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## **Volcanic Emissions, Plume Dispersion and Downwind Radiative Impacts following Mount Etna Series of Eruptions of 21-26 February 2021**

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2 **Volcanic Emissions, Plume Dispersion and Downwind Radiative Impacts following**  
3 **Mount Etna Series of Eruptions of 21-26 February 2021**

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24 **Key Points:**

- 25 ● Three distinct paroxysmal events took place at Mount Etna from 21 to 26 February 2021  
26 and the plumes were transported towards the north
- 27 ● Plumes dispersion is characterized with Lagrangian modelling initialized with measured  
28 SO<sub>2</sub> emissions and with satellite observations
- 29 ● A series of LiDARs detected and characterized the plumes downwind and their  
30 observations are used to estimate the plumes radiative forcing
- 31

## 32 **Abstract**

33 During the extended activity of Mount Etna volcano in February-April 2021, three distinct  
34 paroxysmal events took place from 21 to 26 February, which were associated with a very  
35 uncommon transport of the injected upper-tropospheric plumes towards the north. Using a synergy  
36 of observations and modelling, we characterised the emissions and three-dimensional dispersion  
37 for these three plumes, we monitor their downwind morphological and optical properties, and we  
38 estimate their radiative impacts at selected locations. With a satellite-based source inversion, we  
39 estimate the emitted sulphur dioxide (SO<sub>2</sub>) mass at an integrated value of 55 kt and plumes  
40 injections at up to 12 km altitudes, which combine to qualify this series as extreme in the eruption  
41 strengths spectrum for Mount Etna. We then combine Lagrangian dispersion modelling, initialised  
42 with measured temporally-resolved SO<sub>2</sub> emission fluxes and altitudes, with satellite observations  
43 to track the dispersion of the three individual plumes. The transport towards the north allowed the  
44 height-resolved downwind monitoring of the plumes at selected observatories in France, Italy and  
45 Israel, using LiDARs and photometric aerosol observations. Volcanic-specific aerosol optical  
46 depths in the visible spectral range ranging from about 0.004 to 0.03 and local daily average  
47 shortwave radiative forcing ranging from about -0.2 to -1.2 W/m<sup>2</sup> (at the top of atmosphere) and  
48 from about -0.2 to -3.5 W/m<sup>2</sup> (at the surface) are found. Both the aerosol optical depth and the  
49 radiative forcing of the plume depends strongly on its morphology (position of the sampled section  
50 of the plume) and composition (possible presence of fine ash).

## 51 **Plain Language Summary**

52 The volcanic plumes from Mount Etna's eruptions disperse usually towards the east, to the  
53 Central-Eastern Mediterranean, where no ground stations are available to monitor and  
54 characterise them. During the extended intense volcanic activity of Mount Etna in February-  
55 April 2021, some of the plumes dispersed towards the north, in an area where many and well-  
56 equipped ground observatories are available. This was a unique and very uncommon possibility  
57 to study the emissions, dispersion dynamics, shape/composition and impacts of Mount Etna's  
58 emissions. We analysed these rare events using all available information layers: satellite  
59 observations, numerical simulation of the plume's dispersion and downwind ground-based  
60 observatories. By coupling these information pieces, we could reconstruct these events and  
61 untangle their impacts from those of a major Saharan dust outbreak that occurred simultaneously.  
62 These events were found quite extreme for the Mediterranean volcanism (extreme emissions and  
63 at altitudes as high as 12 km). We found that this kind of volcanic eruptions have a relevant (and  
64 not yet studied) impact on the upper-tropospheric composition and regional climate in the  
65 Mediterranean area. The plumes, after regional-to-continental dispersion, exhibit a complex  
66 filamentary morphology and, and most likely, a complex composition coming from the interplay  
67 of sulphur compounds and ash emissions.

## 68 **1 Introduction**

69 Volcanic activity, spanning from passive degassing to explosive eruptions, can have relevant  
70 downwind impacts on the atmospheric composition (e.g. von Glasow et al., 2009), aerosol  
71 properties (e.g. Sellitto et al., 2017), air quality and health (e.g. Michaud et al., 2005), the formation  
72 and lifetime of clouds (e.g. Malavelle et al., 2017) and the radiative balance (e.g. Andersson et al.,  
73 2015, Sellitto et al., 2020, Kloss et al., 2021). Associated with the volcanic activity are both  
74 primary emissions of gases and particles, like water vapour, carbon dioxide, sulphur dioxide (SO<sub>2</sub>)  
75 and ash, and secondary aerosols, in particular the tiny-sized sulphate aerosols (SA) forming from

76 the gaseous- and liquid-phase oxidation/nucleation from SO<sub>2</sub> primary emissions. All these  
77 effluents can have environmental and climatic impacts downwind the dispersion pathways of the  
78 volcanic plumes. The SA, in particular, are the main contributors of the volcanogenic modulation  
79 of the Earth's radiative balance and, then, of the local, regional or global climate system (Sellitto  
80 and Briole, 2015). This is due to their high reflectivity properties in the solar spectral range,  
81 mirrored by their very high single scattering albedo (SSA) (Krotkov et al., 1997), and their  
82 relatively long atmospheric residence time, linked to their tiny typical sizes (Stevenson et al.,  
83 2003). While the importance of moderate-to-strong volcanic eruptions, with plume injection in the  
84 stratosphere, on the Earth's climate system is now relatively well established (Santer et al., 2014;  
85 Ridley et al., 2017), the role on the regional climate system of smaller eruptions, with injection in  
86 the troposphere, as well as local impacts of persistent passive degassing activity (Sellitto et al.,  
87 2020), is not yet well understood and quantified (Oppenheimer et al., 2011).

88 The downwind impacts of tropospheric volcanic plumes and their spatio-temporal extent depend  
89 critically on the interplay of three main aspects: 1) the internal geochemical and geophysical  
90 processes linked to magma degassing and plume injection, and, once the plume is released, 2) its  
91 atmospheric physico-chemical evolution, and 3) the local and regional atmospheric dynamics  
92 driving the plume dispersion. To characterise these three components of the plumes release and  
93 evolution, as well as to estimate downwind impacts, a coordinated synergy of ground-based and  
94 satellite observations, and the modelling of transport and physical-chemical evolution, are  
95 necessary. Such synergies have been exploited in a number of studies in the past (e.g. Haywood et  
96 al., 2010; Webley et al., 2012; Sellitto et al., 2016; Kloss et al., 2021).

97 In this paper, we couple ground-based and satellite observations with dispersion and impact  
98 modelling to study a specific phase of the extended activity of Mount Etna occurring during the  
99 period February-April 2021. We focus, in particular, on the series of paroxysmal eruptions during  
100 the period from 21 to 26/02/2021. This period was characterized by an uncommon local dynamics  
101 that allowed a rare transport of the emitted plumes towards the north, where a number of ground-  
102 based observatories could be used to monitor the morphological, optical and radiative properties  
103 of the plumes. Vertically-resolved information can be retrieved from stations equipped with  
104 LiDAR (Light Detection And Ranging) systems. From several points of view, this is a unique case-  
105 study to gather better insights into the evolution of a moderate volcanic eruption: the extended  
106 period of the activity, the unusual intensity of the eruptions, the relatively high altitude of the  
107 atmospheric injection and the transport towards areas with a relatively large density of potential  
108 downwind observations. Here we show that, in such conditions, a synergistic  
109 observational/modelling approach can contribute a more complete end-to-end characterisation of  
110 the volcanic events, from emissions to dispersion to downwind impacts.

111 The paper is organized as follows. In Sect. 2 we introduce and describe the array of observations  
112 and modelling tools used in this work. In Sect. 3 we provide a qualitative description of the eruptive  
113 series under investigation, and we frame this volcanic activity into the wider Mount Etna's internal  
114 geophysical context. In Sect. 4 we show results for the characterisation of the emission, dispersion  
115 and impacts of this series of events. We draw conclusions in Sect. 5.

## 116 2 Data and Methods

### 117 2.1 Volcanic SO<sub>2</sub> emission flux rates and total mass estimations from satellite-based source 118 inversion

119 The SO<sub>2</sub> emission flux rates time series and total mass have been obtained by exploiting the  
120 measurements collected from the Spinning Enhanced Visible and InfraRed Imager (SEVIRI). The  
121 SEVIRI instrument is a multispectral imager on board the Meteosat Second Generation (MSG)  
122 geostationary satellite. It has 12 spectral channels from visible to Thermal InfraRed (TIR) spectral  
123 ranges and a spatial resolution of 3 km at sub-satellite point. The temporal resolution varies from  
124 5 (rapid scan mode over Europe and Northern Africa) to 15 min (Earth full disk) (see  
125 <https://www.eumetsat.int/meteosat-second-generation> for more details on MSG platform and  
126 SEVIRI instrument). The SEVIRI measurements are collected in real time from the Multimission  
127 Acquisition SysTem (MAST) developed at Istituto Nazionale di Geofisica e Vulcanologia (INGV)  
128 (Stelitano et al., 2020). All the SEVIRI images used in this work have been resampled in a regular  
129 grid of 3×3 km<sup>2</sup> and processed every 15 minutes.

130 The SO<sub>2</sub> columnar abundance is computed by applying the Volcanic Plume Retrieval (VPR)  
131 procedure (Guerrieri et al., 2015). This approach is based on the computations of the plume  
132 transmittances in the SEVIRI TIR bands centred at 8.7, 10.8, and 12 μm, obtained through linear  
133 equations based on the original image radiances and a new image with volcanic plume signal  
134 removed; the latter is computed by means of a linear regression of the radiance values outside the  
135 edges of the plume itself. The SO<sub>2</sub> estimates are obtained from 8.7 μm transmittances, after  
136 eliminating the contribution of ash and ice particles computed from 10.8 and 12 μm channels. The  
137 main advantage of VPR is that the only input required is the volcanic cloud top height.

138 From the SO<sub>2</sub> abundance, the SO<sub>2</sub> emission flux rate is obtained by applying the “traverse  
139 approach” (Corradini et al., 2008; Merucci et al., 2011) considering a transect placed at 30±1.5 km  
140 from the summit craters and a wind speed derived from the ARPA (Agenzia Regionale per la  
141 Protezione Ambientale) database (Scollo et al., 2009). Such a distance for the reference transect  
142 has been selected to minimize the retrieval uncertainties induced by both the opacity of the pixels  
143 too close to the craters and the dilution of the pixels too far from the emission. Knowing the wind  
144 speed, the SO<sub>2</sub> flux at 30 km is then reported to 0 km over the vents (Corradini et al., 2020; 2021).  
145 From SO<sub>2</sub> emission rate, the SO<sub>2</sub> total emitted mass is obtained by temporal integration.

146 The volcanic cloud top height is obtained by applying the consolidated “dark pixel” procedure  
147 (Prata et al., 2001; Scollo et al., 2014; Corradini et al., 2018), based on the comparison between  
148 the minimum SEVIRI 10.8 μm brightness temperature of a pixel contained in a fixed area over the  
149 summit craters and an atmospheric temperature profile measured approximately in the same area  
150 and at the same time of satellite acquisition (Scollo et al., 2014; Corradini et al., 2018). In this  
151 case an area of 19×19 SEVIRI pixels and ARPA database were used respectively. This method is  
152 based on the assumption that the plume is opaque and in thermal equilibrium with its environment.

### 153 2.2 Model simulations of plume dispersion and impacts

#### 154 2.2.1 Plume dispersion with the FLEXPART Lagrangian model

155 We simulate the dispersion of the SO<sub>2</sub> plumes emanated by the Mount Etna activity during the  
156 period 21-26/02/2021 using the Lagrangian dispersion model FLEXPART (Pisso et al., 2019). The  
157 simulations are initialized with the emission rates and altitude estimated with SEVIRI, using the  
158 methods described in Sect. 2.1. As meteorological inputs, our FLEXPART simulations use

159 European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-5 reanalysis data at 30  
 160 km horizontal resolution and 137 height levels (100–200 m vertical resolution in the troposphere,  
 161 1–2 km in the lower stratosphere). The FLEXPART outputs are averaged over 30 min intervals  
 162 and are given at  $0.1^\circ \times 0.1^\circ$  horizontal grid and 9 altitude levels, from the surface to 13 km altitude,  
 163 with a vertical resolution of 1 km, from 5 to 13 km, and a unique vertically-broad layer, from  
 164 surface to 5 km. Dry and wet deposition, as well as chemical depletion, are considered in the  
 165 simulations, using the default FLEXPART sink parameterization for SO<sub>2</sub>.

## 166 2.2.2 Plume radiative forcing estimations with the UVSPEC radiative transfer model

167 We estimate the radiative impact of the Mount Etna plumes for the events under investigation  
 168 using the radiative transfer model UVSPEC and the LibRadtran package (Emde et al., 2016). The  
 169 equinox-equivalent clear-sky daily-average shortwave (integrated between 300 and 3000 nm)  
 170 surface and top of the atmosphere (TOA) direct radiative forcings (RF) are estimated with a similar  
 171 methodology as done by Sellitto et al. (2016, 2020); please refer to these previous works for more  
 172 details on the setup of the radiative model.

173 The background atmospheric state is set using the AFGL (Air Force Geophysics Laboratory)  
 174 winter mid-latitudes climatological standard. The surface albedo is set to 0.15, independent from  
 175 wavelength, which is a typical average shortwave albedo for the vegetated surfaces underlying the  
 176 locations associated with our study. Baseline and volcanically-perturbed radiative transfer  
 177 calculations are carried out using different aerosol layers configurations: using aerosol extinction  
 178 profiles at downwind LiDAR stations, as volcanically-perturbed conditions, and with the same  
 179 aerosol extinction profiles with the identified volcanic plume layer removed, as non-volcanic  
 180 baseline (see specific configurations in Sec. 4.5). Different hypotheses must be considered for the  
 181 non-measured optical parameters of volcanic aerosols. The spectral variability of the volcanic  
 182 aerosol extinction, the single scattering albedo and the angular distribution of the scattered  
 183 radiation are not directly measured and can be quite uncertain due to the possible presence of both  
 184 SA and a fraction of residual ash. Values of the Ångström exponent (a compact measure of the  
 185 spectral variability of the particle extinction and linked to the average size of the particles in the  
 186 plume), of the single scattering albedo (linked to the absorption properties of the aerosol layer)  
 187 and of the asymmetry parameter (linked to the angular distribution of the aerosol layer) are selected  
 188 for each case described in Sect. 4.5; please refer to this section for the details of these different  
 189 hypotheses. Different runs are realized, for all experiment, for different solar zenith angles (SZA)  
 190 to sample the full day. The daily-average shortwave TOA RF for the volcanically-perturbed  
 191 aerosol layer is calculated as the SZA-averaged upward diffuse irradiance for the baseline  
 192 simulation minus the volcanically-perturbed run, integrated over the whole shortwave spectral  
 193 range. The shortwave surface radiative forcing is calculated as the SZA-average downward global  
 194 (direct plus diffuse) irradiance for the volcanically-perturbed run minus the baseline, integrated  
 195 over the whole spectral range.

## 196 2.3 Satellite Observations of volcanic plumes

### 197 2.3.1 SO<sub>2</sub>, SA and dust observations with IASI

198 The Infrared Atmospheric Sounder Interferometer (IASI) is a Fourier transform spectrometer  
 199 covering the large infrared spectral range between 645 and 2760 cm<sup>-1</sup>, with a relatively high  
 200 spectral resolution (0.5 cm<sup>-1</sup>) and a relatively small radiometric noise (noise equivalent spectral  
 201 radiance of about 20 mW/(cm<sup>-1</sup>m<sup>2</sup>sr) around 1000 cm<sup>-1</sup>). The IASI instrument observes the Earth's

202 atmosphere and surface with a downward-looking geometry, with circular footprints of down to  
203 12 km radius at the nadir spaced by 25 km, and with a swath of 2200 km (Clerbaux et al., 2009).  
204 The instrument series are on board MetOp-A -B and -C spacecrafts since 2006, 2012 and 2018,  
205 respectively. Each instrument of the series provides a near-global coverage every 12 h. For this  
206 work, we have used IASI observations of SO<sub>2</sub>, SA and dust.

207 The IASI daily SO<sub>2</sub> dataset includes SO<sub>2</sub> columns and volcanic plume altitude. The SO<sub>2</sub> partial  
208 columns at 6 altitude intervals between 5 and 25 km are retrieved from IASI observations by  
209 exploiting the correlation between brightness temperature differences and SO<sub>2</sub> total columns with  
210 assumptions on the SO<sub>2</sub> plume altitudes (Clarisse et al., 2012). From these partial columns  
211 profiles, the total column is obtained by vertical integration. In addition, the altitude of the SO<sub>2</sub>  
212 plume is estimated by using a sensitive trace gas detection method for high spectral infrared  
213 measurements (Clarisse et al., 2014).

214 The detection of SA and its aerosol-type-specific optical depth is provided by the AEROIASI-  
215 H<sub>2</sub>SO<sub>4</sub> retrieval algorithm (Guermazi et al., 2021). The AEROIASI-H<sub>2</sub>SO<sub>4</sub> algorithm is based on  
216 a self-adapting Tikhonov-Phillips regularization method, built around the radiative transfer model  
217 Karlsruhe Optimized and Precise Radiative transfer Algorithm (KOPRA) (Stiller et al., 2000). This  
218 method iteratively fits each IASI spectrum at 20 spectral micro-windows by adjusting the vertical  
219 profile of SA number concentration, jointly with water vapour profiles and surface temperature.  
220 The SA are assumed as spherical droplets of an aqueous solution of sulphuric acid (H<sub>2</sub>SO<sub>4</sub>), and  
221 the corresponding refractive indices are taken from Biermann et al. (2000). The information on the  
222 total SA-specific optical depth is mainly provided by the selective absorption of the undissociated  
223 H<sub>2</sub>SO<sub>4</sub> in the SA solution droplets. The interference of the co-existent SO<sub>2</sub> is avoided by using  
224 an operational spectral micro-window around 900 cm<sup>-1</sup>, a spectral region characterised by H<sub>2</sub>SO<sub>4</sub>  
225 absorption bands but outside the main SO<sub>2</sub>-sensitive region around 1100-1200 cm<sup>-1</sup>.

226 For the characterization of the three-dimensional distribution of desert dust plumes during the  
227 event analyzed in the paper, we use the AEROIASI-Dust approach (Cuesta et al., 2015, 2020). The  
228 AEROIASI-Dust algorithm derives vertical profiles of desert dust, in terms of extinction  
229 coefficient at 10 μm, from individual thermal infrared spectra measured by IASI satellite sensor in  
230 cloud-free conditions, both over land and ocean. This method iteratively fits each IASI spectrum  
231 at 12 spectral micro-windows by adjusting the vertical profile of dust aerosol abundance jointly  
232 with surface temperature. The information on the vertical distribution of dust is mainly provided  
233 by their broadband radiative effect, which includes aerosol thermal emission depending at each  
234 altitude on the vertical profile of temperature. As for AEROIASI-H<sub>2</sub>SO<sub>4</sub>, the AEROIASI-Dust  
235 algorithm uses the radiative transfer model Karlsruhe Optimized and Precise Radiative transfer  
236 Algorithm (KOPRA) (Stiller et al., 2000). The current paper uses AEROIASI retrievals from  
237 version described by Cuesta et al., (2020) but using a complex refractive index derived by Di  
238 Biagio et al. (2017) with a dust sample from a desert in Mali (which shows a smaller difference  
239 between IASI and simulated spectra than that used by Cuesta et al. (2020)).

### 240 2.3.2 SO<sub>2</sub> and AI observations with Sentinel-5p TROPOMI

241 The TROPOspheric Monitoring Instrument (TROPOMI), onboard the Copernicus Sentinel 5  
242 Precursor (S5p) spacecraft, is a joint project of the Netherlands Space Office and the European  
243 Space Agency (Veefkind et al., 2012). The TROPOMI is a satellite-based spectrometer operating  
244 in the ultraviolet/visible/near infrared spectral region and at a nadir-viewing geometry. It provides  
245 observation of key atmospheric constituents at the unprecedented high spatial resolution of down

246 to  $7 \times 3.5 \text{ km}^2$ . In the present work, we use the offline Level 2 datasets of Aerosol Absorbing Index  
247 and the total SO<sub>2</sub> column.

248 The UltraViolet Aerosol Absorbing Index (UVAI) is a compact parameter used to visualize the  
249 presence of absorbing aerosols in the sampled airmasses. It is based on the spectral ratio of the  
250 measured top of the atmosphere reflectance and a pre-calculated theoretical reflectance for a  
251 Rayleigh scattering-only atmosphere, at a given pair of UV wavelength. Positive residuals of the  
252 observed and modelled reflectances are linked to the presence of UV-absorbing aerosols, like dust  
253 and smoke. Negative residuals may indicate the presence of non-absorbing aerosols, while values  
254 close to zero are found in the presence of clouds or no aerosols. As the UVAI is dependent on  
255 different aerosol layer optical and morphological properties, and on the underlying surface  
256 reflectance, in this paper we use this parameter only as a general indication of the presence of dust.  
257 The total column SO<sub>2</sub> is retrieved using a DOAS (Differential Optical Absorption Spectroscopy)  
258 method. The SO<sub>2</sub> slant column density (SCD), i.e. the SO<sub>2</sub> concentration along the mean light  
259 path through the atmosphere, is derived by fitting cross-sections in a spectral range characterised  
260 by the absorption of SO<sub>2</sub> (in this case, at different spectral fitting micro-window between 310 and  
261 390 nm, depending on the SO<sub>2</sub> burden). After different calibrations and corrections of the derived  
262 SO<sub>2</sub> SCD, this latter is converted to vertical columns using the air mass factor from radiative  
263 transfer calculations. For more details about the TROPOMI total column SO<sub>2</sub> product, please refer  
264 to Theys et al. (2017).

### 265 2.3.3 SO<sub>2</sub> observations with OMPS-NM

266 The Ozone Mapping and Profiler Suite Nadir Mapper (OMPS-NM) flies on the Suomi National  
267 Polar-orbiting Partnership (Suomi-NPP) satellite since 2012 and measures the Earth backscattered  
268 UV/visible radiation at a nadir-viewing geometry (Ditman et al., 2002). In this work, the Level 2  
269 height-resolved SO<sub>2</sub> product is used. For this product, a direct vertical column fitting algorithm is  
270 used to retrieve the SO<sub>2</sub> column amounts in the lower (centered at 2.5 km), middle (centered at  
271 7.5 km) and upper (centered at 11 km) troposphere, as well as the lower stratosphere (centered at  
272 16 km).

### 273 2.3.3 Aerosol classification with CALIOP

274 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) is a LiDAR system onboard  
275 the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) spacecraft ; it  
276 flies at about 700 km altitude in a sun-synchronous orbit and is part of the so-called A-Train  
277 (Winker et al., 2010). The CALIOP space LiDAR is in orbit since April 2006 and is still in  
278 operation. It provides vertical profiles of aerosols and clouds at about 01:30 and 13:30 local time.  
279 In this work, we use aerosol detections of the Vertical Feature Mask level 2 product version 4.2,  
280 at varying horizontal resolution, as well as the related aerosol types classification information.

## 281 2.4 Ground-based observations

### 282 2.4.1 MPLNET LiDAR observations at Sede Boker site

283 The National Aeronautics and Space Administration (NASA) Micropulse Lidar Network  
284 (MPLNET; Welton et al., 2001) project was established in 1999 in support of the NASA Earth  
285 Observing System (EOS; Wielicki et al., 1995). During the last two decades, MPLNET  
286 observations have significantly contributed to fundamental studies and applications on climate



287 change and air quality studies and in support for NASA satellite and sub-orbital missions (Lolli et  
288 al., 2018, Bilal et al., 2019, Lolli et al., 2020, Lewis et al., 2020, Campbell et al., 2021). This  
289 federated global network is constituted by homogeneous commercially available Micropulse  
290 LiDARs (MPL) instruments manufactured by Droplet Measurement Technology. The MPLNET  
291 instruments are single-wavelength LiDAR systems that use a diode-pumped Nd:Yag laser at 532  
292 nm, with a 1-minute temporal resolution and 75-meter vertical resolution. The MPLNET network  
293 is active during the whole day and night and operates in all weather conditions, in the limit of laser  
294 signal extinction. The MPLNET network is deployed at global scale and cover all latitudes bands.  
295 For this study, the observations from Sede Boker (30.8° N, 34.8° E, 480 m a.s.l.) permanent  
296 MPLNET site are specifically used. To retrieve the vertically-resolved optical aerosol and cloud  
297 properties from a single wavelength LiDAR, strong assumptions, that limit retrieval accuracy, are  
298 needed. To reduce this uncertainty, MPLNET LiDAR network deploys the instruments, if possible,  
299 together with an Aerosol Robotic Network (AERONET; Holben et al., 1998) sun-photometer to  
300 constrain the LiDAR equation and reduce the retrieval errors (Welton et al, 2000). More details on  
301 the full MPLNET standard data product suite, including aerosols, have been provided by Welton  
302 et al. (2018). The standard, automated, MPLNET products are not designed to accurately capture  
303 elevated volcanic plumes due to their transient nature, high altitude, and unique microphysical  
304 properties relative to the local aerosols that limit the usefulness of the constrained aerosol retrieval  
305 approach. Instead, MPLNET provides a custom retrieval method that bypasses the automated  
306 aerosol height algorithm, and requires manual specification of the plume altitude (for this study  
307 based on inspection of the lidar signal and volume depolarization ratio profiles). Upon  
308 specification of the plume base and top, the optical depth of the plume is determined using the  
309 attenuation of the lidar signal and a molecular scattering (Lewis et al, 2016). The retrieval of plume  
310 properties is calculated using the same MPLNET constrained retrieval algorithm using the plume  
311 optical depth. This process often results in few retrievals during the plume's advection over the  
312 lidar site as calculation of the plume optical depth is difficult with noisy signals. Instead, the Lidar  
313 Ratios from these few retrievals are averaged, and the result is used to determine the plume  
314 properties by fixing the Lidar Ratio using the classic Fernald lidar algorithm (Fernald 1984). The  
315 retrieved lidar ratios were in good agreement with several LiDAR studies on volcanic emissions  
316 (e.g., Prata et al., 2017) and a fixed value of 50 sr was used. Data is freely available on MPLNET  
317 website (<https://mplnet.gsfc.nasa.gov/>).

#### 318 2.4.2 The ACTRIS-Fr OHP LiDAR Observatory

319 The Observatoire de Haute-Provence (OHP) located in southern France (43.9° N, 5.7° E, 670 m  
320 a.s.l.) is one of the NDACC (Network for the Detection of Atmospheric Composition Change)  
321 alpine stations, equipped with a variety of LiDAR instruments for monitoring of the lower and  
322 middle atmosphere.

323 The longest continuous LiDAR data record at OHP is provided by a Rayleigh-Mie-Raman LiDAR  
324 for temperature and aerosol measurements (hereafter referred to as LTA) operating at the  
325 wavelength of 532 nm. The LTA instrument (Keckhut et al., 1993) has provided routine  
326 measurements for over 3 decades with a mean measurement rate of 10-12 nighttime acquisition  
327 nights per month and a typical duration of acquisition of 3-5 hours. To retrieve the vertical profiles  
328 of aerosol backscatter and extinction, the raw lidar returns from three LTA elastic channels,  
329 covering respectively troposphere, lower stratosphere and upper stratosphere, are merged and a  
330 Fernald-Klett inversion method is applied (Fernald, 1984), assuming a constant LiDAR ratio of 50  
331 sr. The retrieved backscatter and extinction coefficients are reported with a time resolution of 1

332 minute and a vertical resolution of 15 m. The scattering ratio is then computed as a ratio of total  
 333 (molecular plus aerosol) to molecular backscattering, where the latter is derived from ECMWF  
 334 meteorological data. A more detailed description of the instrument, aerosol retrieval and error  
 335 budget is provided by Khaykin et al. (2017) and references therein.

336 The ultraviolet tropospheric differential absorption LiDAR (LiO3tr) also operated at OHP at the  
 337 same time as the LTA LiDAR. It records backscatter signal at 316 nm for ozone monitoring  
 338 (Ancellet et al. 1997, Gaudel et al. 2015). The 316 nm signal is very weakly absorbed by ozone in  
 339 the troposphere and can be used for aerosol backscatter monitoring between 3 and 13 km altitude.  
 340 As for the LTA instrument, the Fernald-Klett inversion method (Fernald, 1984) is applied  
 341 assuming a constant LiDAR ratio of 50 sr at 316 nm and a 1.01 scattering ratio close to a molecular  
 342 return at 11 km. Since no overlap correction is applied in order to match the LTA retrieval at 3  
 343 km, a scattering ratio uncertainty of the order of 10% is expected below 4 km.

344 The OHP observatory is also equipped with an automatic LiDAR (CIMEL model CE376)  
 345 sounding in the troposphere from ~200 m to ~12 km of altitude at the wavelength of 532 nm,  
 346 named GAIA and operational since 2019. The GAIA system is also equipped with a depolarization  
 347 channel at the same wavelength to gather more information on the nature and optical properties of  
 348 aerosols, mostly in the boundary layer and in the free troposphere. It provides data at a frequency  
 349 of 1 mn and the datasets are then averaged on 10 min slices. Its spatial resolution is 15 m. The  
 350 scattering ratio is computed as for the LTA LiDAR, but interpolating radiosoundings collected at  
 351 OHP launched every week in the late morning. The data are corrected for instrumental deadtime,  
 352 overlap in the first hundreds of meters of altitude (using an algorithm called CHECK delivered by  
 353 the PHOTONS-AERONET network), sky background and squared altitude to take into account  
 354 the quadratic decrease of the signal in function of altitude. A Haar wavelet method (Brooks 2003,  
 355 Cohn and Angevine 2000) can then be used to retrieve the boundary layer height. The GAIA data  
 356 are inverted using the Klett method (Klett 1981).

357

#### 358 2.4.3 The ACTRIS Napoli LiDAR observatory

359 The Napoli LiDAR observatory (Naples, Italy, 4.18° E, 40.84° N, 118 m a.s.l.) is part of the  
 360 ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure) research infrastructure, a pan-  
 361 European distributed research infrastructure for short-lived atmospheric constituents producing  
 362 high-quality data in the area of atmospheric science (Wandinger et al., 2020). The Napoli  
 363 observatory includes passive and active remote sensing systems and near-surface sampling  
 364 systems, for atmospheric studies. Thanks to its quite central location in the Mediterranean basin,  
 365 it represents a strategic location to study optical and microphysical properties of the aerosols  
 366 coming from local sources and long-range transport.

367 The station is equipped with a multiwavelength elastic/Raman LiDAR device able to detect elastic  
 368 signals at 355 nm, 532 nm and 1064 nm, Raman N<sub>2</sub> echoes at 386 nm and 607 nm and aerosol  
 369 depolarization at 532 nm. Details on this LiDAR system are reported by Boselli et al. (2021).  
 370 Vertical profiles of the aerosol extinction coefficient and linear depolarization ratio are shown and  
 371 discussed in this paper. The aerosol extinction coefficient profiles were retrieved using the  
 372 procedure introduced by Ansmann et al (1990). The calibrated particles linear depolarization  
 373 profiles were obtained from the backscattered light polarized along both perpendicular and parallel

374 directions respect to the laser beam polarization and following the inversion procedure reported  
 375 by Biele et al. (2000) and Freudenthaler et al., (2006).

### 376 2.4.3 The AERONET observations

377 Total column measurements of the aerosol optical depth (AOD), Ångström exponent and single  
 378 scattering albedo at several wavelengths, among others aerosol parameters, are carried out globally  
 379 with Cimel sun photometers, as part of the AERosol RObotic NETwork (AERONET; see the  
 380 AERONET website at <http://aeronet.gsfc.nasa.gov> and the description by Holben et al., 1998, for  
 381 further details). In this manuscript we use the AERONET observations for these three aerosol  
 382 parameters at the selected station of Sede Boker, OHP and Napoli, which are part of the  
 383 AERONET network.

## 384 **3 The Mount Etna activity in February and March 2021**

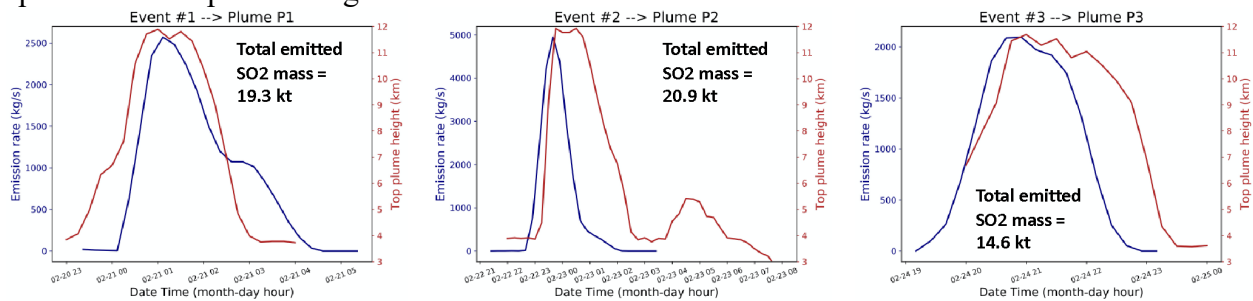
385 Between February and April 2021, Mount Etna experienced an intense eruptive activity from the  
 386 South East Crater (SEC) consisting of astonishing short-lasting lava fountain eruptions coupled  
 387 with lava effusion and episodic pyroclastic density currents. The activity took place while all the  
 388 summit craters, Bocca Nuova, Voragine, North-East crater, were displaying a mild strombolian  
 389 activity and lava effusion at Voragine and Bocca Nuova craters continuously since September  
 390 2019. Differently than the other three summit craters, over the same period SEC was episodically  
 391 erupting but featured a slow and gradual energy increase of its eruptive activity. Eruption,  
 392 eventually intensified between December 2020 and January 2021 to abruptly escalate on the  
 393 afternoon of 16 February 2021, starting to produce a sequence of lava fountaining which ended on  
 394 1st April 2021. Over these two months, 17 paroxysmal episodes were produced with a recurrent  
 395 characterisation, consisting of the resumption of strombolian activity growing in intensity, lava  
 396 flow effusion, and transition from discrete explosions to the continuous gas and lava jetting typical  
 397 of lava fountaining. Overall, each lava fountain lasts few hours while producing the sudden  
 398 spreading in the atmosphere of a volcanic ash and SO<sub>2</sub> plume that rose for a mean altitude of 10  
 399 km asl. Each eruptive episode was accompanied by the dispersal of ash plumes and fall-out  
 400 deposits in the surrounding towns of the volcano up to Catania, which often pose severe hazards  
 401 to aviation and repeated temporary closures of the Catania international airport. Lava flows  
 402 propagated mostly in the eastern flank of the volcano in the Valle del Bove and remained confined  
 403 at a mean altitude of 2,700 m asl. In the present study we focus on the specific phase, during this  
 404 broad activity, occurring between the 21 and 26/02/2021. Three main short-lived paroxysmal  
 405 events, discussed in Sec. 4.1, occurred during this time period.

## 406 **4 Results**

### 407 4.1 Three paroxysms in four days: the SO<sub>2</sub> emissions

408 The method described in Sect. 2.1 was applied to the SEVIRI observations of 21-26/02/2021 to  
 409 estimate the SO<sub>2</sub> emission rate and altitude of plume injection for the three individual paroxysms  
 410 in this timeframe. In Fig. 1, the temporal profile of the emission rate and altitude, for the Events  
 411 #1, 2 and 3 (whose dispersion is subsequently associated to the three plumes referred to as plumes  
 412 P1, P2 and P3 in Sect. 4.2) are shown. The SO<sub>2</sub> emissions associated to the Event #1 started at  
 413 about 23:00 of 20/02 and sharply increased during the first hours of the day 21/02. The emission  
 414 rate for this event peaked at values of about 2000-2500 kg s<sup>-1</sup> at 01:00 to 02:00 of 21/02. During

415 this event, the injection altitude is also sharply increasing from the crater altitude to about 12 km,  
 416 peaking between 01:00 and 02:00 of 21/02. The SO<sub>2</sub> emissions for the Event #1 sharply decreased  
 417 afterwards and the paroxysm was over by about 04:00 of 21/02. The Event #2 started at about the  
 418 same time but one day later than Event #1. The Event #2 had a sensibly shorter duration than Event  
 419 #1 - it started at about 23:00 of 21/02 and was over by 01:00 of 22/02 - but its peak emission rate  
 420 is nearly twice as large than for the Event #1, peaking at about 5000 kg s<sup>-1</sup> on 22/02 00:00. The  
 421 injection altitude for the Event #2 peaks at 12 km, in a similar fashion as the Event #1. At about  
 422 19:00 of 24/02, the Event #3 kicked off. The temporal profile and duration of the SO<sub>2</sub> emission  
 423 rate and injection altitude for the Event #3 is similar to those of the Event #1, with a slightly smaller  
 424 peak value of the emission rate (1500-2000 between 20:00 and 22:00 of 24/02).  
 425 The total emitted SO<sub>2</sub> mass for the three events has been estimated at values of 19.3, 20.9 and 14.6  
 426 kt for the Event #1, 2 and 3, respectively. The Event #2 injected the largest amount of SO<sub>2</sub>, despite  
 427 the shortest duration for the ensemble of the three events, due to the significantly larger peak  
 428 emission rate. The total SO<sub>2</sub> mass emitted from the three added events is 55 kt. Typical values of  
 429 the emission rates for moderate-to-strong eruption of Mount Etna are generally less than  
 430 2000 kg s<sup>-1</sup> (Sellitto et al., 2016, Corradini et al., 2020). This specific activity is then characterised  
 431 by relatively brief but very intense paroxysmal events, if compared to more general behaviour of  
 432 Mount Etna. In addition, the altitude of the plume injection is particularly high. Documented  
 433 extremely high-altitude eruptions of Mount Etna rarely exceed 10-11 km altitude (e.g. Sellitto et  
 434 al., 2016, Corradini et al., 2020). Thus, this series of three paroxysms qualify as an extreme event  
 435 in terms of the injection altitude. The total SO<sub>2</sub> mass emitted during these few days of activity is  
 436 about 50% of the record-breaking sequence of eruptions of 24-31/02/2015, which was estimated,  
 437 with a similar method as the present paper, at about 100.0 kt overall (Corradini et al., 2020). Thus,  
 438 in terms of the SO<sub>2</sub> emission rates and injection altitude, as well as for its total SO<sub>2</sub> emitted mass,  
 439 the sequence of three events during 21-26/02/2021 has to be categorised as one in the higher  
 440 spectrum of eruption strengths of Mount Etna.



441  
 442 *Figure 1: Temporal profile of the emission rates (blue lines) and injection altitude (red lines) for the three paroxysmal events*  
 443 *(Event #1-3 in the figure) of Mount Etna in the period 21-26/02/2021. The total emitted SO<sub>2</sub> mass, for each of the three events, is*  
 444 *also reported in the individual panels.*

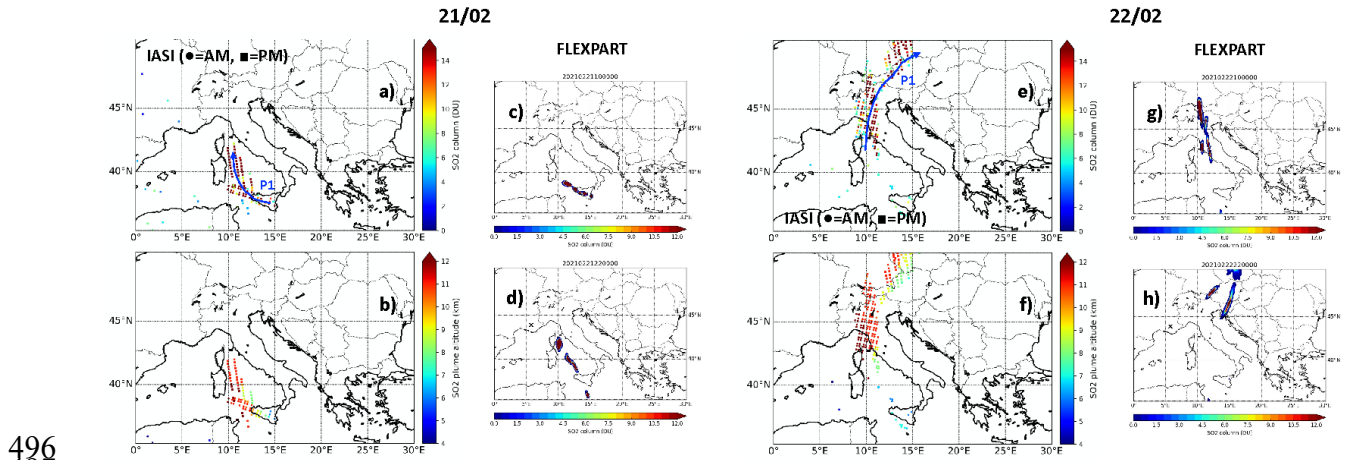
445 **4.2 An uncommon transport toward the north: the SO<sub>2</sub> plume dispersion**

446 The plumes emanating from the three individual paroxysmal events described in the previous  
 447 section are then characterised in terms of their subsequent dispersion.  
 448 Figures 2-4 show the SO<sub>2</sub> column amount and plume altitude obtained from IASI observations,  
 449 and the SO<sub>2</sub> column amount obtained from FLEXPART simulations, for 21-22 (Fig. 2), 23-24  
 450 (Fig. 3) and 25-26/02 (Fig. 4). The FLEXPART SO<sub>2</sub> simulations are obtained using the measured  
 451 SO<sub>2</sub> emissions described in Sect. 4.1. As extensively discussed by Sellitto et al. (2016), the  
 452 synergy of good quality regional observations from satellite and realistic simulations is crucial to  
 453 get more comprehensive insight into confined plumes dispersion, thanks to the information

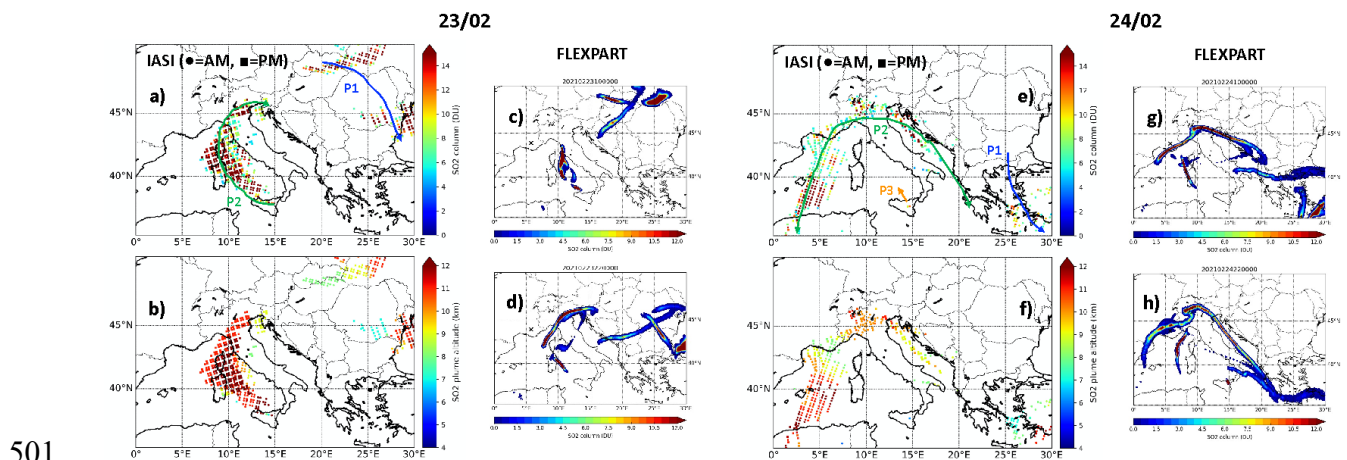
454 complementarity of these two information layers. In fact, the vertical sensitivity of satellite  
455 observations of SO<sub>2</sub> is limited, thus satellite observations can only provide a partial information  
456 on the height distribution of SO<sub>2</sub>; on the other hand, uncertainties in the input parameters and the  
457 physico-chemical in-plume processes can impact on the realism of dispersion simulations. In  
458 addition, the temporal sampling of satellite observations, one or two overpasses per day for low-  
459 Earth orbit spacecraft, is usually not sufficient to smoothly characterise the temporal evolution of  
460 the plume's dispersion. On the contrary, dispersion simulations can be realised at sub-hour  
461 temporal resolution, thus filling the temporal gap between satellite overpasses. An animation of  
462 the FLEXPART simulations for this case study, with the full temporal resolution of 30 minutes,  
463 can be found as Supplement. As it is possible to notice from Figs. 2-4, the description of the plume  
464 dispersion with observations and simulations is very consistent, thus cementing our confidence in  
465 our further interpretation of the plumes dynamics.

466 On 21/02, the plume P1, linked to the paroxysmal Event #1, disperses towards the central  
467 Tyrrhenian Sea, reaching Corsica to the western coast of Tuscany, for the night overpass of IASI  
468 (about 21:30 LT). During this phase, the plume stayed between 10-12 km altitude, with lower-  
469 altitude sections at the southern end of the plume. Values slightly higher than 15 DU are found for  
470 plume P1 during this phase, both from observations and modelling. The plume P1 then dispersed  
471 towards more north-eastern direction on the 22/02, to northern Italy and then Austria and further  
472 north-east, while progressively slightly descending at lower altitudes (down to 8-10 km). Clusters  
473 of relatively large values of the SO<sub>2</sub> column, around 15 DU, are found on 22/02. Plume P1  
474 orientates around an anticyclonic circulation due to the presence of a stable Omega block in Central  
475 Europe (Hoshyaripour, 2021), dispersing towards the south-eastern direction on 23/02 and then  
476 disperses further east, reaching Turkey and the Middle East on 24/02. The SO<sub>2</sub> column  
477 concentration for P1 remained relatively large, with values larger than 10 DU, while the plume  
478 kept descending, down to 7-10 km on 24/02. On 23/02, plume P2 (linked to the paroxysmal Event  
479 #2) kicked off with a similar initial dispersion as for P1, towards the central Tyrrhenian Sea, before  
480 starting to spread on 23/02 afternoon, from southern France to the Adriatic Sea. By 24/02, the  
481 plume P2 has formed an elongated and almost filamentary shape of quite large horizontal extent,  
482 from northern Spain, to the west, to southern Greece, to the east. Plume P2 was also much more  
483 irregular, in terms of both SO<sub>2</sub> column amount and plume altitude than plume P1. Values as high  
484 as 15 DU are found at the western and eastern ends of the plume, with mean altitudes at about 9 to  
485 12 km. Smaller column values, around 5-6 DU, and a mean altitude of about 10 km are found in  
486 the central area of the plume, in sub-alpine northern Italy. Plume P2 then compressed (i.e. the  
487 western and eastern ends of the plume got closer) on 25/02 and formed a curly plume then starting  
488 to disperse towards the north-east on 26/02. During this phase, the eastern end of plume P2 swept  
489 through southern Italy, still keeping at 8-9 km altitude. Plume P3 (linked to the paroxysmal Event  
490 #3) appears on 25/02 morning, again orientating towards the central Tyrrhenian Sea, while rapidly  
491 dispersing towards the east together with plume P2. Plume P3 displayed slightly larger values of  
492 the SO<sub>2</sub> column (larger than 15-18 DU) and lower altitude (7-10 km) than plumes P1 and P2. This  
493 overall description of the dispersion of the plumes P1-3 in the upper-troposphere (UT) is confirmed

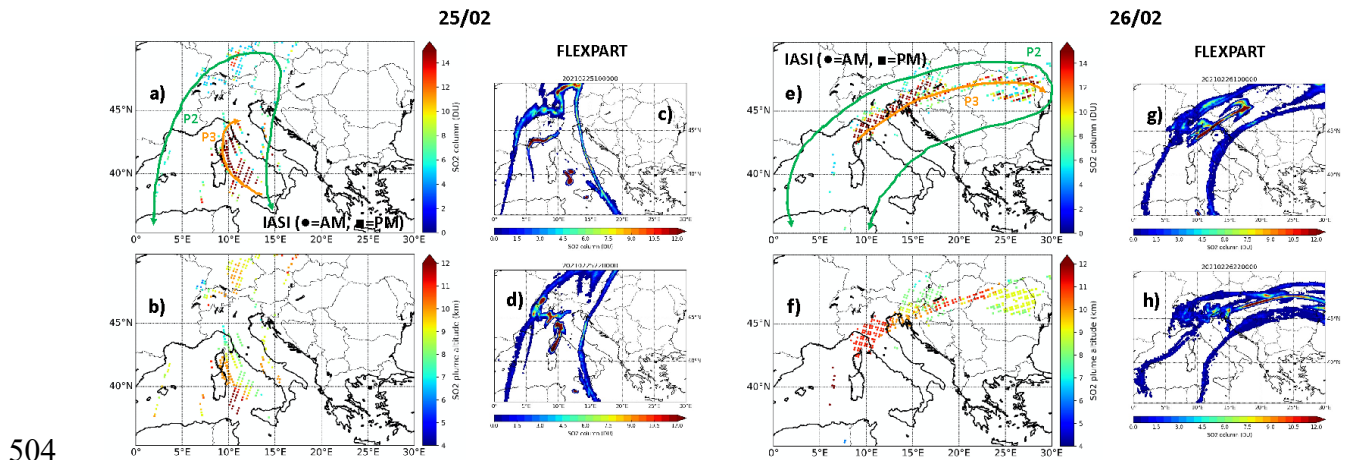
494 by the independent observations of the upper-tropospheric SO<sub>2</sub> column from OMPS-NM  
 495 (Supplementary Fig. 1).



496  
 497  
 498  
 499  
 500  
 Figure 2: IASI SO<sub>2</sub> column (a,e) and mean altitude of the plume (b,f) for the morning (approximate overpass at ~9:30 LT, circles) and afternoon (approximate overpass at ~21:30 LT, squares) MetOp-A spacecraft overpass; corresponding FLEXPART SO<sub>2</sub> column simulations (at 10:00 LT, c,g, and at 22:00 LT, d,h), for 21 and 22/02/2021. Plume P1 is individuated, and its approximate direction of dispersion is indicated as a blue arrow in panels a and e.



501  
 502  
 503  
 Figure 3: same as Figure 2 but for 23 and 24/02/2021. Plumes P1, P2 and P3 are individuated, and their approximate directions of dispersion are indicated as blue, green and orange arrows, respectively, in panels a and e.



504  
 505  
 506  
 Figure 4: same as Figure 2 but for 25 and 26/02/2021. Plumes P2 and P3 are individuated, and their approximate directions of dispersion are indicated as green and orange arrows, respectively, in panels a and e.

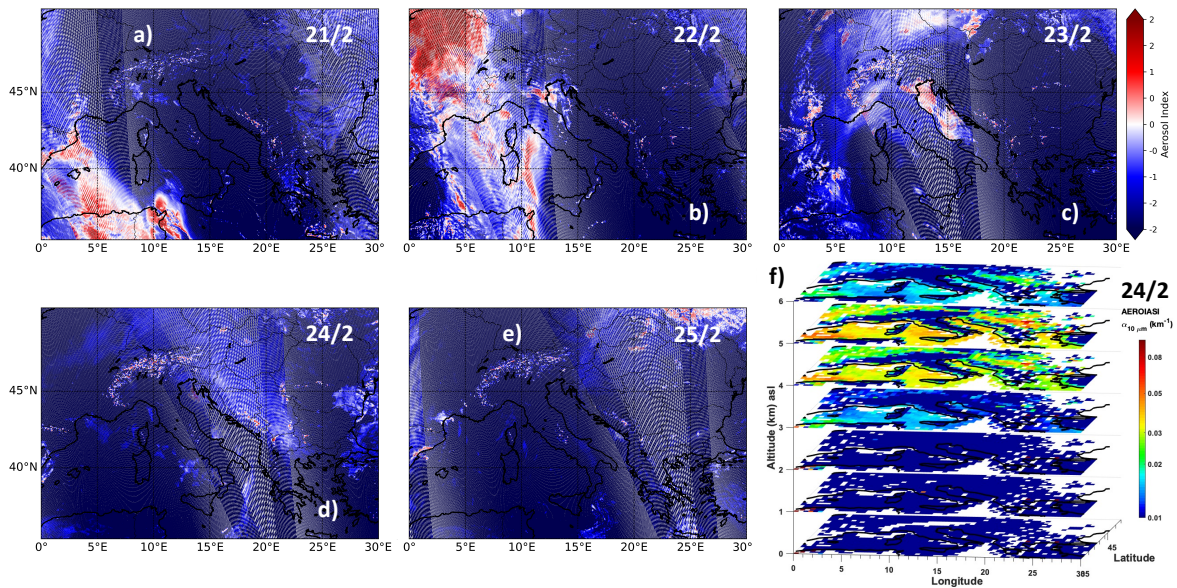
507 More in general, the dispersion of plumes P1-3 was consistently directed towards the northern  
508 quadrant from the perspective of the Mount Etna volcanic source. Using a more-than-one-decade  
509 long dispersion simulations dataset, Sellitto et al. (2017) have estimated the prevalent direction of  
510 dispersion of the plumes generated from Mount Etna emissions. They have found that for more  
511 than 80% of the time Mount Etna's plumes disperse towards the eastern quadrant. The dispersion  
512 towards the northern and north-western quadrant, as is the case for the initial dispersion for these  
513 events, is a rare occurrence, accounting only for less than 5% of the cases. On the other hand, the  
514 northern and north-western sectors are the only downwind areas that assure the presence of  
515 continental long-term active observatories that can allow the downwind observation and  
516 characterisation of Mount Etna's plumes. Thus, this series of paroxysms is a rare occasion to study  
517 more in details the dispersed plume of Mount Etna's volcanic emissions.

#### 518 4.3 Not only volcanic aerosols: the dust events of February 2021

519 During the same period of the series of volcanic events discussed in this manuscript, a major  
520 Saharan dust outbreak to central Europe occurred (Hoshyariapour 2021). Starting from 21/02, a  
521 trailing cold front produced a sustained dust uptake in northern Morocco and Algeria; the dust was  
522 then transported towards western Europe by southerly flow, was lifted by warm conveyor belt, and  
523 entered the mentioned Omega block located in central Europe, thus orienting in an anticyclonic  
524 circulation similar to volcanic plume P1. More specific analysis of this dust event is beyond the  
525 scopes of the present paper. Nevertheless, here we characterise the bulk dust distribution during  
526 this event, so to help the interpretation of the volcanic plumes at downwind observatories of Sect.  
527 4.4.

528 Figures 5a-e display the UVAI observations from TROPOMI during this period. They show the  
529 dust emitted from north-western Sahara towards the Western Mediterranean and North-Eastern  
530 Spain on 21/02, thus reaching Central and Northern France on 22/02 and then circulating around  
531 the Omega block high pressure during the following days, with weaker and weaker UVAI  
532 signature, possibly do to dust removal and dispersion. The vertical distribution of Saharan dust for  
533 this event is further investigated using the height-resolved information of the AEROIASI-Dust  
534 algorithm. In Fig. 5f, the dust-specific aerosol extinction profiles at 10  $\mu\text{m}$ , from AEROIASI-Dust,

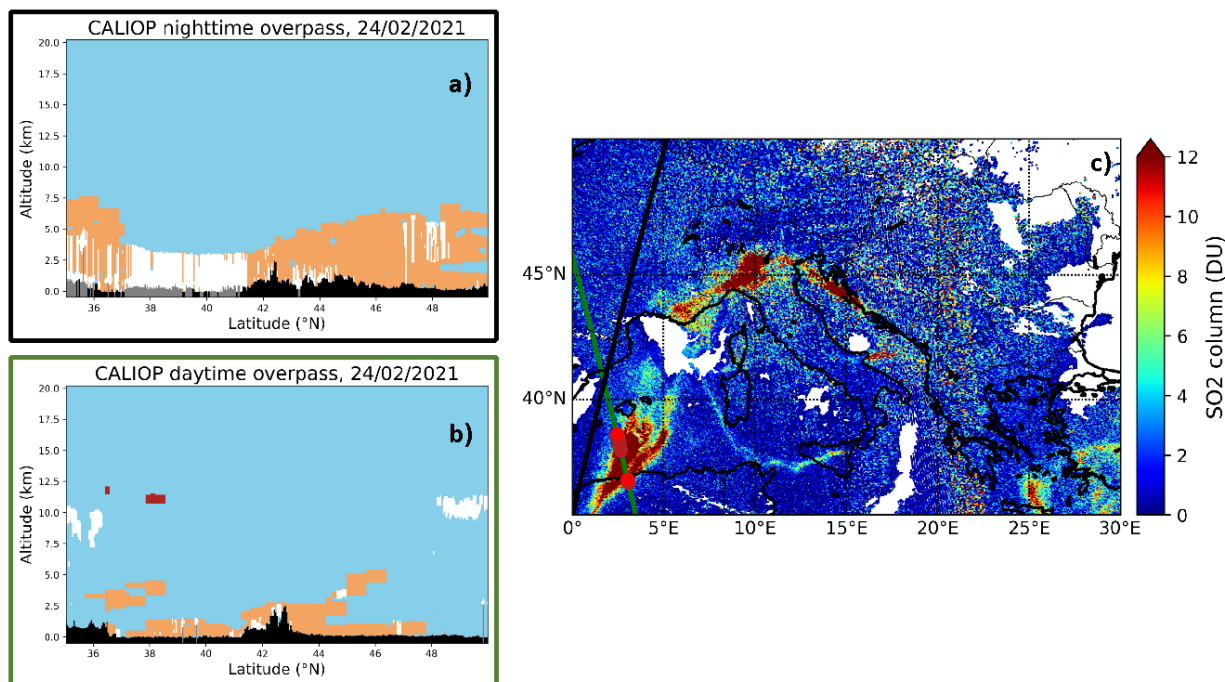
535 is shown for the 24/02. The dust looks confined, in the region of interest of our study, between 3  
 536 and 6 km altitude.



537  
 538 *Figure 5: UVAI retrievals with S5p TROPOMI for the period 21-25/02 (a,e). Dust-specific aerosol extinction at 10 μm from*  
 539 *AEROSI-Dust observations for 24/02, from surface to 6 km altitudes (f).*

540 Figure 6a,b show the CALIOP feature layer classification for CALIOP day and night overpasses  
 541 in the area of interest (see ground tracks of CALIOP in Fig. 6c), on 24/02. Both day and night  
 542 overpasses confirm the presence of dust from near-surface to 5-7 km altitude. The lifting of the  
 543 dust layer in the south-north direction, from North Africa to Central Europe, is clearly visible.  
 544 Figure 6b also shows evidence of the volcanic plume from Mount Etna plume P2. As discussed  
 545 more in details in Sect. 4.4, the plume P2 arrives in Southern France and Spain coastal areas on  
 546 24/02 afternoon, which is consistent with the two CALIOP overpasses of Fig. 6a,b. An early  
 547 observation of the plume P2 on 23/02, in the central Tyrrhenian Sea, together with the underlying  
 548 dust layer, is shown in the Supplementary Fig. 2. The volcanic aerosol layer is reasonably co-  
 549 located with the volcanic SO<sub>2</sub> plume, observed with TROPOMI, for both overpasses (23/02,  
 550 Supplementary Fig. 2b, and 24/02, Fig. 6c). The volcanic plume P2 observed by CALIOP, is  
 551 located at exceptionally high altitudes for Mount Etna's activity, at 12-13 km on the 23/02 and  
 552 then slightly descending to 11-12 km on 24/02. The vertical plume location is very consistent with  
 553 IASI observations and FLEXPART simulation of the SO<sub>2</sub> plume discussed in Sect. 4.2. The  
 554 volcanic and the dust plumes remain completely vertically-separated in this area.





555  
 556 *Figure 6: (a,b) Feature layer classification (sky blue: clear sky, black: Earth's surface, white: clouds, brown: tropospheric*  
 557 *aerosol – here classified as dust, red: stratospheric aerosol feature – here classified as volcanic, grey: no signal) for CALIOP*  
 558 *nighttime, at ~2:00 am (a) and daytime, at ~1:30 pm (b), overpasses for 24/02/2021. (c) Sentinel-5p TROPOMI SO2 column*  
 559 *observations for 24/02/2021. The black and green lines in panel c are the ground tracks of CALIOP observations of panel a*  
 560 *and b, respectively. The position of the stratospheric aerosol (volcanic) feature, identified in panel b, is also indicated in panel c*  
 561 *as red circles.*

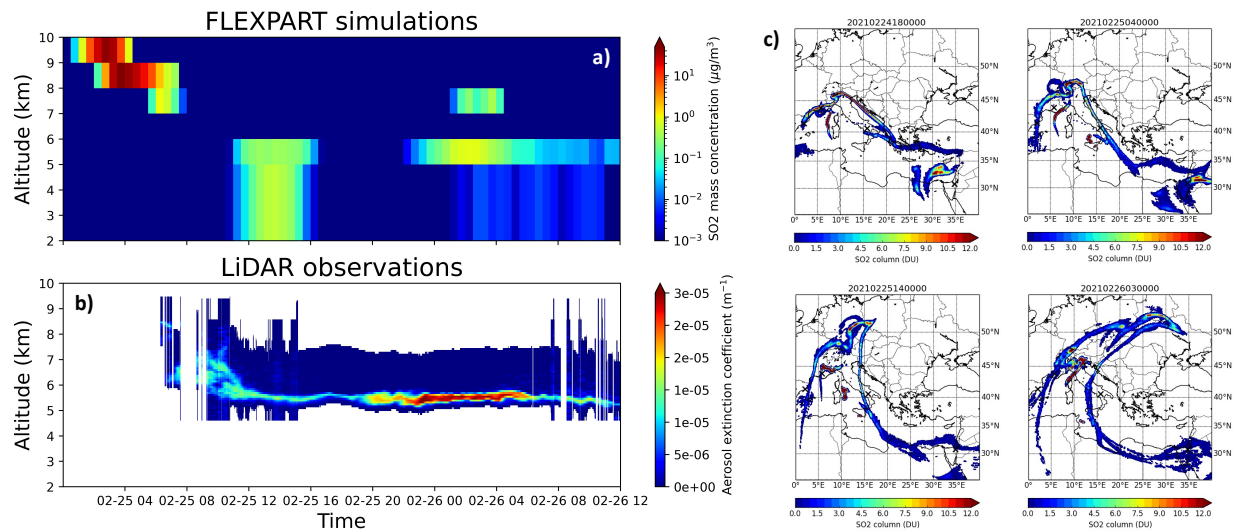
#### 562 4.4 The volcanic plumes themselves: observations at downwind observatories

563 The uncommon transport of Mount Etna's plumes towards the north, for these three events, is a  
 564 rare occasion to monitor and characterise the volcanic aerosols in Mount Etna's plumes at well-  
 565 equipped observatories, including vertical information with LiDAR systems. As mentioned above,  
 566 in the large majority of cases, Mount Etna's plumes disperse towards the east, in a mainly marine  
 567 region where very few ground observation stations are present. On the contrary, there is significant  
 568 density of ground stations northwards, i.e. in continental Europe.

569 Plumes P1 and P2 have been observed downwind at different stations. Here in the following, we  
 570 analyse the plume's observations at three stations: Sede Boker, Israel, OHP, France, and Napoli,  
 571 Italy.

572 Figure 7a shows the volcanic plume vertical distribution over Sede Boker station, in terms of the  
 573 SO2 volcanic tracer, from FLEXPART simulations. Figure 7b shows the series of LiDAR aerosol  
 574 observations at Sede Boker, for the same period. The plume P1, after a relatively long dispersion  
 575 phase over central-easter Europe, driven by anticyclonic flow of the Omega block discussed in  
 576 Sect. 4.2 (see Fig. 2 and 3, as well as the animation in the Supplementary material), overpasses the  
 577 Sede Boker station starting from 25/02 early in the morning and, with discontinuous broken  
 578 dispersed plumes sections, until 26/02 in the early afternoon, as visible from Fig. 7c. More dense  
 579 clusters of the plume P1 overpass the station during the period spanning from 25/02 night to 26/02  
 580 early morning. During this plume's overpass, P1 has aged 4-5 days since their volcanic emissions.  
 581 The LiDAR observations show an aerosol layer linked to the plume P1 in the same period indicated  
 582 by the FLEXPART simulations. Even if a part of the plume observation is missed by the LiDAR,

583 during the night between 24 and 25/02 (due to low LiDAR signal quality), a consistent vertical  
 584 behaviour is shown by simulations and observations. Both information layers show an initially  
 585 higher plume, located at 9-10 km, then descending to 5-6 km altitude. The plume looks  
 586 filamentary, with a geometrical vertical thickness of less than 500 meters (Figs. 7a and b) and a  
 587 horizontally-confined shape (Fig. 7c). The mean aerosol depolarization for the plume is  $32.3 \pm 1.4\%$   
 588 (Tab. 1). This value is very elevated for volcanic aerosols and might indicate the presence of a  
 589 large fraction of fine ash, for plume P1. The three-dimensional morphology of this aerosol layer,  
 590 as detected by LiDAR, as well as the consistency of observations with volcanic tracer dispersion  
 591 simulations, suggest that a significant presence of dust can be excluded at these altitudes.

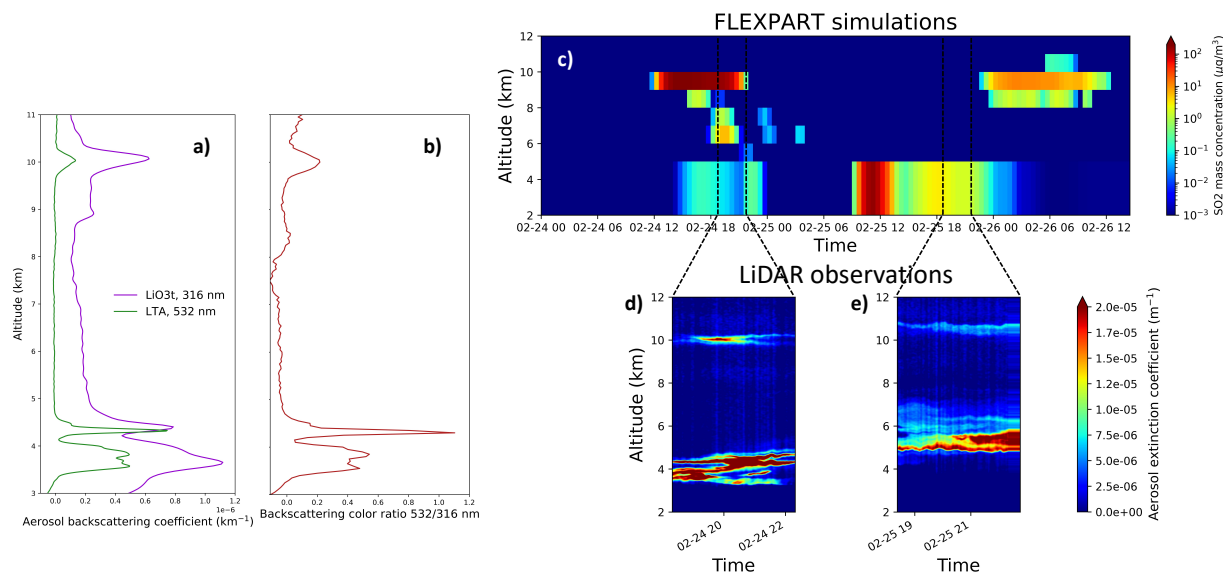


592  
 593 *Figure 7: FLEXPART SO<sub>2</sub> mass concentration profiles, from 25/02/2021 00:00 to 26/02/2021 12:00, over Sede Boker station (a).*  
 594 *Volcanic-aerosol-specific LiDAR observations of the aerosol extinction coefficient, for the same period as in panel a, at Sede Boker*  
 595 *station (b). Selected maps of the SO<sub>2</sub> total column dispersion, between 24/02 and 26/02/2021, from FLEXPART simulations (c).*  
 596 *The position of the Sede Boker station is shown as a black cross in panel c.*

597 The western and eastern peripheries of the dispersed plume P2 (hereafter referred to as P2/W and  
 598 P2/E, respectively) are almost synoptically observed at OHP (P2/W) and Napoli (PE/E) ground  
 599 stations. Starting from 24/02, the plume P2 takes an elongated and then folded-shaped geometry,  
 600 with the western periphery P2/W slowly moving south-westwards over Southern France and  
 601 Eastern Spain, and the eastern periphery P2/E sweeping Southern Italy in a north-western direction  
 602 (Fig. 3-4).

603 Basing on FLEXPART simulations, the P2/W section arrives at OHP station basically in two  
 604 separate phases: during evening/night of 24/02 and during night/early morning between 25 and  
 605 26/02 (Fig. 8c). The FLEXPART simulations over OHP show a denser volcanic plume, in terms  
 606 of the SO<sub>2</sub> tracer, at altitudes of about 9-10 km altitude, with less dense volcanic plume at lower  
 607 altitudes (2-4 km). This is generally consistent with the observations of the LTA LiDAR system  
 608 at OHP (Figs. 8d-e). The LTA LiDAR was operational from about 18:30 to 22:30 on 24/04 and  
 609 25/02; it observes an aerosol layer at about 10-11 km altitude on both days and during the full  
 610 period, with a maximum value of the aerosol extinction between 19:00 and 21:00 on 24/02,  
 611 consistently with FLEXPART simulations. The very weak volcanic aerosol signature at 10-11 km  
 612 for LTA LiDAR observations 25/02 is not present in FLEXPART simulations. A previous  
 613 observations session was carried out during the late afternoon/evening of 23/02 (not shown here);  
 614 a volcanic-related aerosol plume is not observed during this period, consistently with FLEXPART

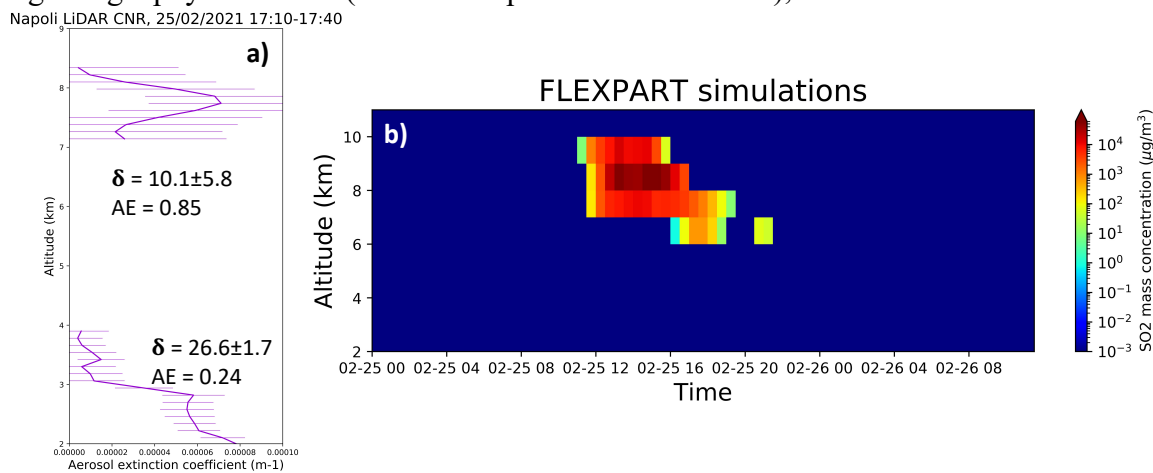
615 time-series analyses of the plume P2/W dispersion. While the LTA LiDAR system was operational  
 616 only during night-time, the co-located GAIA system observed the atmosphere over OHP during  
 617 day-time as well (data not shown here), even if with a significantly lesser signal/noise ratio at those  
 618 altitudes. While GAIA observes the same night-time volcanic aerosol signature as LTA (discussed  
 619 above), no aerosol signature is present during the days of 24 and 25/02, so it is reasonable to  
 620 exclude that volcanic aerosols were present in the atmosphere over OHP during the day. Besides  
 621 the relatively high-altitude aerosol layer observations, the LTA LiDAR observed a lower-altitude  
 622 aerosol plume at 3-5 km (see Fig. 8a). This aerosol signal can be easily associated with the dust  
 623 event described in Sect. 4.3, even if a component of volcanic aerosol cannot be excluded based on  
 624 the FLEXPART vertical profiles of Fig. 8c. For a limited time, the LTA and the LiO3t LiDARs  
 625 operated simultaneously at OHP. Using a combination of the aerosol backscattering profiles  
 626 retrieved by LTA (at 532 nm) and the LiO3t (at 316 nm) LiDARs at OHP, a colour ratio has been  
 627 calculated in the free troposphere on 24/02 between about 19:00 and 20:30. The aerosol  
 628 backscattering profile observations at both wavelengths show a vertically thin plume at about 10-  
 629 11 km and a wider plume between 3 and 5 km. Besides this quite distinct vertical structure, the  
 630 colour ratio shows very different values for the two layers, with 0.1-0.2 for the highest first layer  
 631 and up to 1.0-1.2 for the lowest layer. In addition, depolarisation ratio observations with the GAIA  
 632 system for the highest layer display very small, near-zero values. These evidences indicate the  
 633 presence of distinctly smaller mean particle size, with a possible spherical shape, for the highest  
 634 layer with respect to the lowest, thus corroborating the possibility that volcanic aerosols, with an  
 635 SA prevalence, and dust, respectively, dominate the two layers. Spectroscopic evidence of the  
 636 presence of SA at OHP is found using the AEROIASI-H2SO4 retrieval algorithm (Supplementary  
 637 Fig. 3), this suggesting that the volcanic plume P2/W is composed of SA, even if the presence of  
 638 an ash fraction cannot be excluded.



639  
 640 *Figure 8: Mean aerosol backscattering coefficient profiles, between 19:00 and 20:30 of the day 24/02/2021, observed by LiO3t*  
 641 *(violet line) and LTA (green line) LiDARs (a), and the backscattering colour ratio (b), at OHP station. FLEXPART SO2 mass*  
 642 *concentration profiles, from 24/02/2021 00:00 to 26/02/2021 12:00, over OHP station (c) and available night-time LTA*  
 643 *observations of the aerosol extinction coefficient in the same period (d: 24/02; e: 25/02). Time intervals of LTA observations are*  
 644 *indicated by black dashed lines in panel c.*

645 The P2/E plume section is observed at Napoli station. Based on the FLEXPART simulations, the  
 646 P2/E volcanic plume overpasses Napoli during the period spanning from late morning to early

647 evening of 25/02, at an altitude of 7-10 km, with a maximum density between 15:00 to 18:00 (Fig.  
 648 9b). Consistently, the LiDAR system in Napoli observed a vertically-confined aerosol plume in  
 649 the afternoon of 25/02, with a maximum around 17:30, at 7-8 km (Fig. 9a). A second aerosol  
 650 plume, with a wider vertical structure, is observed between 2 and 3 km. The LiDAR system in  
 651 Napoli operates at two wavelengths and has depolarisation information (depolarization ratio  $\delta$ ).  
 652 Using the two wavelengths, the bulk Ångström exponent (AE) has been calculated. Both AE and  
 653  $\delta$  show distinct values for the two layers: AE of 0.85 and 0.24, and  $\delta$  of  $10.1 \pm 5.8\%$  and  $26.6 \pm 1.7\%$ ,  
 654 for the higher and lower layer, respectively. This indicates significantly smaller (AE observations)  
 655 more spherical-shaped ( $\delta$ ) for the higher layer, thus corroborating the identification of the highest  
 656 layer as volcanic and the lowest as dust. The AE and  $\delta$  for the highest layer are nevertheless quite  
 657 different from what expected for a pure SA-dominated layer, indicating the possibility of the  
 658 presence of a fraction of larger and aspherical ash particles. It is also important to notice that: 1)  
 659 the SO<sub>2</sub> plume tracer of P2/E in Napoli (Fig. 9b) has peak values nearly two orders of magnitude  
 660 larger than for P2/W at OHP (Fig. 8c), and 2) the aerosol extinction is almost ten times larger for  
 661 P2/E (Fig. 9a) than P2/W (Fig. 8d). This clearly indicates the large variability of the morphology  
 662 and properties of one volcanic plume, e.g. when looking at different plume's sections.  
 663 Based on the results shown in this section, two main evidences emerge: 1) precise (e.g. in terms  
 664 of the input parameters) and detailed dispersion simulations, as well as complementary satellite  
 665 observations at a wider regional scale, as support towards the interpretation of downwind  
 666 observations, and 2) synoptic downwind observations at different locations, can be used to  
 667 characterise in details confined plumes of known source and their variability in three-dimensional  
 668 morphology, composition and properties. In the present case, for example, two separate and non-  
 669 mixing vertical layers of volcanic aerosols and dust could be identified. For the volcanic aerosol  
 670 layer, the variability of the vertical structure and composition at different sections of the plumes  
 671 (see the case of plumes P2/W and P2/E), as well as the different properties of individual plumes  
 672 for a given geophysical event (see the comparison of P1 and P2), could be characterised.



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 675  
 676  
 677  
 Figure 9: Mean aerosol extinction coefficient profiles at 355 nm, between 17:10 and 17:40 of the day 25/02/2021, observed by LiDAR (a) and FLEXPART SO<sub>2</sub> mass concentration profiles, from 25/02/2021 00:00 to 26/02/2021 12:00, over Napoli station (b). The mean Ångström exponent (AE) and depolarisation ratio ( $\delta$ ), for the two identified vertically-separated aerosol layers are also reported in panel a.

## 678 4.5 Optical properties and the impact on the radiative balance

679 Using the observations from ground-based LiDAR and the complementary observations and  
680 atmospheric modelling of the previous sections, the optical and radiative properties of plumes P1  
681 and P2/E and W, at Sede Boker, OHP and Napoli stations, have been estimated.

682 The volcanic-specific aerosol optical depth ( $AOD_V$ ), averaged during the core overpass periods  
683 (see details in the caption of Tab. 1) at the three ground stations, estimated using LiDAR data, are  
684 listed in Tab. 1. These values are also compared with the total column (i.e. volcanic plus dust and  
685 other possible aerosols in the line of sight of the instrument)  $AOD_T$ ,  $AE_T$  and  $SSA_T$  estimated with  
686 co-located AERONET Cimel sun photometers. For plume P1, moderate  $AOD_T$  are found, with  
687 relatively high  $AE_T$ , which points at a limited impact of dust in the overall column. The  $AOD_V$  is  
688 relatively large,  $0.0076 \pm 0.0015$ . As mentioned, the large  $\delta_V$ , aggregated with a relatively large  
689  $AOD_V$ , may point at a consistent presence of ash within the identified volcanic plume P1. This is  
690 somewhat surprising for a plume that, at the time of downwind observation, has already aged about  
691 4-5 days since emissions. It is worth noticing that this plume shows a consistent descent during  
692 the dispersion (emitted at about 12 km and detected at Sede Boker at 5 to 10 km) which can be  
693 linked to the larger sedimentation rate of the more massive ash particles than SA. As already  
694 discussed in the previous section, plume P2 looks very inhomogeneous, when comparing the  
695 western and eastern ends P2/W and P2/E. This is mirrored by inhomogeneities in the measured  
696  $AOD_T$ . The P2/W plume has almost ten times smaller  $AOD_T$  with respect to P2/E (P2/W:  
697  $0.0036 \pm 0.0014$ , P2/E:  $0.031 \pm 0.006$ ). As discussed in Sect. 4.4, this large difference in the  $AOD_T$   
698 of P2/W and E can be attributed to the fact that these plumes may be largely dominated by SA, the  
699 first, and may present a fraction of ash, the second. With  $AOD_T$  between 0.25 and 0.30, the total  
700 column looks largely dominated by dust, with the volcanic component taking only a small part of  
701 the column. Relatively small  $AE_T$  seems to confirm this hypothesis through the indication of the  
702 dominance of very large (dust) particles.

703 Using the libRadtran-UVSPEc radiative transfer model (RTM), driven by aerosol extinction  
704 profiles measured by LiDARs at the three stations, the daily average radiative forcing (RF) of the  
705 isolated volcanic plumes has been estimated with the methodology described in Sect. 2.2.2, and is  
706 reported in Tab. 2. The aerosol extinction profiles used as input to the RTM are the average of the  
707 observations at the core overpass time, i.e. the same period used to calculate the  $AOD_V$  in Tab.1  
708 and detailed in its caption. Based on the considerations above and the complementary observations,  
709 we have made the following hypotheses for non-measured optical parameters of the plumes. For  
710 plume P1 at Sede Boker, a fraction of fine ash was probably present in the volcanic plume. Thus,  
711 we have run the RTM using an interval of AE typical of small-sized particles (1.8-2.0) and a SSA  
712 typical of partially absorbing particles (0.90-0.95). A moderately dusty background lower-  
713 tropospheric atmosphere is considered for altitudes between surface to 3 km. For plume P2/W at  
714 OHP, a SA-dominated plume is considered. Thus, we have run the RTM using an interval of AE  
715 typical of small-sized particles (1.8-2.0) and a SSA typical of very reflective aerosols (0.95-1.00).  
716 A very dusty background lower-tropospheric atmosphere is considered for altitudes between  
717 surface to 5 km, based on LiDAR observations of the lowest layer of e.g. Fig. 8a-b. For plume  
718 P2/E at Napoli, a fraction of ash in the volcanic plume is considered. Thus, we have run the RTM  
719 using the observed AE (0.85), which is typical of larger particles than what found for P1 and P2/W,  
720 and SSA typical of partly absorbing aerosols (0.90-0.95). As for plume P2/W, a very dusty  
721 background lower-tropospheric atmosphere is considered for altitudes between surface to 5 km,  
722 based on LiDAR observations of the lowest layer of e.g. Fig. 9a. With these assumptions, a TOA  
723 RF of  $-0.22 \pm 0.06$ ,  $-0.17 \pm 0.03$  and  $-1.19 \pm 0.29$  W/m<sup>2</sup>, and a surface RF of  $-0.66 \pm 0.09$ ,  $-0.21 \pm 0.04$

724 and  $-3.51 \pm 0.47$  W/m<sup>2</sup> are found for P1, P2/W and P2/E, respectively. Associated with a larger  
 725 AOD<sub>v</sub>, the RF of P2/E is significantly larger than for P1 and P2/W. The surface RF is much larger  
 726 than the TOA RF, for both P1 and P2/E, due to the presence of absorbing aerosols in these plumes.  
 727 This means that part of the incoming shortwave radiation does not reach the surface due to the in-  
 728 plume absorption, which adds to the radiation scattered back to space. For plume P2/W, the TOA  
 729 and surface RF have a very similar value, which points at a very limited absorption within the  
 730 plume due to the likely dominant presence of purely reflective SA. The RF values obtained for  
 731 these plumes can be compared with RFs estimated in the past for proximal and distal observations  
 732 of Mount Etna's emissions. Using ground LiDAR observations as input to the RTM, Sellitto et al.  
 733 (2020) have estimated the RF of a typical passive degassing plume at a proximal location, about 7  
 734 km downwind Mount Etna craters and have found values of about  $-4.5$  and  $-7.0$  W/m<sup>2</sup>, at TOA  
 735 and surface. Mild explosions and a small fraction of ash were present during these observations.  
 736 For an almost purely SA, after a dispersion of about 350 km downwind Mount Etna, Sellitto et al.  
 737 (2016) have found a RF efficiency, i.e. the RF per AOD unit, of about  $-40$  to  $-50$  W/m<sup>2</sup>/AOD. The  
 738 RF efficiency of P2/E (about 35 W/m<sup>2</sup>/AOD) and P1 (about 29 W/m<sup>2</sup>/AOD) are consistent with  
 739 estimations of Sellitto et al. (2020) for an ashy passive degassing plume (about 35 W/m<sup>2</sup>/AOD).  
 740 The RF efficiency of P2/W (about 47 W/m<sup>2</sup>/AOD) is consistent with estimations of Sellitto et al.  
 741 (2016) for purely SA plume. Compared with larger volcanic eruptions, our estimates are e.g. of  
 742 the same order of magnitude or even larger than the impact of the Raikoke eruption in 2019 ( $-0.3$   
 743 to  $-0.4$  W/m<sup>2</sup>) (Kloss et al., 2021). The Raikoke had the largest volcanic impact on the radiative  
 744 balance documented in at least the last 3 years. Of course, Raikoke eruption is much more relevant  
 745 than the activity of Mount Etna discussed in the present study, due to its larger scale (hemispheric)  
 746 impact. This demonstrates that, even if Mount Etna eruptions are of a limited regional interest,  
 747 they can have a quite relevant impact at the regional scale.

748  
 749 *Table 1: Summary of optical properties of volcanic plumes P1 and P2, volcanic-specific layers (V index in the table) from LiDAR*  
 750 *observations and total-column values (T index in the table) from AERONET sun-photometric observations. \*=mean value of*  
 751 *LiDAR observations between 25/02 20:00 and 26/02 06:00; \*\*=mean value of sun-photometric observations of 25 and 26/02;*  
 752 *\*\*\*=mean value of sun-photometric observations of 26/02 between 05:00 and 06:00; §=mean value of LTA LiDAR observations*

753 of 24/02 between about 19:00 and 21:30; <sup>§§</sup>=mean value of sun-photometric observations of 24/02; += mean value of LiDAR  
 754 observations of 25/02 between 17:10 and 17:40 (as in Fig. 9); <sup>++</sup>=mean value of sun-photometric observations of 25/02

	AOD <sub>V</sub> (532 nm)	δ <sub>V</sub> (532 nm)	AE <sub>V</sub>	AOD <sub>T</sub> (532 nm)	AE <sub>T</sub>	SSA <sub>T</sub>
P1: Sede Boker	0.0076±0.0015* (maximum: 0.035)	32.3±1.4%*	-	0.099±0.004** 0.116±0.005***	1.16±0.01***	0.88±0.01***
P2/W: OHP observatory	0.0036±0.0014 <sup>§</sup> (maximum: 0.0062)	-	-	0.26±0.06 <sup>§§</sup>	0.21±0.10 <sup>§§</sup>	0.96±0.03 <sup>§§</sup>
P2/E: Napoli Observatory	0.031±0.006 <sup>+</sup>	10.1±5.8% <sup>+</sup>	0.85 ± 0.60 <sup>+</sup>	0.310±0.070 <sup>++</sup>	0.92±0.12 <sup>++</sup>	0.94±0.02 <sup>++</sup>

755

756 Table 2: Summary of radiative forcing estimations of volcanic plumes P1 and P2, see text for the radiative transfer modelling  
 757 setup and hypotheses

	TOA radiative forcing (W/m <sup>2</sup> )	Surface radiative forcing (W/m <sup>2</sup> )
P1: Sede Boker	-0.22±0.06	-0.66±0.09
P2/W: OHP observatory	-0.17±0.03	-0.21±0.04
P2/E: Napoli Observatory	-1.19±0.29	-3.51±0.47

758

## 759 5 Conclusions

760 The synergy of volcanic plumes observation and modelling at different spatio-temporal scales is  
 761 crucial to characterize their emission, dispersion and downwind impacts. Mount Etna experiences  
 762 an intense eruptive activity between February and April 2021. Within this phase, three peculiar  
 763 extreme events, in terms of the large SO<sub>2</sub> emission rates (55 kt of SO<sub>2</sub> emitted overall) and  
 764 injection altitude (up to 12 km), took place between 21 and 26/02, that we have studied with a  
 765 combination of regional-scale satellite observations and Lagrangian dispersion modelling, and  
 766 local downwind measurement at selected ground stations coupled with offline RTM modelling.  
 767 These three events and the subsequently formed plumes of volcanic effluents (plumes P1-3)  
 768 displayed a very uncommon dispersion towards the north, which allowed the downwind  
 769 observation and characterisation of the plumes due to the presence of several observatory sites in  
 770 this quadrant. Ground-based LiDAR observations and complementary information from  
 771 observations and modelling at the sites of Sede Boker, Israel (plume P1), OHP (western end of  
 772 plume P2 – P2/W) and Napoli (eastern end of plume P2 – P2/E) revealed a complex inter- (P1  
 773 versus P2) and intra-plume (P2/W versus P2/E) variability of the morphology, composition, optical  
 774 properties and radiative impacts of Mount Etna plumes. Plume P1 has been detected at about 10  
 775 then descending to 5-6 km after only 4-5 days atmospheric aging, with depolarisation ratio  
 776 consistent with a possible presence of in-plume ash. Plume P2 displayed a dramatic variability  
 777 from east to west ends, with a thin and likely SA-dominated plume to the west and a dense and  
 778 likely ash-bearing plume to the east. Both the AOD (0.004 to 0.03) and local clear-sky daily

779 average shortwave radiative forcing (  $-0.2$  to  $-1.2$   $\text{W/m}^2$ , at TOA and  $-0.2$  to  $-3.5$   $\text{W/m}^2$  at the  
 780 surface) point at a very relevant impact on the upper-tropospheric aerosol layer and the regional  
 781 climate at the continental scale, in the Mediterranean. In addition, the synergy of observations and  
 782 modelling presented in the present work allowed to empirically disentangle the information about  
 783 volcanic aerosols and other aerosol sources, like mineral dust.

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#### 803 **References**

- 804 Ancellet G. and Beekmann M. (1997) Evidence for changes in the ozone concentrations in the  
 805 free troposphere over southern France from 1976 to 1995, *Atmospheric Environment*, 31, 17,  
 806 2835-2851, [https://doi.org/10.1016/S1352-2310\(97\)00032-0](https://doi.org/10.1016/S1352-2310(97)00032-0).
- 807 Andersson, S., Martinsson, B., Vernier, JP. et al. (2015) Significant radiative impact of volcanic  
 808 aerosol in the lowermost stratosphere. *Nat. Commun.* 6, 7692.  
 809 doi:<https://doi.org/10.1038/ncomms8692>
- 810 Ansmann, A., Riebesell, M., and Weitkamp, C. (1990) Measurement of atmospheric aerosol  
 811 extinction profiles with a Raman lidar. *Opt. Lett.*, 15, 746–748. doi:10.1364/OL.15.000746.
- 812 Biele, J., Beyerle, G., and Baumgarten, G. (2000) Polarization lidar: corrections of instrumental  
 813 effects. *Opt. Express*, 7, 427–435, doi:10.1364/OE.7.000427.
- 814 Biermann, U.M., Luo, B.P., Peter, T. (2000) Absorption Spectra and Optical Constants of Binary  
 815 and Ternary Solutions of H2SO4, HNO3, and H2O in the Mid Infrared at Atmospheric  
 816 Temperatures. *J. Phys. Chem. A*, 104, 783–793.
- 817 Bilal, M.; Nazeer, M.; Nichol, J.; Qiu, Z.; Wang, L.; Bleiweiss, M.P.; Shen, X.; Campbell, J.R.;  
 818 Lolli, S. Evaluation of Terra-MODIS C6 and C6.1 Aerosol Products against Beijing, XiangHe,  
 819 and Xinglong AERONET Sites in China during 2004-2014. *Remote Sens.* 2019, 11, 486.  
 820 <https://doi.org/10.3390/rs11050486>.



- 821 Boselli, A., D'Emilio, M., Sannino, A., Wang, X., and Amoruso, S. (2021) Remote sensing and  
 822 near surface observations of aerosol properties measured in real time during a big fire event on  
 823 Mount Vesuvius, *Remote Sensing*, 13(10), <https://doi.org/10.3390/rs13102001>.
- 824 Campbell, J.R. J.R., D.L. Hlavka, E.J. Welton, C.J. Flynn, D.D. Turner, J.D. Spinhirne, V.S.  
 825 Scott, and I.H. Hwang, 2002. Full-time, Eye-Safe Cloud and Aerosol Lidar Observation at  
 826 Atmospheric Radiation Measurement Program Sites: Instrument and Data Processing, *J. Atmos.*  
 827 *Oceanic Technol.*, 19, 431-442.
- 828 Campbell, J. R., Dolinar, E. K., Lolli, S., Fochesatto, G. J., Gu, Y., Lewis, J. R., ... & Welton, E.  
 829 J. (2021). Cirrus cloud top-of-the-atmosphere net daytime forcing in the Alaskan subarctic from  
 830 ground-based MPLNET monitoring. *Journal of Applied Meteorology and Climatology*, 60(1),  
 831 51-63.
- 832 Clarisse, L., Hurtmans, D., Clerbaux, C., Hadji-Lazaro, J., Ngadi, Y., and Coheur, P.-F. (2012)  
 833 Retrieval of sulphur dioxide from the infrared atmospheric sounding interferometer (IASI),  
 834 *Atmos. Meas. Tech.*, 5, 581–594, <https://doi.org/10.5194/amt-5-581-2012>.
- 835 Clarisse, L., Coheur, P.-F., Theys, N., Hurtmans, D., and Clerbaux, C. (2014) The 2011 Nabro  
 836 eruption, a SO<sub>2</sub> plume height analysis using IASI measurements, *Atmos. Chem. Phys.*, 14,  
 837 3095–3111, <https://doi.org/10.5194/acp-14-3095-2014>.
- 838 Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D.,  
 839 Pommier, M., Razavi, A., Turquety, S., et al. (2009) Monitoring of atmospheric composition  
 840 using the thermal infrared IASI/MetOp sounder. *Atmos. Chem. Phys.*, 9, 6041–6054.
- 841 Corradini S.; Guerrieri L.; Lombardo V.; Merucci L.; Musacchio M., Prestifilippo M.; Scollo S.;  
 842 Silvestri M.; Spata G.; Stelitano D. (2018) Proximal monitoring of the 2011-2015 Etna lava  
 843 fountains using MSG-SEVIRI data. *MDPI Geosciences*, 8, 140;  
 844 [doi:10.3390/geosciences8040140](https://doi.org/10.3390/geosciences8040140)
- 845 Corradini S., Guerrieri L., Stelitano D., Salerno G., Scollo S., Merucci L., Prestifilippo M.,  
 846 Musacchio M., Silvestri M., Lombardo V., Caltabiano T. (2020) Near Real-Time Monitoring of  
 847 the Christmas 2018 Etna Eruption Using SEVIRI and Products Validation, *Remote Sens.*, 12,  
 848 1336; [doi:10.3390/rs12081336](https://doi.org/10.3390/rs12081336).
- 849 Corradini S., Guerrieri L., Brenot H., Clarisse L., Merucci L., Pardini F., Prata A.J., Realmuto  
 850 V.J., Stelitano D., Theys N. (2021) Tropospheric Volcanic SO<sub>2</sub> Mass and Flux Retrievals from  
 851 Satellite. The Etna December 2018 Eruption”, *Remote Sens.*, 13, 2225.  
 852 <https://doi.org/10.3390/rs13112225>.
- 853 Cuesta, J., Eremenko, M., Flamant, C., Dufour, G., Laurent, B., Bergametti, G., Höpfner, M.,  
 854 Orphal, J., Zhou, D. (2015) Three-dimensional distribution of a major desert dust outbreak over  
 855 East Asia in March 2008 derived from IASI satellite observations. *J. Geophys. Res. Atmos.*, 120,  
 856 7099–7127.
- 857 Cuesta, J., Flamant, C., Gaetani, M., et al. (2020) Three-dimensional pathways of dust over the  
 858 Sahara during summer 2011 as revealed by new Infrared Atmospheric Sounding Interferometer  
 859 observations. *Q J R Meteorol Soc.*; 146: 2731– 2755. <https://doi.org/10.1002/qj.3814>
- 860 Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E.,  
 861 Nowak, S., Caquineau, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D.,  
 862 Williams, E., and Doussin, J.-F. (2017) Global scale variability of the mineral dust long-wave

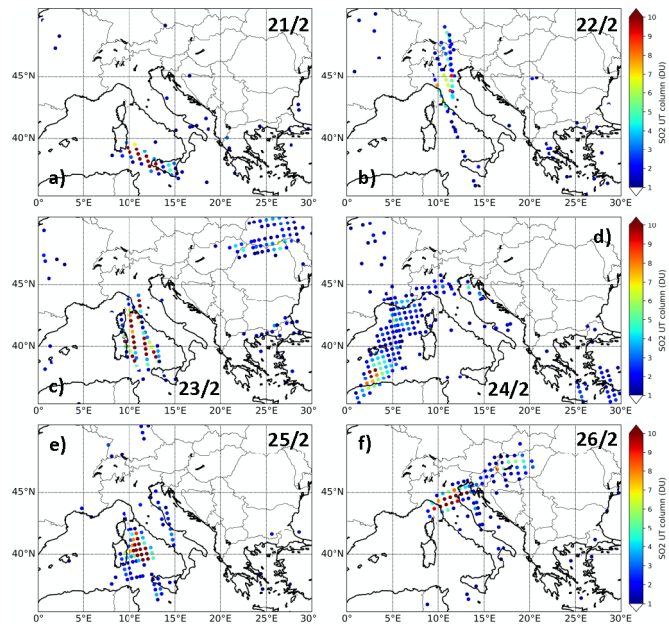
- 863 refractive index: a new dataset of in situ measurements for climate modeling and remote sensing,  
 864 *Atmos. Chem. Phys.*, 17, 1901–1929, <https://doi.org/10.5194/acp-17-1901-2017>.
- 865 M. Dittman et al. (2002), Nadir ultraviolet imaging spectrometer for the NPOESS Ozone  
 866 Mapping and Profiler Suite (OMPS), *Proc. SPIE*, 4814.
- 867 Emde, C., Buras-Schnell, R., Kylling, A., Mayer, B., Gasteiger, J., Hamann, U., Kylling, J.,  
 868 Richter, B., Pause, C., Dowling, T., and Bugliaro, L. (2016) The libRadtran software package for  
 869 radiative transfer calculations (version 2.0.1), *Geosci. Model Dev.*, 9, 1647–1672,  
 870 <https://doi.org/10.5194/gmd-9-1647-2016>.
- 871 Fernald, F. G. "Analysis of atmospheric lidar observations: some comments." *Applied optics*  
 872 23.5, 1984: 652-653.
- 873 Freudenthaler, V., Esselborn, M., Wiegner, M., Heese, B., Tesche, M., and Ansmann, A. (2009)  
 874 Depolarization ratio profiling at several wavelengths in pure Saharan dust during SAMUM 2006.  
 875 *Tellus* 2009, 61B, 165–179. Doi: 10.1111/j.16000889.2008.00396.x.
- 876 Gaudel A., Ancellet G., Godin-Beekmann S. (2015) Analysis of 20 years of tropospheric ozone  
 877 vertical profiles by lidar and ECC at Observatoire de Haute Provence (OHP) at 44°N, 6.7°E,  
 878 *Atmospheric Environment*, 113, 78-89, <https://doi.org/10.1016/j.atmosenv.2015.04.028>.
- 879 Guerrazi, H., Sellitto, P., Cuesta, J., Eremenko, M., Lachatre, M., Mailler, S., Carboni, E.,  
 880 Salerno, G., Caltabiano, T., Menut, L., Serbaji, M.M., Rekhiss, F., Legras, B. (2021)  
 881 Quantitative Retrieval of Volcanic Sulphate Aerosols from IASI Observations. *Remote Sens.*,  
 882 13, 1808. <https://doi.org/10.3390/rs13091808>
- 883 Guerrieri, L., Merucci, L., Corradini, S., Pugnaghi, S. (2015) Evolution of the 2011 Mt. Etna ash  
 884 and SO<sub>2</sub> lava fountain episodes using SEVIRI data and VPR retrieval approach. *J. Volcanol.*  
 885 *Geotherm. Res.*, 291, 63–71.
- 886 Haywood, J. M., Jones, A., Clarisse, L., Bourassa, A., Barnes, J., Telford, P. et al. (2010),  
 887 Observations of the eruption of the Sarychev volcano and simulations using the HadGEM2  
 888 climate model, *Journal of Geophysical Research*, 115, D21212, doi:10.1029/2010JD014447.
- 889 Holben, B.; Eck, T.; Slutsker, I.; Tanré, D.; Buis, J.; Setzer, A.; Vermote, E.; Reagan, J.;  
 890 Kaufman, Y.; Nakajima, T.; et al. (1998) AERONET—A Federated Instrument Network and  
 891 Data Archive for Aerosol Characterization. *Remote Sens. Environ.*, 66, 1–16.
- 892 Hoshyariapour, A. (2021) February 2021: A dusty month for Europe  
 893 <https://blogs.egu.eu/divisions/as/2021/04/03/february-2021-a-dusty-month-for-europe/>
- 894 Khaykin, S. M et al. (2017) Variability and evolution of the midlatitude stratospheric aerosol  
 895 budget from 22 years of ground-based lidar and satellite observations, *Atmos. Chem. Phys.*, 17,  
 896 1829–1845, <https://doi.org/10.5194/acp-17-1829-2017>.
- 897 Kearney, C.S.; Watson, I.M. (2009) Correcting satellite-based infrared sulfur dioxide retrievals  
 898 for the presence of silicate ash., *J. Geophys. Res. Space Phys.*, 114, 22208.
- 899 Keckhut P., Hauchecorne A., and Chanin, M.L. (1993) A critical review on the data base  
 900 acquired for the long term surveillance of the middle atmosphere by French Rayleigh lidars, *J.*  
 901 *Atmos. Oceanic Technol.*, 10, 850-867.

- 902 Lewis, J.R., J.R. Campbell, E.J. Welton, S.A. Stewart, and P.C. Haftings (2016). Overview of  
 903 MPLNET Version 3 Cloud Detection. *J. Atmos. Oceanic Tech.*, 33, 2113 – 2134, doi:  
 904 10.1175/JTECH-D-15-0190.1.
- 905 Lolli, S.; D’Adderio, L.; Campbell, J.; Sicard, M.; Welton, E.; Binci, A.; Rea, A.; Tokay, A.;  
 906 Comerón, A.; Baldasano, R.B.J.M.; et al. Vertically Resolved Precipitation Intensity Retrieved  
 907 through a Synergy between the Ground-Based NASA MPLNET Lidar Network Measurements,  
 908 Surface Disdrometer Datasets and an Analytical Model Solution. *Remote Sens.* 2018, 10, 1102
- 909 Lolli, S.; Vivone, G.; Lewis, J.R.; Sicard, M.; Welton, E.J.; Campbell, J.R.; Comerón, A.;  
 910 D’Adderio, L.P.; Tokay, A.; Giunta, A.; Pappalardo, G. Overview of the New Version 3 NASA  
 911 Micro-Pulse Lidar Network (MPLNET) Automatic Precipitation Detection Algorithm. *Remote*  
 912 *Sens.* 2020, 12, 71. <https://doi.org/10.3390/rs12010071>
- 913 Klett JD. Stable analytical inversion solution for processing lidar returns. *Appl Opt.* 1981 Jan  
 914 15;20(2):211-20. doi: 10.1364/AO.20.000211. PMID: 20309093.
- 915 Kloss, C., Berthet, G., Sellitto, P., Ploeger, F., Taha, G., Tidiga, M. et al. (2021). Stratospheric  
 916 aerosol layer perturbation caused by the 2019 Raikoke and Ulawun eruptions and their radiative  
 917 forcing, *Atmospheric Chemistry and Physics*, 21, 535–560, [https://doi.org/10.5194/acp-21-535-](https://doi.org/10.5194/acp-21-535-2021)  
 918 2021.
- 919 Krotkov, N. A., Krueger, A. J. & Bhartia, P. K. (1997). Ultraviolet optical model of volcanic  
 920 clouds for remote sensing of ash and sulfur dioxide. *Journal of Geophysical Research:*  
 921 *Atmospheres*, 102, 21891–21904. <https://doi.org/10.1029/97JD01690>.
- 922 Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S. et al. (2017)  
 923 Strong constraints on aerosol–cloud interactions from volcanic eruptions. *Nature*, 546, 485–491.
- 924 Merucci, L.; Burton, M.; Corradini, S.; Salerno, G. (2011) Reconstruction of SO<sub>2</sub> flux emission  
 925 chronology from space-based measurements. *J. Volcanol. Geotherm. Res.*, 206, 80–87.  
 926 doi:10.1016/j.jvolgeores.2011.07.002.
- 927 Michaud, J., Krupitsky, D., Grove, J. & Anderson, B. (2005). Volcano related atmospheric  
 928 toxicants in Hilo and Hawaii Volcanoes National Park: Implications for human health.  
 929 *Neurotoxicology*, 26, 555–563. <https://doi.org/10.1016/j.neuro.2004.12.004>.
- 930 Oppenheimer, C., Scaillet, B., & Martin, R. S. (2011). Sulfur Degassing From Volcanoes:  
 931 Source Conditions, Surveillance, Plume Chemistry and Earth System Impacts, *Reviews in*  
 932 *Mineralogy and Geochemistry*, 72, 363–421, doi:10.2138/rmg.2011.73.13, 2011.
- 933 Pisso, I. et al. (2019) The Lagrangian particle dispersion model FLEXPART version 10.4,  
 934 *Geosci. Model Dev.*, 12, 4955–4997, <https://doi.org/10.5194/gmd-12-4955-2019>.
- 935 Prata, A. T., Young, S. A., Siems, S. T., & Manton, M. J. (2017). Lidar ratios of stratospheric  
 936 volcanic ash and sulfate aerosols retrieved from CALIOP measurements. *Atmospheric*  
 937 *Chemistry and Physics*, 17(13), 8599-8618.
- 938 Ridley, D. A., Solomon S., Barnes, J. E., Burlakov, V. D., Deshler, T., Dolgii, S. I. et al. (2014),  
 939 Total volcanic stratospheric aerosol optical depths and implications for global climate change,  
 940 *Geophysical Research Letters*, 41, 7763– 7769, doi:10.1002/2014GL061541.

- 941 Santer, B. D., Bonfils, C., Painter, J. F., Zelinka, M. D., Mears, C., Solomon, S. et al. (2014).  
 942 Volcanic contribution to decadal changes in tropospheric temperature. *Nature Geoscience*, 7,  
 943 185–189.
- 944 Scollo, S.; Prestifilippo, M.; Spata, G.; D’Agostino, M., Coltelli, M. (2009) Monitoring and  
 945 forecasting Etna volcanic plumes. *Nat. Hazards Earth Syst. Sci.*, 9, 1573–1585.
- 946 Sellitto, P. & Briole, P. (2015). On the radiative forcing of volcanic plumes: Modelling the  
 947 impact of Mount Etna in the Mediterranean. *Annals of Geophysics*, 58.  
 948 <https://doi.org/10.4401/ag-6879> (2015).
- 949 Sellitto, P., di Sarra, A., Corradini, S., Boichu, M., Herbin, H., Dubuisson, P., et al. (2016).  
 950 Synergistic use of Lagrangian dispersion and radiative transfer modelling with satellite and  
 951 surface remote sensing measurements for the investigation of volcanic plumes: the Mount Etna  
 952 eruption of 25–27 October 2013, *Atmospheric Chemistry and Physics*, 16, 6841–6861,  
 953 <https://doi.org/10.5194/acp-16-6841-2016>.
- 954 Sellitto, P., Zanetel, C., di Sarra, A., Salerno, G., Tapparo, A. et al. (2017a) The impact of Mount  
 955 Etna sulfur emissions on the atmospheric composition and aerosol properties in the central  
 956 Mediterranean: A statistical analysis over the period 2000–2013 based on observations and  
 957 Lagrangian modelling. *Atmospheric Environment*, 148, 77–88.  
 958 <https://doi.org/10.1016/j.atmosenv.2016.10.032>.
- 959 Sellitto, P., Salerno, G., La Spina, A., Caltabiano, T., Scollo, S., Boselli, A. et al. (2020). Small-  
 960 scale volcanic aerosols variability, processes and direct radiative impact at Mount Etna during  
 961 the EPL-RADIO campaigns. *Scientific Reports*, 10, 15224. <https://doi.org/10.1038/s41598-020-71635-1>
- 962
- 963 Stelitano D., Corradini S., Merucci L., Guerrieri L. (2020) Procedure automatiche per la  
 964 rilevazione e la stima dei parametri delle nubi vulcaniche da satellite, MISCELLANEA INGV,  
 965 Progetto “Sale Operative Integrate e Reti di monitoraggio del futuro: l’INGV 2.0”. Report finale,  
 966 ISSN 1590-2595, 57. <https://doi.org/10.13127/misc/57/20>
- 967 Stevenson, D. S., Johnson, C. E., Collins, W. J. & Derwent, R. G. (2003). The tropospheric  
 968 sulphur cycle and the role of volcanic SO<sub>2</sub>. *Geological Society of London Special Publication*,  
 969 213, 295–305. <https://doi.org/10.1144/GSL.SP.2003.213.01.18>.
- 970 Stiller, G. The Karlsruhe Optimized and Precise Radiative Transfer Algorithm (KOPRA) (2000)  
 971 Wissenschaftliche Berichte, FZKA; Forschungszentrum Karlsruhe: Karlsruhe, Germany, 2000.
- 972 Theys, N., I. De Smedt, H. Yu, T. Danckaert, J. van Gent, C. Hörmann, T. Wagner, P. Hedelt, H.  
 973 Bauer, F. Romahn, M. Pedergnana, D. Loyola, M. Van Roozendaal (2017) Sulfur dioxide  
 974 operational retrievals from TROPOMI onboard Sentinel-5 Precursor: Algorithm Theoretical  
 975 Basis, *Atmos. Meas. Tech.*, 10, 119–153, doi:10.5194/amt-10-119-2017.
- 976 Veefkind, J.P., Aben, I., McMullan, K., Förster, H., De Vries, J., Otter, G., Claas, J.,  
 977 Eskes, H.J., De Haan, J.F., Kleipool, Q., Van Weele, M., Hasekamp, O., Hoogeveen, R.,  
 978 Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and  
 979 Levelt, P.F. (2012). TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for  
 980 global observations of the atmospheric composition for climate, air quality and ozone  
 981 layer applications. *Remote Sensing of Environment*, 120: 70–83.  
 982 doi:10.1016/j.rse.2011.09.027

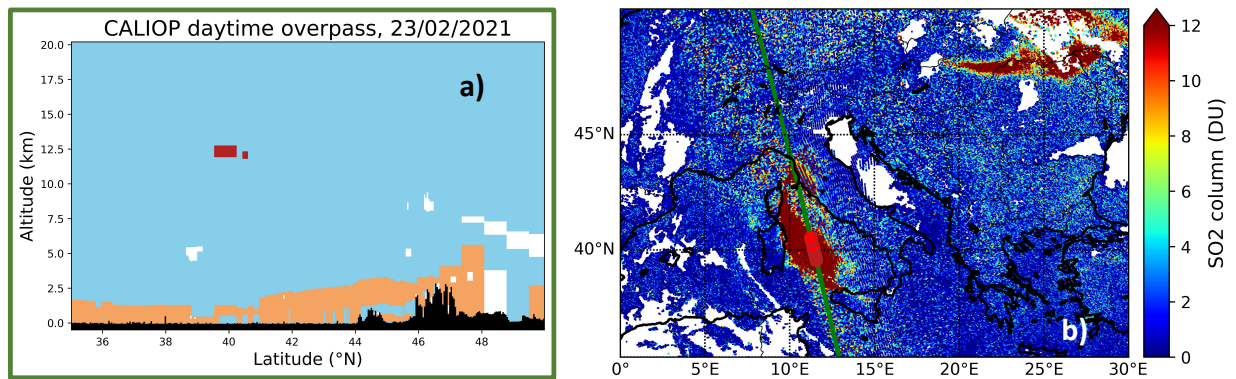
- 983 von Glasow, R., Bobrowski, N. & Kern, C. (2009), The effects of volcanic eruptions on the  
 984 atmospheric chemistry. *Chemical Geology*, 263, 131–142.  
 985 <https://doi.org/10.1016/j.chemgeo.2008.08.020>.
- 986 Wandinger, U., Nicolae, D., Pappalardo, G., Mona, L., Comerón, A. (2020) ACTRIS and its  
 987 aerosol remote sensing component, EPJ Web Conferences 237, 05003,  
 988 <https://doi.org/10.1051/epjconf/202023705003>.
- 989 Webley, P. W., Steensen, T., Stuefer, M., Grell, G., Freitas, S., & Pavolonis, M. (2012).  
 990 Analyzing the Eyjafjallajökull 2010 eruption using satellite remote sensing, lidar and WRF-  
 991 Chem dispersion and tracking model, *Journal of Geophysical Research: Atmospheres*, 117,  
 992 D00U26, doi:10.1029/2011JD016817.
- 993 Welton, E.J. K.J. Voss, H.R. Gordon, H. Maring, A. Smirnov, B. Holben, B. Schmid, J.M.  
 994 Livingston, P.B. Russell, P.A. Durkee, P. Formenti, M.O. Andreae (2000). Ground-based Lidar  
 995 Measurements of Aerosols During ACE-2: Instrument Description, Results, and Comparisons  
 996 with other Ground-based and Airborne Measurements, *Tellus B*, 52, 635-650.
- 997 Welton, E.J.; Campbell, J.R.; Spinhirne, J.D.; Stanley Scott, V., III. (2001). Global monitoring of  
 998 clouds and aerosols using a network of micropulse lidar systems. SPIE Conf. Proc., 4153, 151–  
 999 158
- 1000 Welton, E.J., S.A. Stewart, J.R. Lewis, L.R. Belcher, J.R. Campbell, and S. Lolli (2018). Status  
 1001 of the NASA Micro Pulse Lidar Network (MPLNET): Overview of the network and future plans,  
 1002 new Version 3 data products, and the polarized MPL. EPJ Web of Conferences, 176,  
 1003 <https://doi.org/10.1051/epjconf/201817609003>.
- 1004 Wielicki, B.A., Cess, R.D., King, M.D., Randall, D.A., Harrison, E.F (1995) Mission to planet  
 1005 Earth: Role of clouds and radiation in climate. *Bull. Am. Meteorol. Soc.*, 76, 2125–2154
- 1006 Winker, D.M., Pelon, J., Coakley, J.A., Ackerman, S.A., Charlson, R.J., Colarco, P.R., Flamant,  
 1007 P., Fu, Q., Hoff, R.M., Kittaka, C. et al. (2010) The CALIPSO Mission. *Bull. Am. Meteorol.*  
 1008 *Soc.* 91, 1211–1230.

1009 **Supplementary Information**



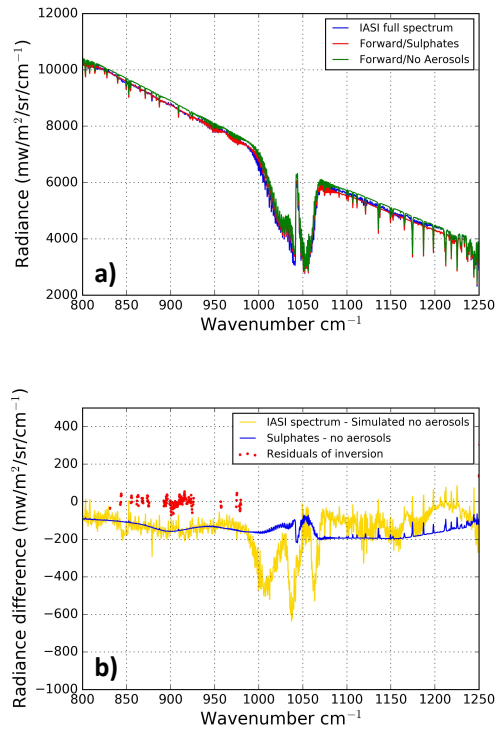
Supplementary Figure 1: OMPS-NM upper-tropospheric SO<sub>2</sub> observations from 21 (panel a) to 26/02/2021 (panel f).

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Supplementary Figure 2: (a) Feature layer classification (same color code as in Fig. 6) for CALIOP daytime overpass, at ~1:30 pm, for 23/02/2021. (b) Sentinel-5p TROPOMI SO<sub>2</sub> column observations for 23/02/2021. The green line in panel b is the ground track of CALIOP observations of panel a. The position of the stratospheric aerosol (volcanic) feature, identified in panel a, is also indicated in panel b as red circles.

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Supplementary Figure 3: The IASI spectrum (blue) taken at OHP (longitude: 5.74°E, latitude: 43.93°N) observed during daytime IASI overpass on 24/02/2021, the AEROIASI-H<sub>2</sub>SO<sub>4</sub> fitted spectrum (red) and a fitted spectrum without SA (green). Spectral radiance differences of the IASI observed spectrum and the fitted spectrum without SA (yellow), the fitted spectra with and without SA (blue) and the fitting residuals (red dots). The SA size distribution is a log-normal with 0.5 μm mean size and 0.5 standard deviation. A mixing ratio of the sulphuric acid of 57% has been considered. The observed spectral signature of SA is observed in the range 800-950 cm<sup>-1</sup>. For more details please refer to Guerrazi et al. (2021). The thermal infrared SA-specific AOD retrieval for this observation is 0.05.