 2023; Longpré et al., 2025). These data are nevertheless substantially higher than the value  
 813 reported by Dayton et al. (2024). Using these values in the MonteCarlo degassing simulation of Dayton et al.  
 814 (2024), the full degassing of  $0.25 \text{ km}^3$  of magma would produce emissions of  $1.93 \pm 0.21 \text{ Mt SO}_2$ . This is  
 815 compatible with the TROPOMI-derived total SO<sub>2</sub> emissions ( $1.81 \pm 0.18 \text{ Mt}$ ).

816 A possibly unaccounted repository for initial S in the degassing balance could be the rare sulfide droplets,  
 817 previously described to be present in the eruptive products matrix (Fig. B1; Day et al., 2022; Pankhurst et al.,  
 818 2022) but also, more recently in clinopyroxene (CPx) cores and in magnetites (Andujar et al., 2025). These  
 819 droplets separated from the silicate melt upon reaching the sulfide saturation during a pre-eruptive  
 820 crystallization episode (Day et al., 2022), as confirmed by our own saturation calculations using the O’Neil  
 821 (2021) SCSS model (see Appendix B2). Importantly for the sulfur budget, although part of the primitive magma  
 822 S content, as recorded in MI, the sulfur they contain is not included in matrix glass analyses (since it is  
 823 physically segregated) and is not released as gas during eruption. The sulfide abundance could range between  
 824 0.03 vol.% (QEMSCAN quantification in Pankhurst et al., 2022) and 0.066 vol.% (0.001 mass fraction in the  
 825 crystallizing assemblage in the models of Day et al., 2022). Assuming a density of  $4500 \text{ kg} \cdot \text{m}^{-3}$  (Saumur et al.,  
 826 2015) and an average sulfur content of  $\sim 35\%$  in the analyzed sulfides (Fig. B1), this range of abundance would  
 827 represent a potential sulfide cargo in the erupted lava until day 20 (Day et al., 2022) of  $\sim 30$  to  $60 \text{ kt}$  of non-  
 828 degassed sulfur (equivalent to  $\sim 60$  to  $120 \text{ kt}$  of SO<sub>2</sub>). Accounting for this contribution would further improve the  
 829 agreement between the petrologic budget ( $1.81$ - $1.87 \text{ Mt}$  of SO<sub>2</sub>) and satellite-based estimates ( $1.81 \pm 0.18 \text{ Mt}$  of  
 830 SO<sub>2</sub>).

831 Surprisingly, applying the same approach for the first week of the eruption (LU1 in Bonadonna et al.,  
 832 2023) encompassing the TROPOMI-derived SO<sub>2</sub> emission peak, we observe a mismatch of a factor of 3  
 833 between the expected SO<sub>2</sub> degassing and that measured by TROPOMI. This arises from the very low thermal

836 lava volume ( $4.3 \text{ Mm}^3$  estimated using the radiant flux, corrected with the factor of 2 proposed by Plank et al.,  
837 which can be due to the transient time required for the surface thermal structure to become steady  
838 (Coppola et al., 2016). Alternatively, using the cumulative volume of  $43.0 \pm 6.1 \text{ Mm}^3$  on the 26-09-2021  
839 reported by Belard and Pinel (2022) and derived from multiple Pléiades stereoscopic surveys during the first  
840 period of the eruption) and assuming a volume of  $15 \pm 0.12 \text{ Mm}^3$  for the edifice (Romero et al., 2022), we found  
841 cumulated  $\text{SO}_2$  emissions of about  $580 \pm 66 \text{ kt}$ , which is closer to the TROPOMI-derived estimates for this  
842 period (about 560 kt).

### 843        5.3.2 $\text{CO}_2$ mass balance and estimation of the reservoir volume

844        Applying the same MonteCarlo approach used for sulfur and assuming full  $\text{CO}_2$  degassing, we estimate  
845 that  $\sim 4.4 \pm 0.8 \text{ Mt}$  of  $\text{CO}_2$  would have been released from the erupted material alone. This is consistent with the  
846 estimate of  $5.4 \pm 1.0 \text{ Mt}$  by Dayton et al. (2024). However, plume measurements indicate significantly higher  
847 total  $\text{CO}_2$  emissions during the eruption, amounting to  $19.4 \pm 1.8 \text{ Mt}$ . This discrepancy, combined with the near-  
848 constant fluxes throughout the eruption, supports the presence of a  $\text{CO}_2$ -rich fluid phase in the reservoir  
849 (Hansteen et al., 1998; Burton et al., 2023) coexisting with a  $\text{CO}_2$ -saturated melt, capable of contributing an  
850 additional 15 Mt of  $\text{CO}_2$ . Based on FI densities reported by Dayton et al. (2023), we estimate that this additional  
851 15 Mt of  $\text{CO}_2$  corresponds to a fluid volume of  $\sim 25-17 \text{ Mm}^3$  at the pressure of the shallow (deflating) reservoir  
852 pressure and at that of the deeper reservoir, respectively.

853        The  $\sim 1\%$  pressure drop relative to the pressure at the beginning of the eruption observed by Charco et  
854 al. (2024) provides an opportunity to derive a first-order constraint on the volume of the deflating reservoir.  
855 Assuming this pressure loss is attributed to a volume change due to magma extraction, we can estimate the total  
856 volume of “hydraulically” connected magma/mush feeding the eruption.

857        To estimate the total volume (magma+fluid) extracted from the reservoir, 1) we corrected the eruptive products  
858 volume for vesicularity (Dense Rock Equivalent or DRE volume, taking as a reference a melt density of  $\sim 2700$   
859  $\text{kg.m}^{-3}$ ; see previous section and Dayton et al., 2024), 2) we added the volume of the magma-filled dykes and sill  
860 network (as described by De Luca et al., 2022) and 3) finally, we corrected for the effect of magma  
861 compressibility. According to Rivalta and Segall (2008), the volume ratios (intrusion/associated reservoir  
862 deflation) necessary to estimate magma compressibility range between 1.2 and 7.7. For the Tajogaite eruption,  
863 the most likely value is  $\sim 5$  (reservoir from 10 to 15 km deep, saturation depth  $> 25 \text{ km}$ ; see Fig. 3 in Rivalta and  
864 Segall, 2008). Using such values for correcting our magma volume and adding our extracted (additional) fluid  
865 volume estimate allows estimating a total volume (magma + fluids) extracted from the reservoir of  $\sim 60 \text{ Mm}^3$   
866 (from 45 to  $200 \text{ Mm}^3$  for the full range of volume ratios). Considering the extraction of this volume produced  
867 the pressure drop in the deflating reservoir, we roughly estimate the volume of magma/mush to equate, at least  
868 to  $6 \text{ km}^3$  ( $4-20 \text{ km}^3$  range). This estimate provides a first-order volume of magma/mush that could have been  
869 “hydraulically” connected to the surface during the eruption. It includes at least the shallow reservoir, but may  
870 also encompass deeper zones of the plumbing system if they were effectively connected during the eruptive  
871 episode.

### 872        5.3.3 Halogens mass balance

873        Fluorine and chlorine generally have high solubility in magmas and only begin to exsolve at shallow  
874 depths, close to the fragmentation level (e.g.: Aiuppa, 2009). This is likely the case for the 2021 La Palma  
875 eruption, where rapid magma ascent (Romero et al., 2022; Boneschi et al., 2024) limited halogen degassing due  
876 to kinetic constraints. As a result, the melt retained most of its original halogen content, and the difference  
877 between melt inclusions and matrix glass Cl and F contents is hardly resolvable from analytical uncertainty  
878 (Dayton et al., 2024). We thus assessed the consistency of our fluxes using another approach, estimating the  
879 expected Cl and F degassed amounts from the total observed emissions.

880        The adsorption of halogen-derived salts onto ash surfaces is likely to be a non-negligible sink for  
881 hydrogen halides of the volcanic plume (Bagnato et al., 2013) and should be considered in our balance. We thus  
882 propose a rough estimate of the scavenged halogen mass using the median (and standard error) content of Cl  
883 (335  $\pm$  34 ppm) and F (422  $\pm$  49 ppm) from a compilation (N=57) of published lixiviation experiments

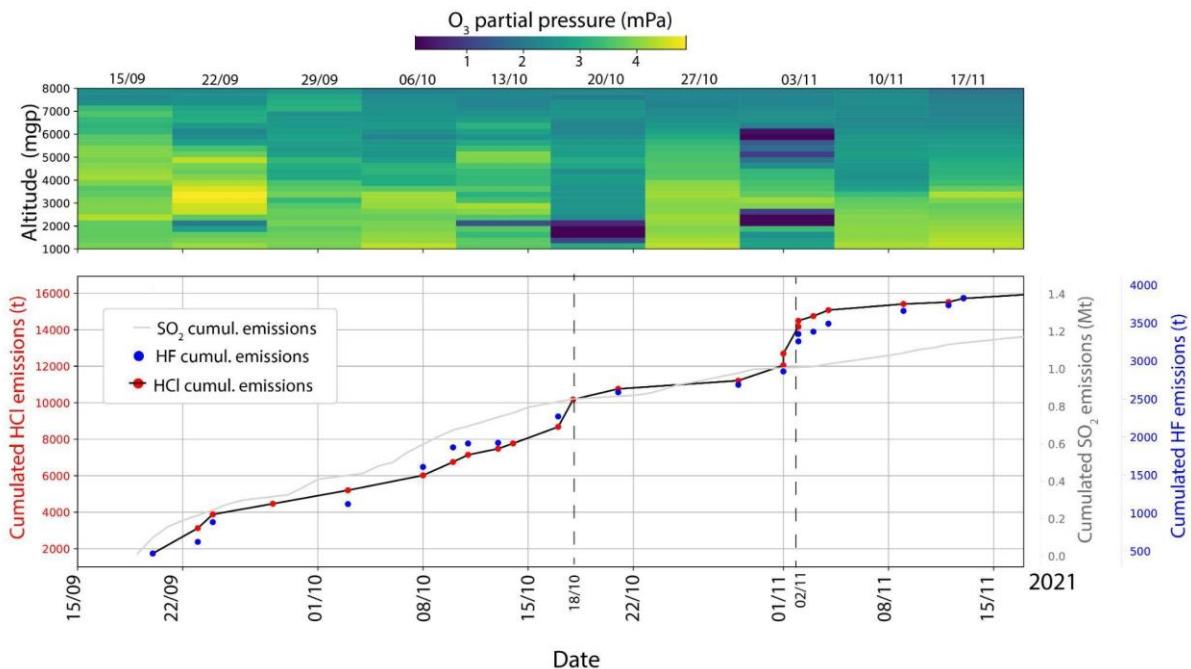
884 (Ruggieri et al. 2023; Sanchez-España et al., 2023; Rodriguez et al., 2025, submitted) and the mass of tephra  
885 emitted throughout the eruption (Bonadonna et al., 2022), including the cone (Civico et al., 2022). We obtain  
886 estimates of  $31 \pm 8$  kt HCl and  $39 \pm 11$  kt HF possibly scavenged from the plume, that we need to sum to our  
887 measured HCl and HF budgets ( $49 \pm 12$  kt and  $13 \pm 2$  kt of HCl and HF, respectively), giving surface emissions  
888 of  $80 \pm 15$  and  $52 \pm 11$  kt for HCl and HF, respectively.

889 Using the average Cl and F contents in MIs of Dayton et al. (2024), these emissions can be explained  
890 with Cl and F losses of  $\sim 195$  and  $130$  ppm from the melt, respectively. This is  $35\%$  and  $9\%$  of the initial melt  
891 content in Cl and F, respectively. This Cl difference should be resolvable analytically, but the F difference is  
892 indeed within the analytical uncertainty of electron microprobe for volcanic glasses (Rose-Koga et al., 2021)  
893 and at the limit of that for the Secondary Ion Mass Spectrometry analyses of Dayton et al. (2024). We propose a  
894 complementary estimation of the Cl loss from the melt using petrological data of the MI and matrix glasses of  
895 the eruption (Burton et al., 2023; Dayton et al., 2024; Longpré et al., 2025). The determination of the amount of  
896 Cl degassing from the melt is indeed obscured by the magma evolution in the plumbing system, as shown by the  
897 bivariate diagram between  $K_2O$  and the Cl contents (Fig. B3), where the matrix glass Cl contents are  
898 consistently higher than that of MI, impeding simple quantifications by difference as for S balance. In this  
899 diagram, MIs define a trend (Pearson's  $R=0.943$ ) that can be used to estimate the average Cl amount degassed  
900 from magma. We find an error-weighted mean Cl content difference between the simulated undegassed magma  
901 compositions and the matrix glasses of  $189 \pm 10$  ppm (95%;  $N=633$ ;  $\sigma=135$ ), within uncertainties of our  
902 degassing balance approach. The total HCl emissions that would arise from such degassing from the volume of  
903 eruptive products is  $77 \pm 7$  kt, indistinguishable from our HCl balance of  $80 \pm 15$  kt within uncertainties. This  
904 approach is not possible for F due to significant variability in MI F contents.

#### 905 **5.4 Potential atmospheric implications of Tajogaite eruption emissions**

906 Volcanic emissions of greenhouse gases and reactive species represent critical inputs for climate  
907 models, as they contribute to baseline radiative forcing, perturb the oxidative capacity of both the troposphere  
908 and stratosphere, influence aerosol–cloud microphysical interactions, and play a significant role in the  
909 geochemical cycling of key elements such as sulfur, carbon, and halogens between the Earth's surface and  
910 atmosphere (Von Glasow et al., 2009 and references therein). Accurate quantification of these natural fluxes is  
911 essential for distinguishing anthropogenic signals from background variability in atmospheric composition. The  
912 Tajogaite eruption provides a striking example of how a single volcanic event can temporarily dominate  
913 regional atmospheric budgets. Its  $SO_2$  emissions were approximately 15 times greater than Spain's total  
914 anthropogenic  $SO_2$  emissions for the year 2021 (123 kt; MITECO, 2023), and even exceeded the total EU  
915 anthropogenic  $SO_2$  emissions for that year (1.4 Tg; EEA, 2023). Assuming a conservative 20% conversion rate  
916 of S to sulfate aerosols (see Section 5.1 and Fig. 6), the eruption is estimated to have produced approximately  
917 0.5 Mt of sulfate particles. However, since the plume remained below 8 km altitude, well within the troposphere,  
918 these aerosols were likely short-lived and regionally confined, with limited potential to affect atmospheric  
919 radiation budgets, and a negligible climatic forcing from aerosol loading. In terms of carbon emissions, the  $CO_2$   
920 released by the eruption amounted to approximately 10% of Spain's anthropogenic  $CO_2$  emissions for 2021  
921 (<https://www.miteco.gob.es/es/calidad-y-evaluacion-ambiental/temas/sistema-espanol-de-inventario-sei-informe-interactivo-inventario-nacional-emisiones-atmosfera.html>). Emissions of CO were also substantial,  
923 corresponding to about 7% of the 2021 national anthropogenic CO inventory (1.64 Mt; MITECO, 2023).  
924 Halogen emissions were particularly notable. The total atmospheric HCl output was around ten times higher  
925 than the annual UK emissions since 2017 (UK National Atmospheric Inventory) and represented roughly 20%  
926 of total European anthropogenic HCl emissions in 2014 (220 kt; Zhang et al., 2022), which are primarily  
927 associated with the energy sector (38%) and open waste burning (23%). Similarly, the eruption's HF  
928 atmospheric emissions exceeded UK national totals for the same period by an order of magnitude. In contrast to  
929 the purely atmospheric pathway, a significant fraction of the halogens was likely scavenged from the plume by  
930 ash particles. This process, which accounts for an estimated  $31 \pm 8$  kt of HCl and  $39 \pm 11$  kt of HF, provides a  
931 distinct mechanism for their re-entry into the geosphere through ash deposition. Subsequently, these ash-bound  
932 halogens are remobilized by initial rainfall events (Medina et al., 2025), where they can enter and be transported  
933 through natural elemental cycles of Cl and F in soil, aquifer, and marine environments.

934 Volcanic emissions of chlorine are known to significantly influence tropospheric ozone ( $O_3$ ), as these  
 935 halogens participate in catalytic cycles that destroy  $O_3$ , particularly in the presence of sunlight and moisture  
 936 (Gerlach, 2004). Studying these emissions allows assessing the chemical forcing of volcanoes on the  
 937 troposphere, test atmospheric chemistry models using real events, and improve our understanding of the climatic  
 938 and chemical role of volcanic eruptions, even moderate ones like Tajogaite. In this study, we looked for signs of  
 939 such an impact in the local  $O_3$  total column from FTIR spectroscopy but found no clear evidence. However, Fig.  
 940 7 displays the time series of  $O_3$  partial pressure up to 8000 m, retrieved from electrochemical concentration cell  
 941 (ECC) ozonesonde measurements conducted by AEMET (García et al., 2021) from Puerto de la Cruz, Tenerife.  
 942 These are shown together with cumulative  $SO_2$ , HF, and HCl emissions up to 18 November 2021, corresponding  
 943 to the period with the most continuous and densely sampled flux measurements. A noticeable coincidence was  
 944 observed between the two sharp increases in the cumulative HCl (and HF) emissions, occurring on the  
 945 18/10/2021 and 02/11/2021 and local ozone depletion (with  $O_3$  values near zero) at plume altitudes. This  
 946 coincidence is not observed for low HCl/ $SO_2$  ratios.



947  
 948 **Figure 7: Relationship between the partial pressure of  $O_3$  at the volcanic plume altitude (from AEMET radiosonde**  
 949 **data) and the volcanic gas fluxes.  $SO_2$ , HCl, and HF emissions are shown as cumulative curves to highlight key**  
 950 **temporal variations. The  $O_3$  partial pressure is derived from radiosonde measurements conducted at the AEMET**  
 951 **Tenerife station.**

952 Although geometric constraints and weather conditions affected the continuity of our cumulative flux estimates,  
 953 this preliminary observation suggests that halogen-induced  $O_3$  loss may occur locally, at least where the plume  
 954 was present, even if only transiently. The observed  $O_3$  loss appears to be short-lived, with concentrations  
 955 recovering shortly after, arguing against a persistent or widespread effect. To better assess the intensity and  
 956 duration of this impact, more continuous time series and refined flux retrievals are required. Nonetheless, this  
 957 initial evidence from the Tajogaite eruption provides a valuable basis for future investigations.

## 958 6. Summary and Conclusion

959 In this study, we explored the variability of the chemical composition of the Tajogaite eruption  
 960 volcanic gas plume by combining ground-based FTIR and UV direct-sun measurements with surface gas  
 961 observations at two sites: Fuencaliente, on La Palma, and the Izaña high-altitude Atmospheric Observatory, a  
 962 reference station for atmospheric studies located in Tenerife. New retrieval methods are presented to derive the  
 963 HF and HCl volcanic contribution in the total columns obtained from the solar FTIR spectra for both low  
 964 (EM27/SUN) and high (IFS-125HR) spectral resolution measurements performed up to 140 km from the  
 965 eruptive fissure. The good agreement between the different products (total columns and ratios) obtained from

966 the different instruments (FTIR, DOAS and surface measurements) demonstrates the robustness of our results,  
967 even at such distant and low-concentration locations as the 140 km-far IZO Observatory. Our compositional  
968 ratios measured during the eruption are also consistent with the limited data reported in the literature (Asensio-  
969 Ramos et al., 2025; Erickson et al., 2024; Burton et al., 2023), including for previous basaltic eruptions in the  
970 world (e.g. Aiuppa et al., 2009). We derived SO<sub>2</sub> volcanic emission fluxes from the TROPOMI data and  
971 assessed the long-term variability of the emission fluxes of the other volcanic species, based on our  
972 compositional data. We found total emissions of CO<sub>2</sub>, SO<sub>2</sub>, HCl, HF and CO of 19.4±1.8, 1.8±0.2, 0.05±0.01,  
973 0.013±0.002 and 0.123±0.005 Mt, respectively. These emissions were found to be non-negligible in the annual  
974 Spanish national and European inventory balance compared to anthropogenic emissions. Furthermore, while the  
975 SO<sub>2</sub> and halogen halides emission fluxes decreased throughout the eruption along with the lava emission fluxes,  
976 the CO<sub>2</sub> emission fluxes were found to be almost constant, implying a comparatively increasing discharge with  
977 respect to the daily emitted lava volumes. This is consistent with a significant amount of CO<sub>2</sub> being already  
978 exsolved in the reservoir, as previously observed by Burton et al. (2023), Dayton et al. (2023) and Dayton et al.  
979 (2024). Global degassing balances were performed for C, S, Cl and F, showing a good consistency between the  
980 plume measurements and the petrological data. This study highlights the potential of employing existing global  
981 atmospheric FTIR, DOAS and surface measurement networks to explore remotely (>100 km) the variability of  
982 volcanic plumes chemical composition and its implications at different timescales. By demonstrating their  
983 effectiveness in tracking volcanic emissions in real time, our findings underscore the value of these networks for  
984 both operational volcano monitoring and scientific investigations during and after eruptive crises. Such  
985 measurements are crucial for assessing the role of volcanic emissions as natural sources in the global cycling of  
986 carbon, sulfur, and halogens. This study emphasises the value of solar absorption measurements for  
987 volcanology, atmospheric research, and air-quality monitoring during eruptions, and suggests their potential  
988 application during major eruptions even when access is more restricted.

## 989 7. Appendices

### 990 Appendix A

#### 991 Comparison between the new HF and HCl products derived from the IFS-125HR and EM27/SUN 992 measurements

993 The appendix A provides a summary of the comparison between the standard NDACC FTIR HCl and HF  
994 products and those developed in this study (Fig. A1), the new IFS-125HR SO<sub>2</sub> retrievals (Fig. A1c), as well as  
995 the comparison between all the IFS-125HR and EM27/SUN products (Fig. A2). As illustrated by the  
996 comparison (Table A1), the standard and optimised approaches show an excellent agreement under background  
997 conditions with a mean bias of approximately 3% and 15% for HCl and HF, respectively, while the scatter is  
998 limited to 4% for both trace gases. These values fall within the expected uncertainty estimations of the IFS-  
999 125HR products (García et al., 2021). However, for volcanic emissions, the NDACC approaches, in contrast to  
1000 our optimized approach, are not able to capture the volcanic HCl and HF contributions in the lower/middle  
1001 troposphere, resulting in a mean difference of 88% and 100% for HCl and HF, respectively. Column  
1002 enhancements as large as 6.00 x 10<sup>20</sup> and 2.05 x 10<sup>20</sup> molec/m<sup>2</sup> for HCl and HF, respectively, were reported  
1003 during the volcanic process, which accounts for the high variability observed between mean, median and scatter  
1004 values under volcanic emissions.

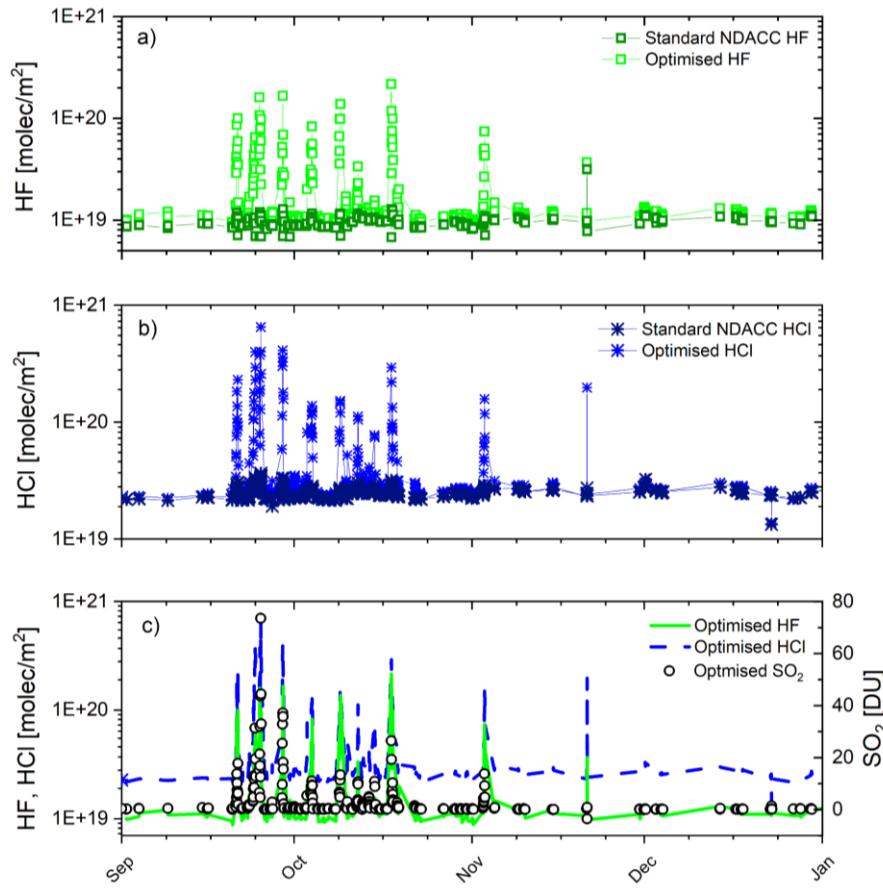


Figure A1: Time series of the standard NDACC and optimised HF (a) and HCl (b) total column amounts measured at IZO from the IFS-125HR instrument between 1 September and 31 December 2021. (c) Time series of the optimised HF, HCl and SO<sub>2</sub> IFS-125HR products at IZO for the same period.

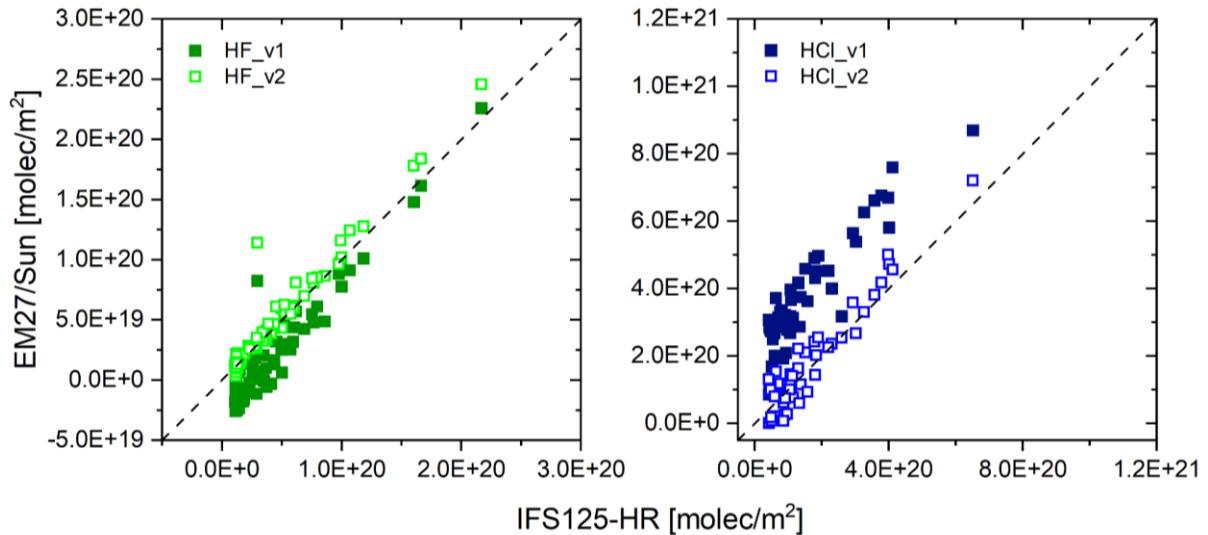
Figure A1 also presents the SO<sub>2</sub> total column amounts retrieved from the IFS-125HR measurements at IZO. The excellent agreement found between the SO<sub>2</sub>, HCl and HF retrievals consistently capturing volcanic plumes probes the reliability and quality of the optimised IFS-125HR products, which has been also documented by side-by-side Pandora and FTIR SO<sub>2</sub> observations (Taquet et al. 2023). As found for the NDACC FTIR sites of IZO and Altzomoni (García et al., 2022), the Pandora and FTIR comparison shows an excellent correlation for the whole SO<sub>2</sub> range observed (Pearson correlation coefficients larger than 0.99) and the scatter between techniques is comparable to background signal (less than 0.7 and 2.0 DU for IZO and Altzomoni, respectively). For further details about SO<sub>2</sub> IFS-125HR retrieval refer to García et al. (2022).

**Table A1: Summary of the comparison between the standard NDACC and optimised HF and HCl total column amounts measured at IZO from the IFS-125HR instrument for the period (1) 1-15 September and 1-31 December 2021 under background conditions; and (2) between 19 September and 31 November affected by volcanic emissions. N stands for the number of measurements and STD corresponds to the standard deviation of the data distribution.**

IFS-125HR Products	Background conditions (1-15 September & 1-31 December, N=68 for HCl and N=67 for HF)			Volcanic emissions (19 September - 31 November, N=414 for HCl and N=405 for HF)		
	Mean	Median	STD	Mean	Median	STD
NDACC HCl [molec.m <sup>-2</sup> ]	2.47E19	2.50E19	1.46E18	2.47E19	2.40E19	2.69E18
Optimised HCl	2.55E19	2.57E19	2.02E18	5.07E19	2.7E19	6.86E19

[molec.m <sup>-2</sup> ]						
NDACC - Optimised HCl [molec.m <sup>-2</sup> ]	8.11E17	7.49E17	9.79E17	2.60E19	2.88E18	6.66E19
NDACC - Optimised HCl [%]	3.2	3.0	3.9	88	12	204
NDACC HF [molec.m <sup>-2</sup> ]	9.92E18	9.77E18	6.85E17	9.51E18	9.19E18	1.46E18
Optimised HF [molec.m <sup>-2</sup> ]	1.14E19	1.14E19	6.95E17	1.95E19	1.12E19	2.34E19
NDACC - Optimised HF [molec.m <sup>-2</sup> ]	1.50E18	1.45E18	3.64E17	1.00E19	1.68E18	2.32E19
NDACC - Optimised HF [%]	15.2	14.9	4.1	100	18	288

1021 Figure A2 shows the comparison between the new HCl and HF products (HCl\_v2 and HF\_v2) derived from the  
 1022 EM27/SUN measurements, as well as those (HCl\_v1 and HF\_v1) estimated using the same retrieval as Butz et  
 1023 al. (2017) and the new products derived from the IFS-125HR measurements at IZO. Fit parameters are  
 1024 presented in Table A2. An excellent correlation was found for the newly developed EM27/SUN products  
 1025 (HCl\_v2 and HF\_v2), highlighting the improvements of the retrieval methods especially in case of far  
 1026 measurement sites.



1027  
 1028 **Figure A2: Intercomparison between the IFS-125HR and EM27/SUN HF and HCl total columns obtained from side-  
 1029 by-side measurements during the Tajogaite eruption. The diagonal (y=x) is plotted as a dashed line. The fit  
 1030 parameters from linear regression are given in Table A2.**

1031 **Table A2: Fit parameters obtained from the linear regression between the EM27/SUN HF and HCl products and  
 1032 those from the IFS-125HR.**

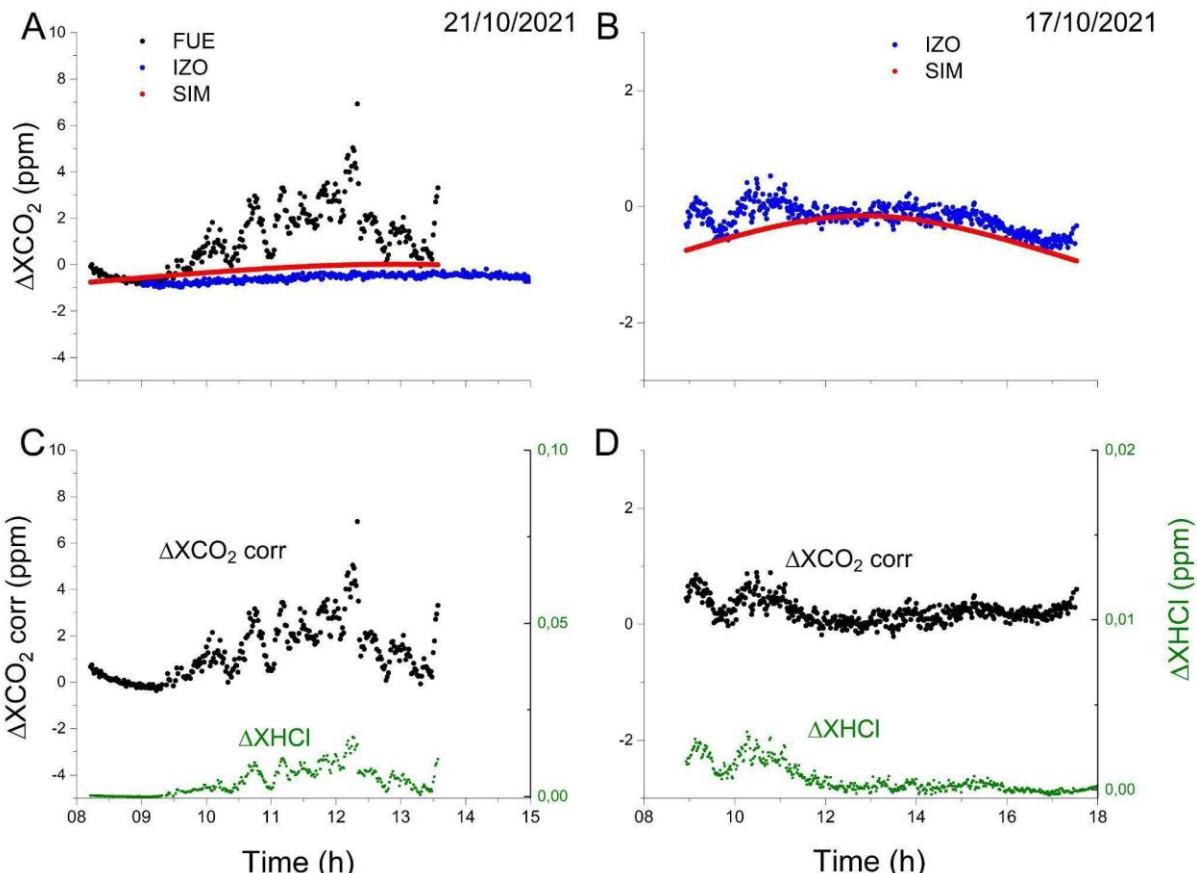
EM27/Sun (10 min average centered in the IFS125-HR measurements) vs. IFS-125HR	Linear regression using least squares fitting method
HF_v1	Slope=1.11±0.03

	Offset=(-2.7±0.2)×10 <sup>19</sup> molec/m <sup>2</sup> R=0.91
HF_v2	Slope=1.11±0.025 Offset=(-2.0±11.8)×10 <sup>17</sup> molec/m <sup>2</sup> R=0.98
HCl_v1	Slope=1.14±0.07 Offset=(20.4±1.5)×10 <sup>19</sup> molec/m <sup>2</sup> R=0.92
HCl_v2	Slope=1.11±0.05 Offset=(-0.43±1.07)×10 <sup>19</sup> molec/m <sup>2</sup> R=0.95

1033

1034 **Procedure for removing CO and CO<sub>2</sub> background to the total column estimates and extracting volcanic**  
1035 **contribution**

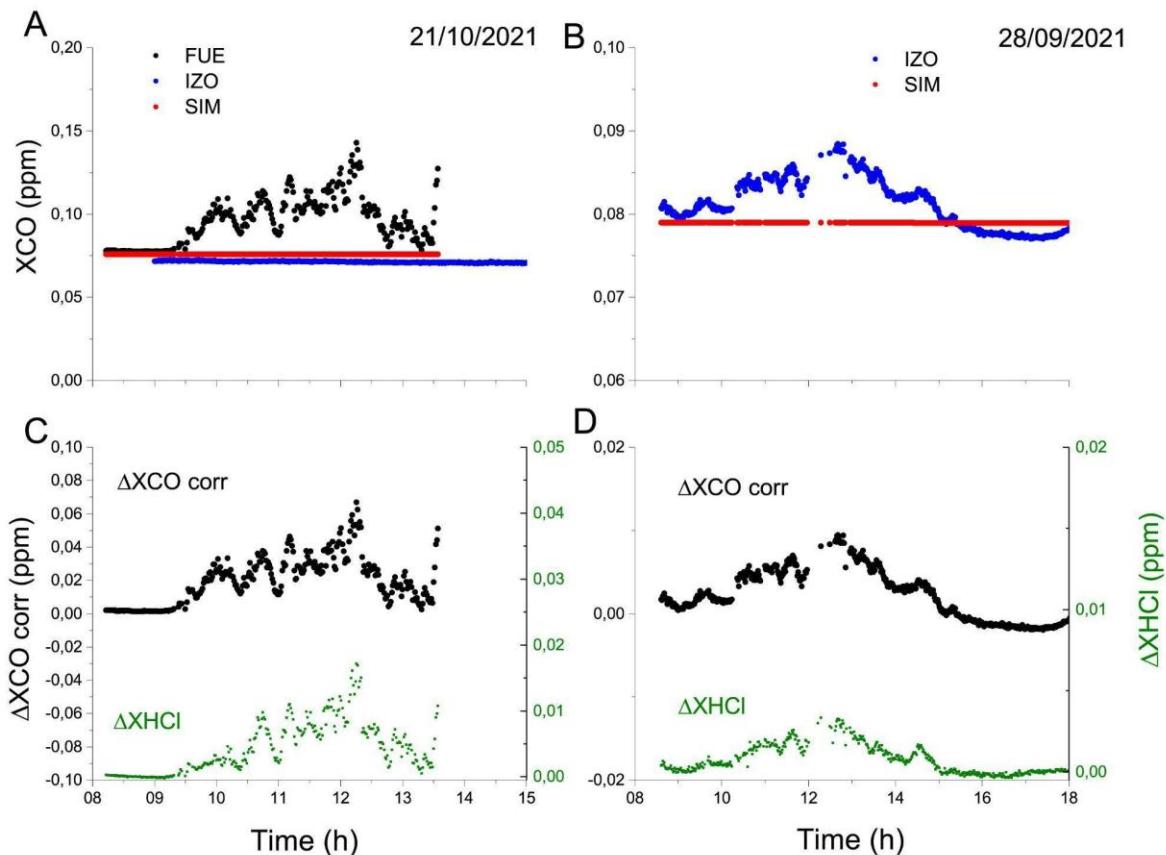
1036 CO and CO<sub>2</sub> analysis from the total column measurements require the simulation and removal of the  
1037 background concentration. Figures A3 and A4 show the procedure we employed for this study, through two  
1038 examples from the FUE and IZO dataset. The time series is first detrended from the annual cycle, using IZO  
1039 long-term time series and a third degree polynomial. The intraday variability of CO<sub>2</sub> background is then  
1040 simulated using the average of the XCO<sub>2</sub> intraday time series from spectra without volcanic plume contribution.  
1041 The background contribution is simulated for each day, fitting an offset. An example of the total procedure is  
1042 illustrated in Fig. A3 and A4 for CO<sub>2</sub> and CO, respectively. The simulated background at FUE (in red, A) was  
1043 compared with IZO measurements (in blue, A) when it was not affected by the volcanic plume. The resulting  
1044 intraday time series of  $\Delta XCO_2$  (Fig A3C and A3D) and  $\Delta XCO$  (Fig A4C and A4D) in presence of volcanic  
1045 plume are well correlated with the  $\Delta HCl$ , which can be considered as a tracer of the volcanic plume.



1046

1047 **Figure A3: Procedure for removing the atmospheric background contribution to estimate the CO<sub>2</sub> and CO**  
1048 **abundance in the volcanic plume. A and B show a typical example of uncorrected  $\Delta XCO_2$  at FUE (in black)**  
1049 **and IZO (in blue) and the corresponding simulated background (red). The background simulations (in red)**  
1050 **obtained using the average diurnal pattern without presence of volcanic plume and adjusting offset is compared with the measurements**

1051 taken at the IZO station (blue) on the same day. The corrected  $\Delta XCO_2$  is presented in black in (C) and (D)  
 1052 concurrently with the  $\Delta XHCl$ , which can be considered as a tracer of the volcanic plume.

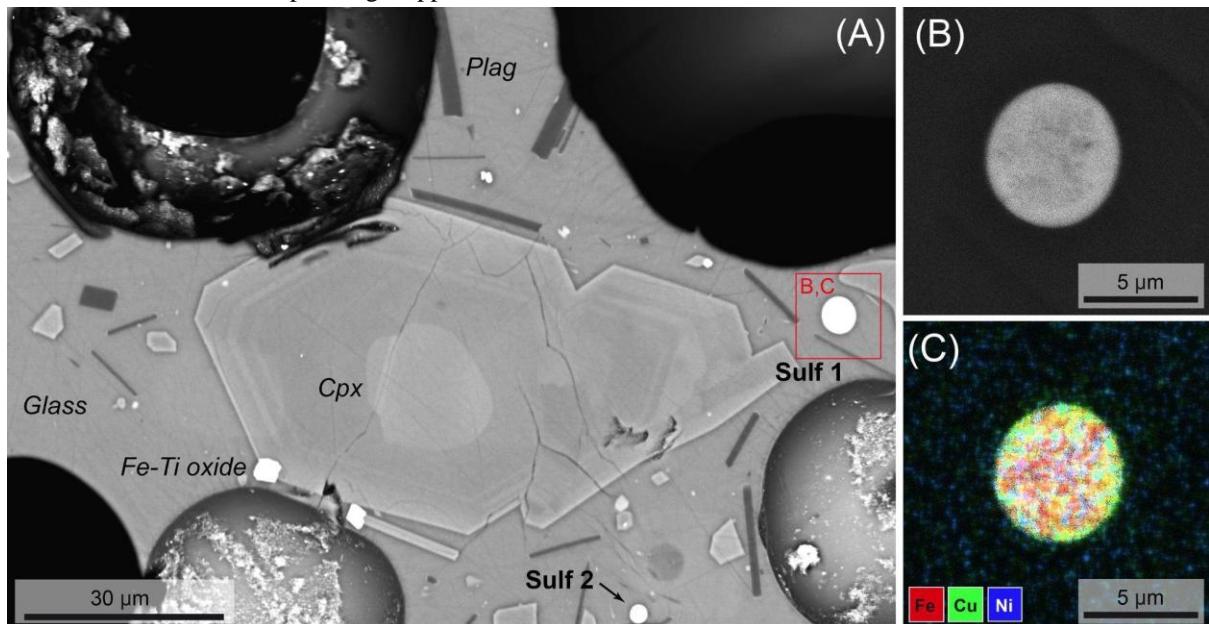


1053

1054 Figure A4: Same as A3 for CO. For CO the procedure only consisted in removing the long-term trend, estimated  
 1055 from the long-term IZO daily average time series.

1056 **Appendix B**

1057 Appendix B presents the tephra compositions acquired with Scanning Electron Microscope and Electron Micro-  
 1058 Probe and elements of the petrologic approaches used in for the estimation of the volatile emissions.



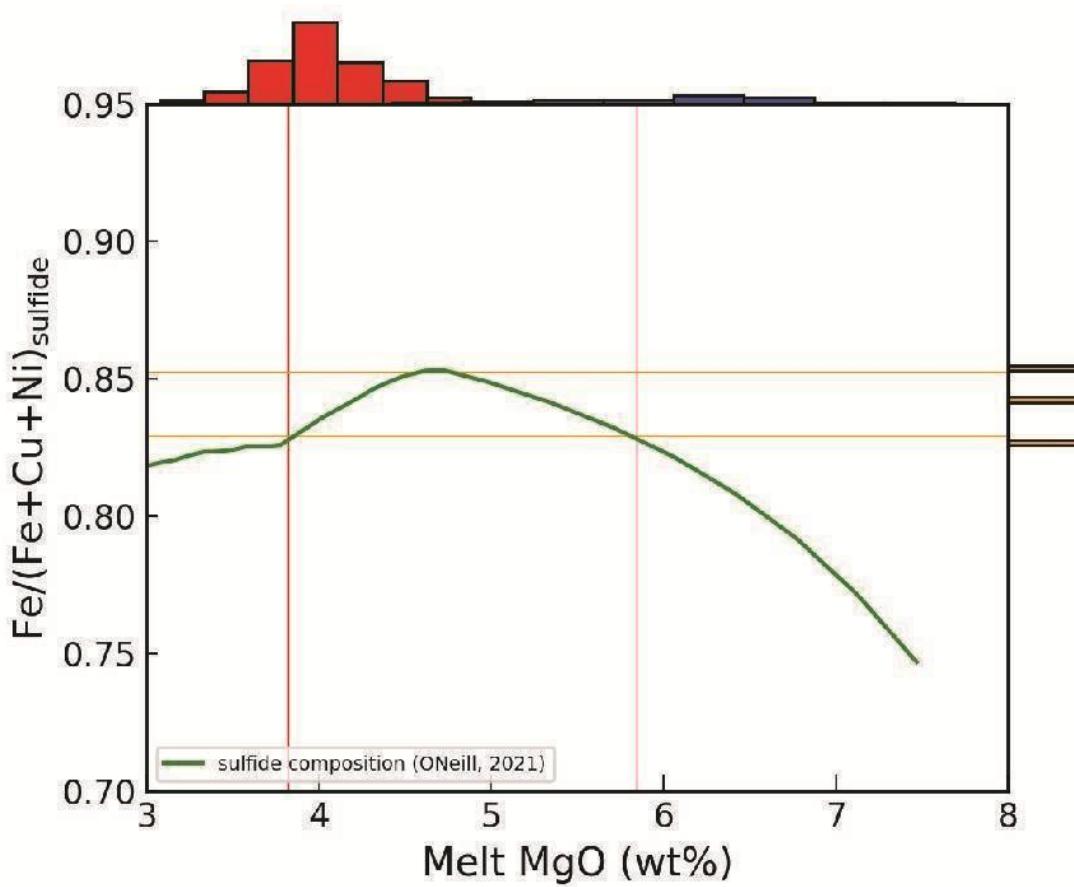
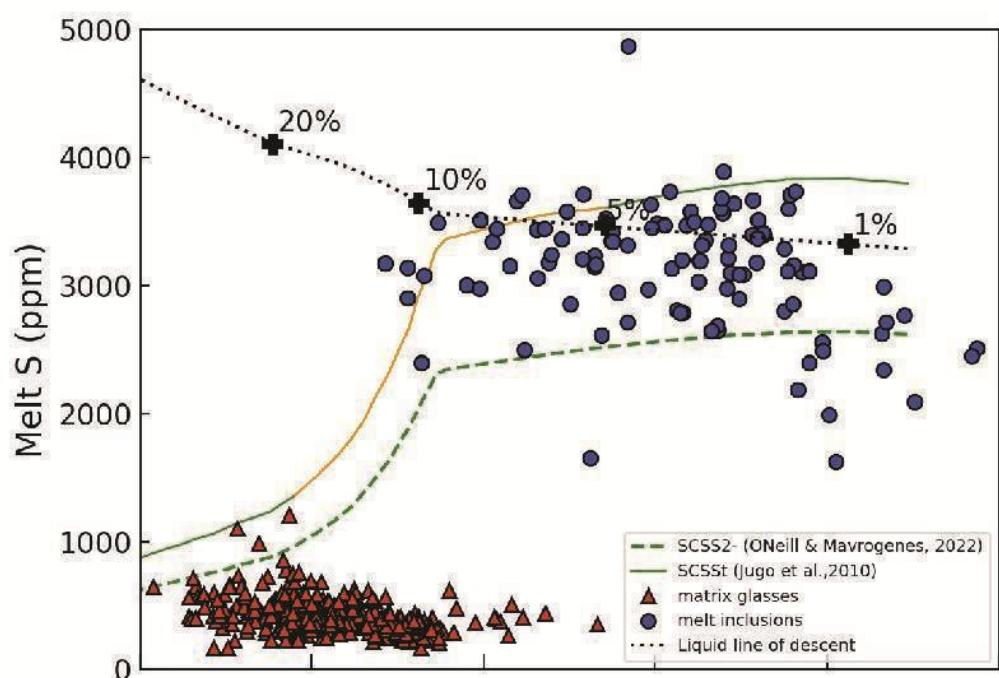
1059

	S	Fe	Ni	Cu
Sulf1	35.7	54.2	2.9	7.3
Sulf2	34.6	53	2.8	9.7

1060

1061 Figure B1: (A) Backscattered electron (BSE) image of a section of LM-2309 tephra sample (Gonzalez-Garcia et al.,  
 1062 2023) displaying two sulfide droplets (Sulf-1 and Sulf-2). (B) Detail BSE image of Sulf-1, and (C) EDX compositional  
 1063 map of Sulf-1, showing zoning in Fe-Cu-Ni sulfides. The images were acquired using (A) a JEOL JSM-7610F gun  
 1064 emission scanning electron microscope operating at 15 kV (IESW, Hannover) (A), and a TESCAN Vega 4 operating  
 1065 at 20kV with EDX Bruker detectors (UCM, Madrid) (B, C). The table below shows their compositions (in wt%),  
 1066 determined by energy dispersive spectroscopy with a Cameca SX-100 electron microprobe (Uni. Bremen).

1067 Figure B2: The upper panel shows the results of the sulfur content at sulfide saturation (SCSS) calculations  
 1068 performed using the model of ONeill (2021) implemented in the open-source Python3 tool PySulfSat (Wieser and  
 1069 Gleeson, 2023). The starting composition is one of the most primitive MI of the literature dataset for the eruption  
 1070 (LM0 G29 Dayton et al. 2024), to which a Petrolog3 (Danyushevsky and Plechov, 2011) crystallization model (with  
 1071 olivine ±clinopyroxene + spinel as crystallizing phase, following Day et al. 2022) is applied at a magma stalling at 3.5  
 1072 kbars and a  $\text{FO}_2$  buffer of NNO+0.4, following Andujar et al. (2025). Given these conditions, the melt is expected to  
 1073 contain a significant proportion of sulfur as sulfate ( $\text{S}^{6+}$ ), rather than sulfide ( $\text{S}^{2-}$ ). Therefore, we used the SCSS<sub>St</sub>  
 1074 model of Jugo et al. (2010), which accounts for mixed sulfur speciation, to evaluate saturation. Only a few inclusions  
 1075 slightly exceed the SCSS<sub>St</sub> curve, consistent with the rarity of sulfide globules in the eruptive products and with the  
 1076 interpretation that sulfide saturation was only reached locally or after some crystallization (Day et al., 2022). The  
 1077 bottom panel shows the modeled composition (Fe/Fe+Ni+Cu) of the sulfide phase precipitating along the liquid line of  
 1078 descent, which is matching the measured compositions between ~4 and 5.8 wt% MgO (after 5-15% crystallization).  
 1079 This range is reported as the orange portion of the liquid line of descent in the upper panel.



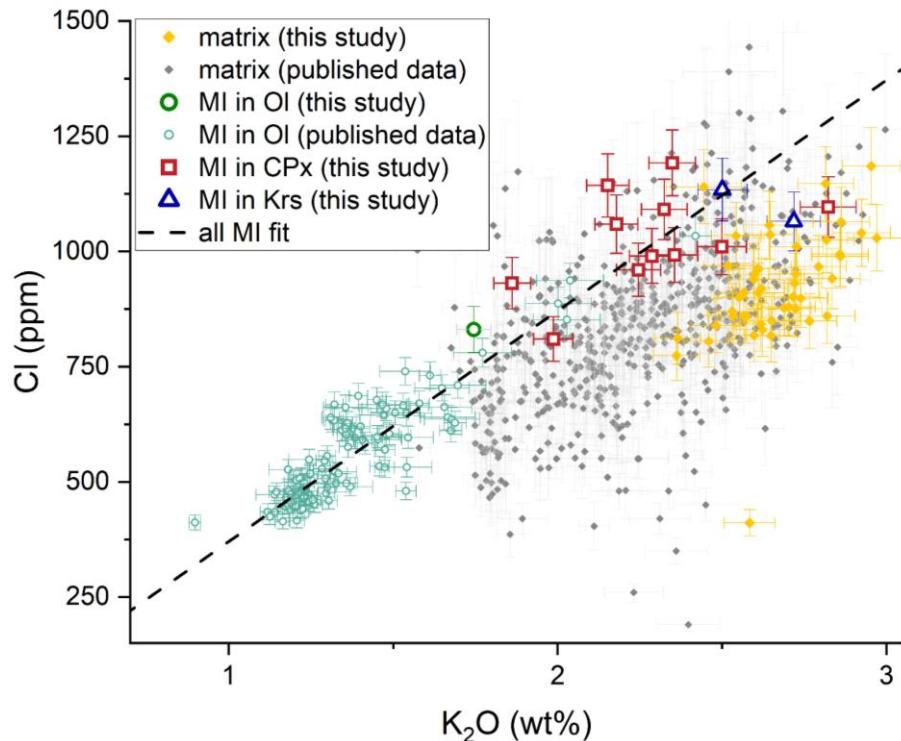


Figure B3: New and previously published Cl content in MIs (green hollow circles) and matrix glass (orange diamonds) with melt evolution, represented by the incompatible K<sub>2</sub>O. MI compositions are from this study, Burton et al. (2023) and Dayton et al. (2024). Ol stands for olivine, CPx, for clinopyroxene and Krs for Kaersutite. Matrix glasses have been measured on tephra samples and are from this study, Burton et al. (2023), Ubide et al., 2023, Dayton et al. (2024), Longpré et al. (2025). The dashed line is the linear regression through the MIs dataset and represents the increase in melt Cl content during crystal fractionation. We calculated the degassed Cl amount as the error-weighted mean difference between each matrix Cl content and the regression line.

## Appendix C

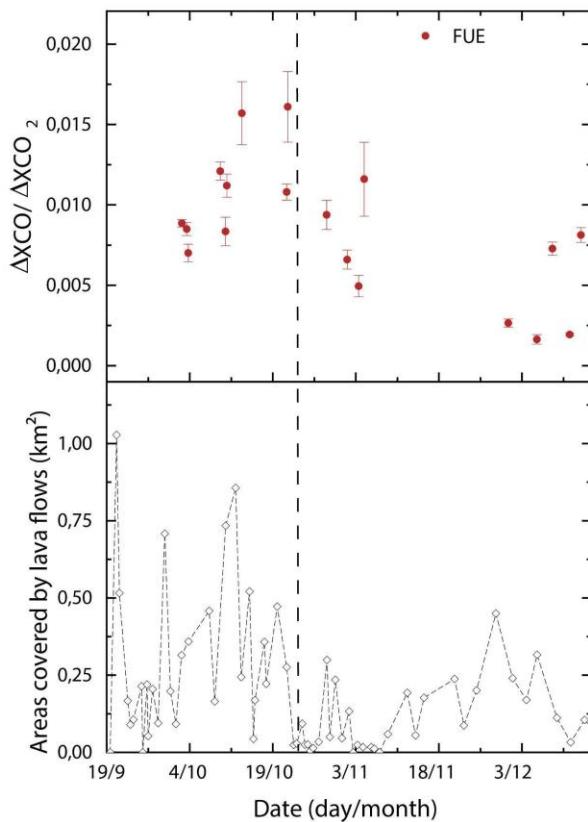
Appendix C details the procedure that we employed to estimate the plume age for the dates reported in Table 5, using the Hysplit forward simulations.

We used high resolution meteorological data derived from the WRF-ARW (Advanced Research Weather Research and Forecasting) model (Powers et al., 2017; Skamarock et al., 2019). The WRF-ARW model is run operationally twice daily, utilizing initial and boundary conditions from HRES-IFS (High Resolution Integrated Forecast System) data provided by ECMWF (European Centre for Medium-Range Weather Forecasts) at a resolution of  $0.09^\circ \times 0.09^\circ$ . The model configuration includes three nested domains with horizontal resolutions of 6 km, 2 km, and 1 km, respectively, and 31 vertical levels, operating in non-hydrostatic mode. Each simulation produces forecasts extending up to 72 hours. The outputs of the WRF-ARW model are converted into the required format for the HYSPLIT model (Hybrid Single-Particle Lagrangian Integrated Trajectory) using the ARW2ARL program. This process produces meteorological data formatted for use in trajectory and dispersion simulations. To prepare the data for HYSPLIT, the WRF-ARW outputs are processed to generate ARL files with a 12-hour temporal span. These files are designed to overlap every 12 hours, ensuring continuous hourly meteorological data coverage. This approach provides a seamless dataset necessary for accurate and uninterrupted backward trajectory and forward simulation calculations. Forward simulations were performed using Hysplit with a standard configuration over a minimum total calculation time of 48h and an hourly time resolution. The plume altitude was taken from the IGN/AEMET data and Milford et al. (2023) for each studied date.

## Appendix D

Appendix D presents the comparison between the  $\Delta XCO/\Delta XCO_2$  measured at FUE using the direct-sun FTIR measurements and the area covered daily by the lava flows, derived from the daily Copernicus EMSR546

1111 mapping. A good agreement is found between the two dataset, indicating a possible contribution of the burning  
 1112 infrastructure and vegetation in our FUE FTIR measurements. It contrasted with the ratios reported by Asensio-  
 1113 Ramos et al. (2025) derived from open-path measurements performed at the N-NW from the eruptive fissure,  
 1114 less affected by this contribution.

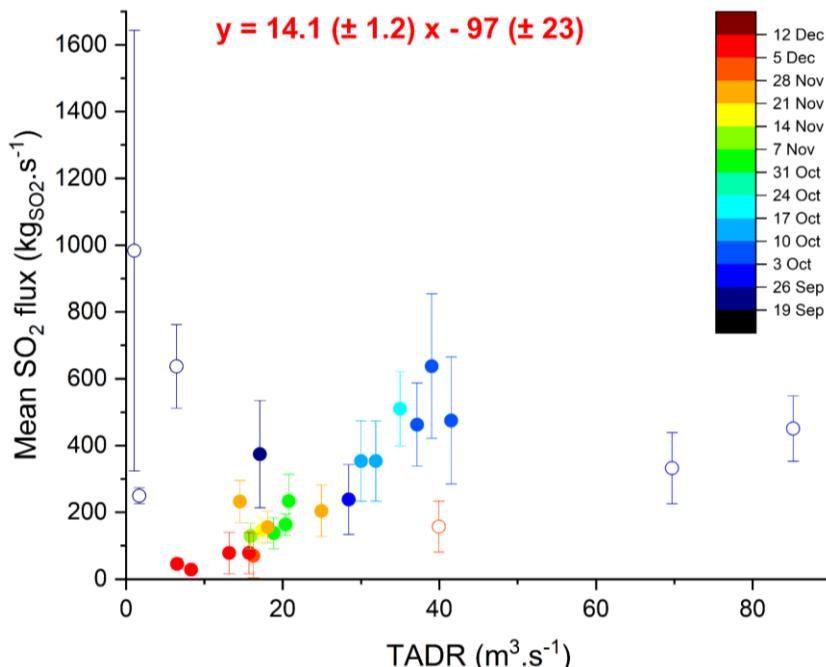


1115

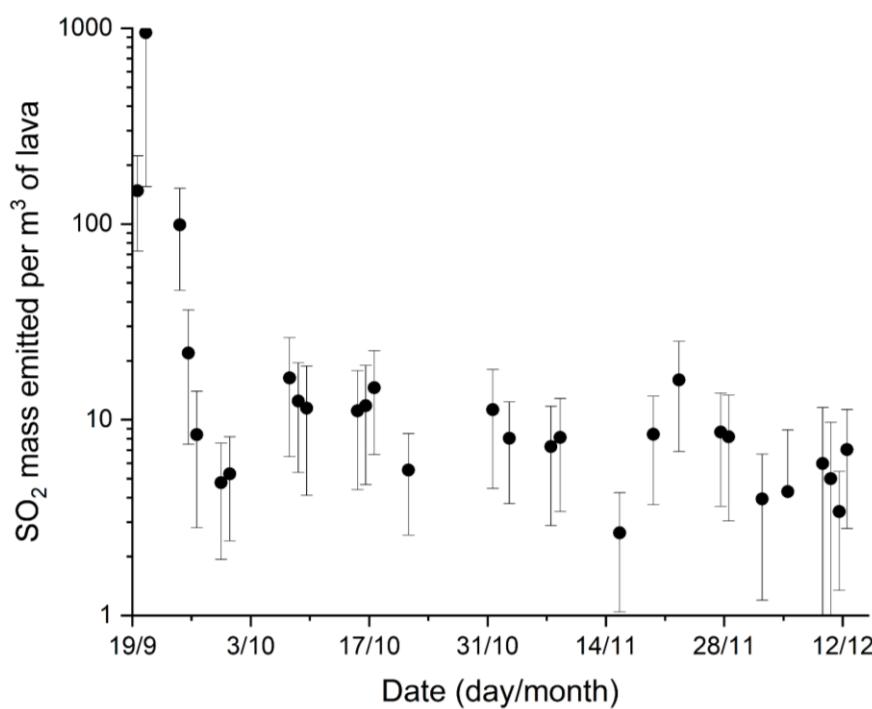
1116 **Figure D1: Comparison between the time series of  $\Delta\text{XCO}/\Delta\text{XCO}_2$  ratios obtained over the whole eruptive period at**  
 1117 **FUE and the daily covered area by the lava flows derived from the daily Copernicus EMSR546 mapping**  
 1118 **(COPERNICUS EMERGENCY MANAGEMENT SERVICE | Copernicus EMS - Mapping).**

1119 **Appendix E**

1120 Appendix E describes the relationship observed between the retrieved daily SO<sub>2</sub> emission fluxes and the lava  
 1121 Time Averaged Discharge Rate (TADR). The TADR estimates the lava volume responsible for the radiant flux  
 1122 measured by satellite (Coppola et al., 2016). We exploit the fact that this volume is also the source of SO<sub>2</sub>  
 1123 emissions (Fig. E1), providing a direct quantification of the amount of S actually degassing (“effective S  
 1124 degassing”), which is usually indirectly derived a posteriori, by the difference between the S content of the  
 1125 primitive magma (melt inclusions) and that remaining in the matrix of the (degassed) eruptive products. This  
 1126 quantity is shown constant through the eruption (E2).



1127  
 1128 **Figure E1: Correlation between the TADR and SO<sub>2</sub> volcanic emission fluxes illustrating an average “effective S  
 1129 degassing” of  $14.1 \pm 1.2$  kg SO<sub>2</sub> per (thermal) cubic meter of lava discharged to the surface. Hollow points correspond  
 1130 to the outliers to the dataset.**



1131  
 1132 **Figure E2: Time series of “effective S degassing”.**

1133                   **8. Data availability**

1134                   FTIR data used in this study are available upon request. In situ surface data at Izaña  
1135                   Atmospheric Observatory contribute to the WMO-GAW Program and are available at the World Data  
1136                   Centre for Greenhouse Gases (WDCGG, <https://gaw.kishou.go.jp/>). TROPOMI data (Copernicus  
1137                   Sentinel-5P) are publicly available from Sentinel-5P data hub at  
1138                   [https://sentinels.copernicus.eu/web/sentinel/data-products/-/asset\\_publisher/fp37fc19FN8F/](https://sentinels.copernicus.eu/web/sentinel/data-products/-/asset_publisher/fp37fc19FN8F/).  
1139                   Petrological dataset is available as Supplementary Material.

1140                   **9. Authors contribution**

1141                   All of the co-authors contributed to the preparation and writing of the manuscript. OG, TB,  
1142                   NT conceptualised the study. OG led the development of the FTIR program at the Izaña Atmospheric  
1143                   Observatory and its long-term operation. OG, RR, AA, VC were in charge of the implementation and  
1144                   operation of the Fuencaliente (La Palma) station during the eruption. They also assured the operation  
1145                   and maintenance of measurements at IZO. OG, NT, WS, ES, SL contributed to the FTIR and DOAS  
1146                   and data analysis. SL and PRS are responsible for the GAW surface measurements at IZO and their  
1147                   processing. NT, TB, RC, WS, OG contributed to the implementation and operation of the combined  
1148                   DOAS-EM27/SUN measurements at Fuencaliente. NT, TB, RC performed the MultiGAS  
1149                   measurements and ash sampling. RC processed the MultiGAS data. DGG, AK performed the SEM  
1150                   and EPMA analyses and helped for the interpretation and discussion of results. CA, MIG contribute to  
1151                   realisation of the Hysplit modeling of the volcanic plume dispersion to estimate the plume age. SR,  
1152                   JLD performed the chemical analysis of the PM<sub>10</sub> and contributed to their interpretation. MIG, SR,  
1153                   PGS, TB, NT contribute to the discussion about the PM<sub>10</sub> measurements. FH helped with the FTIR  
1154                   operating maintenance and data processing. He developed the PROFFAST and PROFFIT retrieval  
1155                   codes and provides continuous support to the group with respect to its use and spectrometer operation.  
1156                   FH and OG led the German–Spanish collaboration and provided precious help with respect to the  
1157                   EM27/SUN measurements within the framework of the COCCON network.

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1195

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