

The eruption run-up at Mt. Etna volcano: Constraining magma decompression rates and their relationships with the final eruptive energy

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16 Abstract

Although explosivity is linked with high decompression rates induced by magma ascent, the 17 quantitative relationships between decompression rate and eruption energy have yet to be properly 18 assessed, especially for open-conduit basaltic volcanoes, where ordinary weak activity can rapidly 19 20 evolve into more intense eruptions. Here, we selected three eruptions of different explosivity from Mt. Etna's recent activity to study the relationships between the observed explosive intensities and 21 22 decompression rates determined through diffusion chronometry, which is based on modeling volatile diffusion along olivine-hosted melt embayments. The approach used in this study has provided 23 important indications on differences in the timescales of decompression-driven degassing for magmas 24 emitted with markedly distinct eruptive dynamics, starting from similar physical and chemical 25 conditions of the magmas involved in the three eruptions. The intense paroxysmal activity at Voragine 26 Crater on December 3, 2015, was fostered by high decompression rate (~0.36-0.74 MPa/s), slightly 27 higher than in the less energetic paroxysm that occurred on February 19, 2013, at New South-East 28 Crater (NSEC) (~0.14-0.29 MPa/s). Decompression rates of magmas emitted during lava fountaining 29 are one order of magnitude greater than values obtained for the mild flank eruption that occurred in 30 December 2018 (~0.045-0.094 MPa/s). Our results indicate that degassing kinetics controlled the 31 intensity of activity at Mt. Etna, thus suggesting that the explosivity does not depend exclusively on 32

the degree of overpressurization of the shallowest reservoir due to injection of gas from the deepestlevels of the plumbing system.

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Keywords: Mt. Etna; decompression rate; melt embayment; diffusion modeling; explosivity.

37

38 1. Introduction

The relatively low viscosity of magmas erupted at basaltic volcanoes generally promotes 39 effusive or weak explosive activity (i.e., Hawaiian and Strombolian). However, basaltic volcanoes 40 can also show more explosive behavior, occasionally Plinian/Subplinian style (Williams, 1983; 41 Walker, 1984; Coltelli et al., 2000, Pérez et al., 2009; Lloyd et al., 2014). Highly explosive eruptions 42 43 represent the major source of hazard at basaltic volcanoes, but the dynamics of magmatic processes and triggering mechanisms of these unusual eruptive phenomena are still poorly understood. 44 45 Furthermore, the unexpected character of these events makes it difficult to correctly interpret precursory signals available from monitoring systems, especially at open-system volcanoes 46 47 characterized by almost persistent activity, where the origin of such signals may be controversial.

Several studies attempted to decipher the processes and factors leading to increased energy 48 during explosive basaltic eruptions (Houghton et al., 2004; Allard et al., 2005; Lloyd et al., 2014; 49 Moretti et al., 2018; Arzilli et al., 2019; La Spina et al., 2021). These studies focused on the role of 50 conduit processes, degassing dynamics, volatile flushing, groundmass crystallization and magma 51 ascent rates. In particular, the decompression rate induced by syn-eruptive magma ascent is believed 52 to be crucial in controlling the explosive intensity of an eruption with respect to other factors (e.g., 53 melt viscosity, crystallinity, conduit geometry). Syn-eruptive ascent not only determines the 54 exsolution of volatiles induced by pressure drop, resulting in increased magma buoyancy, but also 55 controls the style of magma degassing (i.e. equilibrium vs. disequilibrium degassing) and the kinetics 56 of bubble nucleation, growth and coalescence, in turn determining the volatile overpressure (Mangan 57 and Cashman, 1996; Mourtada-Bonnefoi and Laporte, 2004; Gonnermann and Manga, 2007; 58 Rutherford, 2008). Inertial fragmentation or fluid-dynamic breakup of low-viscosity melts are 59 60 induced by rapid expansion of bubbles at high decompression rate (Mangan and Cashman, 1996; La Spina et al., 2021). Kinetics of syn-eruptive degassing also has a significant effect on the rheological 61 62 properties of the magma, which depend mainly on crystallinity (which may increase due to ascentdriven crystallization of the groundmass), the presence of bubbles not escaping from the melt, 63 64 temperature variations, and the increase in melt polymerization due to volatile exsolution. Such parameters strongly control the viscosity of the magma, potentially producing an increase of several 65

orders of magnitude during syn-eruptive ascent and leading therefore to explosive eruptions (Moitra
et al., 2018; Arzilli et al., 2019).

Accurate estimation of magma decompression rates is therefore a priority for modern 68 volcanology. A few estimates are available for both effusive and explosive eruptions involving 69 magmas of different compositions and viscosities mainly at arc volcanoes, where several methods 70 71 based on both geophysical and petrological approaches have been developed (e.g., seismicity, 72 groundmass crystallization, reaction rims in hydrous crystals; Scandone and Malone, 1985; Rutherford and Hill, 1993; Geschwind and Rutherford, 1995). Geochronometers based on fast-73 74 diffusing elements in melt and crystals represent a promising technique for estimating decompression rates caused by syn-eruptive magma ascent rates (Lloyd et al., 2014, 2016; Giuffrida et al., 2018; 75 76 Barth et al., 2019; Viccaro et al., 2021; Myers et al., 2021). In this regard, volatile diffusion along melt pockets partially enclosed within host crystals, called melt embayments or melt tubes 77 78 (Humphreys et al., 2008; Lloyd et al., 2014), takes into account elements directly impacted by ascent-79 driven degassing dynamics, offering the opportunity to investigate timescales relevant to the syneruptive degassing process (from seconds to hours). This approach measures compositional gradients 80 of volatile elements (H₂O, CO₂, and S) that are produced by diffusion from the inner part of the 81 embayment to the outer melt in response to decompression-induced degassing; these gradients can be 82 numerically modeled in order to retrieve timescales of diffusion. 83

This technique has been applied to arc volcanoes involving basaltic and silica-rich magmas (Liu 84 et al., 2007; Humphreys et al., 2008; Lloyd et al., 2014; Moussallam et al., 2019; Myers et al., 2021), 85 as well as to some historical eruptions at Kilauea volcano (Ferguson et al., 2016). However, although 86 87 the hypothesis that intense explosive eruptions are correlated with rapid decompression rates finds general consensus in the scientific community, a true quantification of such relationship has yet to be 88 89 properly addressed. To date, there is a need to investigate more cases of open-system, persistently active basaltic volcanoes, where ordinary Strombolian or effusive activity can suddenly turn into 90 91 unusual and unexpected, highly explosive eruptions.

This study aims to quantify differences in decompression rates related to variable degrees of 92 93 explosivity of basaltic eruptions by modeling volatile diffusion along melt embayments, taking Mt. Etna, one of the world's most active and best-monitored open-conduit volcanoes, as a case study. The 94 95 recent activity of Mt. Etna is characterized by a wide spectrum of eruptive styles, from effusive up to 96 violent ash-dominated lava fountains occurring during paroxysmal eruptions (Aiuppa et al., 2007; 97 Cannata et al., 2018; Viccaro et al., 2019; Borzì et al., 2020; Giuffrida et al., 2021), as well as Subplinian and Plinian events occurred in historical times (Coltelli et al., 2000). Some recent episodes 98 99 from the post-2011 activity were selected for this study mainly for the possibility of integrating

petrological data with those coming from geophysical signals acquired by monitoring networks 100 101 installed on the volcano. The selected eruptions in order of increasing explosive behavior are: 1) the flank eruption of December 24-27, 2018 (DEC 2018 hereinafter), which affected the upper eastern 102 side of the volcanic edifice and was accompanied by intense seismic activity (Borzì et al., 2020), 103 produced an 8-km-high (a.s.l.) eruptive column in the proximal area (Corradini et al., 2020), with a 104 Mass Eruption Rate (MER) of 0.90×10^5 kg/s (inferred using the calculation of Mastin et al., 2009); 105 2) the lava fountaining episode occurred at the New South East Crater (NSEC) on February 19, 2013 106 (NSEC 2018 hereinafter), which is part of the intense 2011-2013 paroxysmal activity (Giuffrida and 107 Viccaro, 2017) and produced a 6-12 km-high (a.s.l.) eruptive plume (Calvari et al., 2018), with MER 108 of 1.72×10⁵ kg/s (Freret-Lorgeril et al., 2018); 3) the paroxysmal eruption occurred at Voragine Crater 109 (VOR) on December 3, 2015 (VOR 2015 hereinafter), which represents the most violent eruptive 110 event at Mt. Etna over the last 20 years (Cannata et al., 2018), producing a 15-km-high (a.s.l.) eruptive 111 column (Calvari et al., 2018) with MER of 1.45×10^6 kg/s. 112

113 The choice of a single volcano and episodes from the same eruptive cycle offers the advantage 114 of reducing the number of variables affecting our modeling, with a fixed plumbing system geometry 115 at depth and comparable physical and chemical conditions of the erupted magmas (e.g., composition, 116 temperature, pressure, viscosity and – at least in part – volatile contents). This strategy allows us to 117 better understand and quantify the relationships between decompression rate and eruptive styles.

118

119 **2. Methods**

Air-quenched tephra samples were collected in distal areas from the point of emission during 120 the eruptions and represent the peak in the explosive activity. Samples were sieved with mesh smaller 121 than 1 cm in order to remove clasts possibly affected by post-eruptive water loss (Lloyd et al., 2013). 122 The fraction between 2 mm and 1 cm was crushed and picked, whereas the fraction smaller than 2 123 mm was sieved without crushing. Olivine crystals of 0.35 - 1.00 mm in length were hand-picked 124 125 from the clasts under a binocular microscope, mounted individually using the Crystalbond 509 mounting adhesive, and polished with silicon carbide papers (P1200 and P2400) and diamond pastes 126 127 (6 μ m, 3 μ m, 1 μ m and $\frac{1}{4}$ μ m) until melt embayments were exposed. Then, olivine crystals were removed from Crystalbond 509 and cleaned in acetone before Raman spectroscopy analyses. Finally, 128 129 they were individually embedded in epoxy resin for electron microprobe analysis (EMPA).

A Renishaw inVia confocal Raman micro-spectrometer (Laboratoire Magmas et Volcans, University of Clermont-Auvergne, France) was used for measuring H₂O concentrations along melt embayments. The micro-spectrometer is equipped with a 532.1 ± 0.3 nm diode laser (200 mW output power), a Peltier-cooled CCD detector of 1024×256 pixels, and a Leica DM 2500 M optical

microscope with a motorized XYZ stage. The laser beam of about 5 mW power was focused at 2 µm 134 under the sample surface using a $100 \times$ objective in high confocality setting (slit aperture of 20 μ m). 135 The spectra were recorded from ~ 100 to 1350 cm⁻¹ (i.e., the spectral region of the alumino-silicate 136 network vibrations) with acquisition time of 120 seconds and from ~2900 to 3800 cm⁻¹ (i.e., the 137 spectral region of the O-H vibrations) for 240 seconds using the WiRETM 4.2 software. Basaltic 138 glasses with different water contents (0 to 2.28 wt.%; Schiavi et al., 2018, and references therein) 139 were used for water quantification using the external calibration procedure (Schiavi et al., 2018) and 140 were analyzed several times during each analytical session in order to correct for the influence of the 141 delivered energy on the band intensities (see Supplementary material for the calibration line). 142 Analytical precision measured on reference glasses was better than 5% relative. The estimated 143 144 accuracy is about 15% relative. Profile compositions of H₂O were measured along the central part of the embayments, from the inner toward the mouth of the melt tubes, with a spacing between each 145 146 spot on the order of $5-8 \ \mu m$.

Major elements, S and Cl in embayments were measured at the Dipartimento di Scienze della 147 Terra, University of Milan, using a JEOL JXA 8200 Superprobe equipped with five wavelength 148 dispersive spectrometers (WDS), one energy dispersive spectrometer (EDS) and one 149 cathodoluminescence detector. The radius of Rowland circle is 140 mm for spectrometers 1, 2 and 3, 150 bearing LDE1, LDE2, LDEB, TAP, PET, and LiF crystals and 100 mm for spectrometers 4 and 5, 151 bearing a PETH and a LIFH crystals. Analytical conditions of 5 µm beam at 15 kV accelerating 152 voltage and ~5 nA beam current were used. Analytical precision for all measured elements is on the 153 order of 5%. 154

A Tescan Vega-LMU scanning electron microscope was also used for collecting major 155 elements in olivine at the Dipartimento di Scienze Biologiche, Geologiche e Ambientali of the 156 University of Catania (Italy). The microscope is equipped with an EDAX Neptune XM4-60 EDS 157 micro-analyzer using an EDS coupled with an EDAX WDS LEXS (low energy WDS) calibrated for 158 light elements. Operating conditions were set at 20 kV accelerating voltage and ~2 nA beam current. 159 Repeated analyses on SPI 02753-AB Serial KF certified standards (Fo-rich olivine) during the 160 analytical runs provided precision for all measured elements of 3-5%. Accuracy is on the order of 161 5%. 162

163

164 **3. Results**

Juvenile fragments (sideromelane and tachylite), crystals and lithics constitute the components of the analyzed pyroclasts (Fig. 1a). Juvenile fragments are abundant in the lava fountaining episodes (66 and 90 vol% respectively for NSEC 2013 and VOR 2015; Fig. 1b-d), whereas the highest percentage of lithics (~50 vol%; Fig. 1e) was observed in DEC 2018 tephra. Crystals consist mainly
of plagioclase and represent <5 vol%.

Three types of external morphologies were recognized in the juvenile fragments: spongy, 170 fluidal, and blocky (cf. Pompilio et al., 2017). Both spongy and fluidal morphologies are present in 171 sideromelane clasts of the NSEC 2013 and VOR 2015 eruptions, although spongy morphology is 172 prevalent in VOR 2015 products. These types of clasts show irregular shapes and contain a large 173 number of small spherical or elliptical coalesced vesicles (Fig. 1b). Fluidal morphology mainly 174 characterizes sideromelane clasts of the NSEC 2013 and DEC 2018 products. Interconnected tubular 175 vesicles are common and are responsible for the elongated shape of these clasts (Fig. 1c). The 176 groundmass of both spongy and fluidal clasts is characterized by low crystallinity. Blocky 177 178 morphologies are mainly observed in tachylite clasts, which are affected by randomly oriented cracks and have low vesicularity (Fig. 1d). Blocky clasts are generally more crystalline compared to spongy 179 180 and fluidal clasts; microlites of plagioclase are predominant in blocky clasts.

Fifteen olivine crystals hosting melt-embayments were selected for this study: 5 from NSEC 181 182 2013 products, 6 from VOR 2015 products and 4 from DEC 2018 products. Olivines occur as subhedral crystals, with rounded edges in the rim portion where the analyzed embayments are located 183 (Fig. 2). Visible portions of surrounding glasses show variable proportions of microcrysts and 184 vesicles: NSEC 2013 and VOR 2015 glasses are more vesiculated and less crystallized than DEC 185 2018 glasses, and plagioclase microcrysts observed in VOR 2015 glasses are chiefly acicular or 186 swallow-tailed (Fig. 1f-h). The examined melt-embayments are mostly tubular-shaped (Fig. 2a-c), 187 but few samples display a more irregular shape (Fig. 2d). A vesicle on the open side of the tube 188 evidences the occurrence of a bubble, which is a key condition to constrain the boundary condition 189 at end of the melt tube in diffusion models. The size of the embayments ranges between 49 and 144 190 µm, with longer embayments more common in NSEC 2013 and VOR 2015 products than in DEC 191 2018. 192

The embayments are hosted in olivine crystals with core compositions ranging from Fo74 to 193 Fo₈₄, whereas the olivine rims show a narrower range from Fo₇₄ to Fo₇₉, with a higher average 194 195 forsterite content for VOR 2015 samples, indicating the existence of compositional zoning (i.e., normal and reverse) from the core of the crystals to the rim. Post-entrapment crystallization (PEC) 196 197 effects on major elements compositions are not considered, since volatiles in embayments are the main objects of interest in this study. Glasses in the studied embayments for the three eruptive 198 199 episodes display similar major element compositions, which are also rather uniform throughout the embayments (Fig. 3; Table S1), whereas matrix glasses outside the host olivine show slightly more 200 201 evolved compositions (Table S1).

202 Water concentration profiles measured using Raman spectroscopy show that the concentrations 203 progressively decrease toward the opening of the embayment (Fig. 4a-c; Table S2). The embayment interior of the NSEC 2013 samples shows water concentrations in the range of 0.9-1.3 wt.%. Higher 204 contents in the range of 1.1-1.7 wt.% characterize the VOR 2015 embayments, except for one sample 205 with a lower concentration (~0.9 wt.%; Fig. 4b). Markedly lower concentrations (0.3-0.7 wt.%) were 206 207 measured in the DEC 2018 embayments. For each eruptive episode, the highest H₂O contents were found in the longest embayments, indicating that diffusive volatile loss affected the shorter ones more 208 extensively. The three eruptive episodes selected for this study differ in the shape of water gradients. 209 The VOR 2015 embayments show slightly decreasing concentrations from the innermost portions 210 toward the opening, while a steep gradient is observed in the last 30-40 µm before the bubble, where 211 212 H₂O concentrations drop to ~0.2-0.5 wt.%. Less steep gradients are observed in the NSEC 2013 embayments starting from 50-70 µm from the opening. The DEC 2018 embayments also display 213 214 gentle gradients, reaching contents of 0.1-0.3 wt.% at the mouth.

Sulfur measurements in the interior of the embayments show that VOR 2015 samples are characterized by slightly higher concentrations, on the order of 718-1300 ppm, than the NSEC 2013 products, with concentrations of 809-1150 ppm, except for one sample characterized by a very low S content (~153-260 ppm) (Fig. 4d-f; Table S1). Compared to NSEC 2013 and VOR 2015 samples, the DEC 2018 products have slightly lower S concentrations ranging from 660 to 885 ppm. Although the concentrations generally decrease toward the open side of the embayments, clear gradient profiles cannot be recognized in some of the analyzed samples.

No gradients are observed for Cl in the studied embayments (Fig. 4g-i; Table S1). NSEC 2013 and VOR 2015 display similar concentrations ranging from 906 to 1721 ppm, with an average value of 1400 ppm. The interior of the embayments from the DEC 2018 eruption is characterized by higher Cl concentrations, which range between 2030 and 2137 ppm.

No CO₂ data are available for the selected embayments from the recent activity at Mt. Etna.
Therefore, the modeling is applied considering only the diffusion of H₂O and S.

228

229 **4. Discussion**

4.1 Definition of the initial and boundary conditions for the diffusion modeling

Open melt tubes are generally considered to be under continuous exchange with the external melt. However, slight differences in their compositions indicate that equilibrium between the embayments and the external melt was not completely attained. The equilibrium between the host olivines and the melt-embayments and surrounding melt was evaluated by plotting Mg# of melts versus Fo measured at the rim of the host crystals (Fig. 5); the equilibrium Fo calculated using either

the K_d ^{Mg-Fe}_{Ol-liq} = 0.30 (Roeder and Emslie, 1970) or the K_d predicted (0.27-0.28) using the model by 236 Toplis (2005) are shown. The compositions of the embayment melts plot outside the equilibrium 237 field, having lower Mg# than that predicted to be in equilibrium with Fo74-79. Similarly, the 238 composition of DEC 2018 matrix glasses is in disequilibrium with the Fo at the olivine rim. By 239 contrast, the Mg# values of the matrix glasses in NSEC 2013 and VOR 2015 samples are in 240 equilibrium with the Fo values measured at the olivine rims. This evidence suggests that 241 crystallization of the host olivine at the interface with the NSEC 2013 and VOR 2015 embayments 242 may have induced some modifications in melt compositions before the final ascent, as highlighted by 243 a slight decrease in MgO in the inner part of the embayment of few samples. The low diffusivities of 244 major elements prevent the restoration of equilibrium between the embayments and the external melt, 245 246 producing thus the observed differences.

Thermodynamic modeling performed using rhyolite-MELTS software (Gualda et al., 2012) 247 tracks the evolution of melt composition in melt inclusions from the same eruptive episodes 248 (Zuccarello et al., 2021). The modeling indicates that the equilibrium between the observed Fo 249 250 compositions at olivine rims and the surrounding melts occurs at 18-24 MPa, 43-77 MPa and 59-116 251 MPa for DEC 2018, NSEC 2013 and VOR 2015 samples, respectively. One way to test the accuracy 252 of these constraints is to determine the saturation pressures of volatiles following the approach described in Lloyd et al. (2014). Specifically, S diffusivity is lower than H₂O diffusivity (Zhang et 253 254 al., 2007), potentially preserving its initial concentrations in the embayments interior during syneruptive degassing in timescales of 10 to 20 minutes, as demonstrated by Lloyd et al. (2014). This is 255 not expected during pre-eruptive degassing, which takes place several hours or days before the 256 eruption, allowing to restore the equilibrium with the external melt. The developing of a combined 257 258 H₂O-CO₂-S degassing modeling, derived from melt inclusion compositions (Fig. 6; Zuccarello et al., 259 2021), allowed us to constrain both initial volatile concentrations and initial pressures for the diffusion 260 modeling, by considering H_2O as the intermediate component between the H_2O -S and H_2O -CO₂ degassing models. Assuming that the concentration of S measured in the interior of the embayment 261 is representative of its original content before ascent, the associated H₂O value derived from the H₂O-262 263 S degassing path (Fig. 6a) can be adopted as the initial concentration for diffusion modeling (Lloyd et al., 2014). Then, the H₂O value inferred from the H₂O-S model is used to constrain the saturation 264 265 pressure on the H₂O-CO₂ degassing path, which is modeled as a closed system with an initial fraction 266 of gas already exsolved of 0.03 (Zuccarello et al., 2021), as defined using VolatileCalc (Newman and 267 Lowenstern, 2002). This saturation pressure is therefore assumed as initial for the diffusion modeling. Sulfur degassing can be modeled through mass balance calculations as suggested by Spilliaert et al. 268 269 (2006), who provide the calculation of bulk partition coefficients (D) of S as a function of

decompression and melt evolution. The closed-system degassing for S (Fig. 6a) is simulated at each 270 step of degassing by using the batch fraction equation with initial $H_2O = 3.37$ wt.% at 300 MPa, 271 according to the H₂O-CO₂ degassing path (Fig. 6b; Zuccarello et al., 2021). Initial exsolution of S 272 occurs at lower pressure (Spilliaert et al., 2006), and the closed-system degassing has been modeled 273 starting from the highest S concentration measured in melt inclusions, 3065 ppm, at a pressure of 180 274 MPa, which is derived from H₂O-CO₂ modeling at a corresponding H₂O content of 2.9 wt.% (details 275 are reported in the Supplementary material). Estimated initial S exsolution pressure and vapor/melt 276 bulk partition coefficient D (which increases progressively from 1 to 60) are consistent with values 277 278 defined for past eruptions at Mt. Etna (Spilliaert et al., 2006).

279 Saturation pressures derived through S degassing model are 80-96 MPa, 62-95 MPa and 95-108 MPa for DEC 2018, NSEC 2013 and VOR 2015, respectively. These values are rather consistent 280 with those inferred from thermodynamic equilibrium between external glasses and olivine rims for 281 282 NSEC 2013 and VOR 2015 and significantly greater for DEC 2018, similar to those estimated for NSEC 2013. We assume that DEC 2018 samples were in equilibrium with the surrounding melt 283 284 before the final ascent, and that syn-eruptive matrix crystallization may have led to subsequent disequilibrium with greater effects on DEC 2018 (Fig. 5) than NSEC 2013 and VOR 2015 products, 285 according with petrographic observations. Therefore, we choose to use as initial pressures those 286 estimated from the volatile degassing model approach. The conversion of the inferred starting 287 pressures to depths is performed by considering an average lithostatic density of 2700 kg/m³, resulting 288 in 0.2 km a.s.l. - 0.9 km b.s.l. and 0.9-1.2 km b.s.l. respectively for the NSEC 2013 and VOR 2015 289 eruptions. These depths are consistent with the sources estimated by strainmeters data, which 290 recorded strain changes of the summit area during lava fountaining episodes caused by sources 291 located at 0 and 1.5 km b.s.l., respectively, for the paroxysmal series occurred at NSEC during the 292 2011-2013 activity (Bonaccorso et al., 2013) and for the paroxysmal activity at VOR on 3-5 293 December 2015 (Bonaccorso and Calvari, 2017). Analogously to what was observed for the 2011-294 295 2013 activity at NSEC, even the source feeding the intrusion of DEC 2018 eruption was located at a depth of 0 km b.s.l. (Aloisi et al., 2020), in agreement with the constraints derived from the degassing 296 297 model. The associated initial H₂O concentrations for NSEC 2013, VOR 2015 and DEC 2018 embayments are constrained respectively in the range of 1.88-2.25 wt.%, 2.28-2.41 wt.% and 2.11-298 2.29 wt.%. 299

Diffusion coefficient is a critical parameter that strongly influences the modeling. Estimation of diffusion coefficient is based on the Arrhenius law and is chiefly a function of temperature, pressure, and melt compositions. For magmas of mafic to intermediate composition, pressure has a minor influence on H₂O diffusivity and can be neglected. Thus, H₂O diffusivity is estimated following
 the equation defined experimentally by Zhang et al. (2007):

$$D_{H_2O} = C_{H_2O} exp\left(-8.56 - \frac{19110}{T}\right)$$
[1]

where C_{H2O} is the concentration of water in wt.%, D_{H2O} the water diffusion coefficient in m²/s and T 305 306 the temperature in K. This equation was developed for basaltic melts, but the influence of variations 307 in melt compositions on D_{H2O} is not considered. The effect of melt compositions on D_{H2O} can be parametrized as a function of the NBO/T ratio (Lloyd et al., 2014). The embayments investigated in 308 309 this study display uniform major element compositions, so an average value of NBO/T can be considered for the H₂O diffusivity correction. We used the method described in Prata et al. (2019) in 310 311 assigning cations to the role of T and for the calculation of NBO/T, providing an average value of NBO/T ~0.53 \pm 0.04 for the melt compositions of the studied embayments, which can be broadly 312 assumed to be intermediate between basalts and more evolved terms such as andesites and trachytes. 313 The parametrization (Fig. 7) was performed by defining two distinct linear relationships between 314 NBO/T and Log D_{H2O} calculated using as end members a basaltic composition (NBO/T = 0.82; Zhang 315 et al., 2007) and either andesitic (NBO/T = 0.26; Behrens et al., 2004) or trachytic compositions 316 (NBO/T = 0.16; Freda et al. (2003). The two parametrizations were performed at T = 1080 °C and 317 $H_2O = 2.41$ wt.%, being respectively the temperature used to run the diffusion modeling (see below) 318 and the highest H₂O value used as initial concentration. Although different from Etnean melt 319 compositions, the andesitic and trachytic terms are the most similar among those available in the 320 321 literature that are useful for the parametrization. The resulting D_{H2O} for NBO/T = 0.53 (±0.04) is lower than the diffusivity calculated through the equation [1] by a factor of 0.25 (± 0.04) and 0.52 322 (±0.04) in the case of basalt-andesite and basalt-trachyte parametrization, respectively. The same 323 324 parametrization at lower H₂O concentrations provided the same value of the correction factor. Two distinct diffusion models have been therefore performed for each embayment, considering the 325 326 correction of D_{H2O} parametrized as a function of either the andesitic or trachytic end-member, i.e. 327 multiplying the equation [1] by 0.25 and 0.52 respectively.

Diffusivity of S is dependent not only on temperature and composition, but also on oxygen fugacity, which affects the speciation of sulfur in the melt. Indeed, changing from oxidizing to reducing conditions results in an increase of about 2.5 orders of magnitude in diffusivity (Zhang et al., 2007). Such a dependence makes it difficult to develop a general equation to define S diffusivity. However, based on experiments on Etna and Stromboli melts, D_S was defined for basaltic liquids at conditions of ~3 log units below the quartz-fayalite-magnetite (QFM) buffer, taking into account the dependence on H₂O concentration (Zhang et al., 2007 and references therein):

$$D_S = exp\left(-8.21 - \frac{27692 - 651.6C_{H_2O}}{T}\right)$$
[2]

Although experiments conditions are more reduced compared to that inferred for Etnean magma (QFM; Zuccarello et al., 2021), the equation [2] was used for calculating S diffusivity for Mt. Etna melts in absence of other experiments on more oxidized systems.

338 A one-dimensional diffusion has been used for modeling the concentration profiles. Initial volatile concentrations are assumed to be homogeneous throughout the embayments and during 339 340 diffusion mass exchange is allowed only from the interior of the embayment to the bubble growing 341 at its mouth. The concentration at the interface between the melt and the bubble at the embayment 342 outlet varies as the volatile content in the surrounding melt changes, according to the pressuredependent degassing model. In contrast, zero-flux is assumed in the interior of the embayment 343 344 adjacent to the host olivine crystal, and no exchange with (or growth of) the host olivine is assumed. Measured H₂O concentrations in the matrix glasses surrounding the host crystals are 0.13-0.35 wt.% 345 346 and have been used to constrain the final concentrations. The corresponding pressures of 1-2 MPa have been used as final conditions for diffusion simulations, assuming instantaneous magma cooling. 347 The diffusion coefficient has been estimated at isothermal conditions during the ascent with a 348 temperature of 1080 °C (±10 °C). This corresponds to the average value constrained by 349 thermodynamic simulations for melts of similar compositions to those measured in the embayments 350 (Zuccarello et al., 2021), which is also consistent with the temperature of Mt. Etna lavas measured at 351 the vent (Calvari et al., 1994). Thus, decompression rate (dP/dt) is the only free parameter allowed to 352 change in the diffusion model, with the goal of identifying the value of decompression rate that 353 minimizes the misfit with the measured H₂O concentrations in the embayment. During 354 decompression, the volatile concentration at the embayment outlet changes as a function of the 355 solubility and the pressure calculated at each step of the simulation. Therefore, variations in dP/dt356 357 affect the shape of the modeled gradients for same initial conditions. Simulations have been performed through a script code in Python language by changing progressively dP/dt within a defined 358 range. Since gradients and diffusivity are better characterized for H₂O than for S, the best fit is 359 determined by minimizing the χ^2 for H₂O modeling as follows: 360

$$\chi^{2} = \sum_{i=1}^{n} \frac{(C_{i} - y_{i})^{2}}{\sigma_{i}^{2}}$$
[3]

where C_i is the measured H₂O concentration at *i* position in the compositional profile, y_i is the modeled concentration and σ_i is the uncertainty associated to the measured concentration (estimated at 8% relative by Raman spectroscopy). Major sources of error are represented by uncertainties in estimated *T*, correction factors for the calculation of D_{H2O} , and initial H₂O concentrations constrained

by the degassing model. A Monte Carlo approach has been employed in order to evaluate the 365 uncertainty for each modeling by running one thousand up to ten thousand simulations, where these 366 parameters have been randomly set within the estimated uncertainties assuming a normal distribution 367 around the constrained central value ($2\sigma_T = \pm 10$ °C, $2\sigma_{\text{corr. factor}} = \pm 0.04$, $2\sigma_{H2Oi} = \pm 0.25$ wt.%). The 368 accuracy associated to the equation [1] determined by Zhang et al. (2007) has not been included in 369 370 the error evaluation since it would systematically affect each decompression rate estimation of the same factor (~3.5-4 in terms of MPa/s). The approach described above estimates a constant 371 decompression rate for each compositional profile modeled in the embayments, assuming that 372 decompression is only driven by magma ascent through the conduit. The conversion to 373 decompression rate is justified by the presence of the bubble at the embayment mouth, presumably 374 375 throughout the magma ascent, given that heterogeneous bubble nucleation requires very low supersaturation pressures (Shea, 2017). In natural systems, it is reasonable that the decompression 376 377 rate from the reservoir to the surface is not constant due to the steep, nonlinear pressure gradients linked with magma vesiculation and fragmentation (Myers et a., 2021), so the estimates provided by 378 379 diffusion modeling can be considered as integrated average values recorded by each embayment investigated in this study. For each modeling, the associated timescale t is calculated as: 380

$$t = \frac{P_i - P_f}{dP/dt}$$
[4]

381 where P_i and P_f are the initial and final pressures and dP/dt is the corresponding decompression rate. 382 The conversion into ascent velocity is calculated as:

$$v = \frac{1}{t} \left(\frac{P_i - P_f}{\rho g} \right)$$
[5]

383 where ρ is the average lithostatic density and g is the gravitational acceleration (9.81 m/s²). 384

385 *4.2 Relationships between magma decompression rates and explosivity*

Microanalyses along olivine-hosted melt embayments highlight H₂O concentration gradients in 386 all the analyzed samples. Many samples display irregular concentration gradients for S, so that only 387 those suitable for diffusion modeling were selected. The results of the volatile diffusion modeling are 388 389 concordant with the expected increase in decompression rates as the explosivity of eruptions increases, where the highest decompression rates have been calculated for the VOR 2015 eruption 390 391 (Fig. 8; Table 1). Modeling performed by using the correction factor of 0.25 yielded decompression 392 rates of 0.28-0.44 MPa/s for VOR 2015 eruption (average $\sim 0.36 \pm 0.09$ MPa/s), 0.09-0.20 MPa/s for 393 NSEC 2013 eruption ($\sim 0.14 \pm 0.10$ MPa/s) and 0.016-0.071 MPa/s for DEC 2018 eruption (~ 0.045 \pm 0.024 MPa/s). Two outliers are represented by the samples NSEC EMB3 and VOR EMB2, with 394 395 decompression rates of 0.35 ± 0.06 MPa/s and 0.039 ± 0.008 MPa/s respectively. Considering the

correction factor of 0.52, we obtained decompression rates of 0.55-0.92 MPa/s for VOR 2015 396 eruption (~ 0.76 ± 0.18 MPa/s), 0.18-0.41 MPa/s for NSEC 2013 eruption (~ 0.29 ± 0.10 MPa/s) and 397 0.035-0.150 MPa/s for DEC 2018 eruption (\sim 0.094 \pm 0.051 MPa/s), where the same outliers from 398 NSEC 2013 and VOR 2015 produced respectively 0.72 ± 0.10 MPa/s and 0.080 ± 0.010 MPa/s. 399 Excluding the outliers, the associated total ascent times range from 1.7 to 6.9 minutes for VOR 2015, 400 from 3.8 to 13.8 minutes for NSEC 2013 and from 9.8 to 83.3 minutes for DEC 2018 eruption. The 401 deviation of the NSEC 2013 outlier from other embayments of the same sample can be attributed to 402 a greater effect of bubble expansion at the mouth of a small embayment in producing an apparently 403 404 higher decompression rate. The longer time estimated for one sample of VOR 2015 eruption (20-40 minutes) could be related to a portion of magma ascending at a lower rate during the increase in 405 406 Strombolian activity before the paroxysmal phase, and then being incorporated into the main flow. Such inference is consistent with the timing of the infrasonic early warning detected 30 minutes 407 before the onset of the paroxysmal phase during the VOR 2015 episode (Ripepe et al., 2018). 408 Alternatively, this olivine crystal could have been incorporated in the slowly ascending portion of the 409 410 multiphase flow in contact with the wall of the conduit, where greater cooling and crystallization may lead to increased viscosity (Taddeucci et al., 2004). Estimated χ^2 values (Table 1) suggest good fits 411 for the H₂O modeling (Fig. 9), considering the significance level at 5% and the degree of freedom 412 (DF) calculated as: $DF = N_D - 1$, where N_D is the number of data points of the modeled embayment. 413 A few samples are characterized by relatively high χ^2 , possibly due to variation in decompression rate 414 that affects natural samples (e.g., the VOR 2015 outlier) or because of slightly irregular H₂O gradients 415 in samples with either a narrow tube neck or a wider mouth opening at the end of the tube, which, 416 coupled with the effect of bubble expansion, can lead to an over-steepening of the H₂O profile 417 predicted by one-dimensional numerical approaches. Therefore, diffusivity is strongly dependent on 418 the embayment shape (deGraffenried and Shea, 2020) and more reliable results from one dimensional 419 diffusion are obtained for tubular-shaped embayments. Concerning S profiles, too high χ^2 values 420 resulted from modeling S diffusion at the same decompression rates estimated for H₂O diffusion, so 421 they cannot be used to determine the goodness of fitting ($\chi^2 > 1000$). 422

The results obtained by modeling volatile diffusion in the embayments are overall consistent with the rates previously estimated for eruptions of comparable explosivity (e.g., Lloyd et al., 2014; Ferguson et al., 2016; Giuffrida et al., 2018). Ascent velocities estimated in this study for the intense NSEC 2013 and VOR 2015 eruptions at Mt. Etna are within the range obtained by Li diffusion modeling on plagioclase for some lava fountaining episodes from the 2011-2013 paroxysmal series at NSEC (4 – 64 m/s; Giuffrida et al., 2018). In addition, volatile diffusion modeling in embayments from the highly explosive 1974 eruption at Volcan de Fuego (Lloyd et al., 2014) and from the Keanakakoi Layer 6 (Kilauea; c. 1650 CE; Ferguson et al., 2016) provided decompression rates of 0.32 – 0.47 MPa/s and ~ 0.45 MPa/s, respectively, which are comparable with those obtained for the lava fountaining of VOR 2015 eruption (0.36 - 0.79 MPa/s). Finally, the maximum timescales of magma ascent calculated for the DEC 2018 eruption (up to 83 minutes) are consistent with the beginning of the seismic sequence which affected the flank of the volcanic edifice a couple of hours before the opening of the fracture (Alparone et al., 2020).

Concerning the lava fountain episodes, the slightly deeper overpressurization of the source 436 feeding the lava fountain during the VOR 2015 eruption may constitute the driving force behind the 437 higher decompression rates estimated for this eruption compared with the NSEC 2013 lava 438 fountaining. On the other hand, the higher MER of VOR 2015 compared with NSEC 2013 (1.45×10⁶ 439 kg/s and 1.72×10^5 kg/s, respectively; Freret-Lorgeril et al., 2018) can be related to a larger conduit 440 radius of the VOR than the NSEC, whose respective values of 3.4 m and 1.6 m were calculated by 441 442 taking into account the conservation of mass, expressed as $Q = \rho u A$, where Q is the MER (kg/s), ρ is the density of the multiphase fluid composed of liquid and gas, u is the ascent velocity (m/s), and A443 444 is the cross-sectional conduit area (m^2) . A larger conduit size should allow more efficient syn-eruptive outgassing, in turn leading to an increase in MER and decompression rate. 445

446

447 5. Concluding remarks and implications

Selected eruptive episodes from the recent, post-2011 activity at Mt. Etna characterized by 448 different explosivity allowed us to get insights into kinetics of magma ascent, evaluating how 449 differences in magma decompression rates influence the energy of eruptions at an open-conduit 450 basaltic volcano. Results from volatile diffusion modeling have demonstrated that the degree of 451 explosivity of eruptions fed by magmas with similar initial physical and chemical conditions is 452 positively correlated with decompression rates. In this regard, the magma erupted during the DEC 453 2018 event produced low-energy explosive activity compared to the two lava fountaining episodes, 454 because of lower magma decompression rates (~ 0.045-0.094 MPa/s) than those estimated for the 455 powerful VOR 2015 (~ 0.36-0.76 MPa/s) and NSEC 2013 (~ 0.14-0.29 MPa/s) eruptions. These 456 457 results are consistent with the more dehydrated features observed in the melt inclusions found in DEC 2018 products, where depleted H₂O concentrations were attributed to melt re-equilibration by 458 459 diffusive water loss (Zuccarello et al., 2021). Significant dehydration can affect magmas under opensystem degassing conditions during storage at pressures <200 MPa, where CO₂ flushing leads to 460 461 simulate a closed-system degassing path. Here, we have shown that the final degree of dehydration is strongly dependent on the kinetics of syn-eruptive ascent to the surface. Indeed, high magma 462 463 decompression rate enables part of the initial H₂O cargo (up to 4.0 wt.% in these products) to be

retained, which coupled with changes in the rheological properties of the magma leads to an increase in gas overpressure. On the contrary, low magma decompression rates favor the escape of volatiles from the melt, inducing therefore more extensive water depletion in melt inclusions by diffusive loss.

Previous studies highlighted a prominent role of gas flushing from the deepest levels of the 467 plumbing system as a major factor controlling explosive dynamics (Allard et al., 2005; Aiuppa et al., 468 2007; Moretti et al., 2018). In this regard, the collapsing foam model (Vergniolle and Jaupart, 1986) 469 has been so far considered more appropriate for explaining the explosive activity at Mt. Etna. 470 However, results obtained from volatile diffusion modeling also highlight an important contribution 471 of syn-eruptive degassing and its kinetics in determining the degree of magma fragmentation (i.e., 472 rise speed model; Parfitt and Wilson, 1995). This inference is also supported by the nature of erupted 473 474 tephra, testified by the presence of highly vesiculated and microlite-poor sideromelane emitted during paroxysmal episodes. As inferred from the investigation of textural characteristics of tephra emitted 475 during other explosive eruptions in the last decades (e.g., the 2000 AD paroxysmal series at the South 476 East Crater; Polacci et al., 2006), a combination of both degassing models can therefore explain the 477 478 dynamics of explosive eruptions at Mt. Etna, where fast syn-eruptive degassing has major effects during the peak of lava fountaining, enabling a more efficient magma fragmentation. This study also 479 emphasizes how the ability of Mount Etna volcano in transferring magmas throughout the upper part 480 of the plumbing system can change over rather limited timespans, even when the same basic and 481 volatile-rich magmas are involved. The chance to degas the original high volatile load more or less 482 efficiently, which is indeed related to variable decompression rates experienced during the final 483 ascent, can have therefore significant influence on the ensuing eruptive behavior, with dramatic 484 485 repercussions in terms of hazard associated.

486

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497 **References**

- Aiuppa, A., Moretti, R., Federico, C., Giudice, G., Liuzzo, M., Papale, P., Shinohara, H., Valenza,
 M., 2007. Forecasting Etna eruptions by real-time observation of volcanic gas composition.
 Geology, 35, 1115-1118. <u>https://doi.org/10.1130/G24149A.1</u>.
- Allard, P., Burton, M., Mure, F., 2005. Spectroscopic evidence for a lava fountain driven by
 previously accumulated magmatic gas. Nature, 433, 407–410.
 <u>https://doi.org/10.1038/nature03246</u>.
- Aloisi, M., Bonaccorso, A., Cannavò, F., Currenti, G., Gambino, S., 2020. The 24 December 2018
 eruptive intrusion at Etna volcano as revealed by multidisciplinary continuous deformation
 networks (CGPS, borehole strainmeters and tiltmeters). J. Geophys. Res: Solid Earth, 125,
 e2019JB019117. https://doi.org/10.1029/2019JB019117.
- Alparone, S., Barberi, G., Giampiccolo, E., Maiolino, V., Mostaccio, A., Musumeci, C., Scaltrito, A.,
 Scarfi L., Tuvè, T., Ursino, A., 2020. Seismological constraints on the 2018 Mt. Etna (Italy)
 flank eruption and implications for the flank dynamics of the volcano. Terra Nova, 1–11.
 https://doi.org/10.1111/ter.12463.
- Arzilli, F., La Spina, G., Burton, M.R., Polacci, M., Le Gall, N., Harteley, M.E., Di Genova, D., Cai,
 B., Vo, N.T., Bamber, E.C., Nonni, S., Atwood, R., Llewellin, E.W., Brooker, R.A., Mader,
 H.M., Lee, P.D., 2019. Magma fragmentation in highly explosive basaltic eruptions induced by
 rapid crystallization. Nat. Geosci., 12, 1023–1028. https://doi.org/10.1038/s41561-019-0468-6.
- Barth, A., Newcombe, M.E., Plank, T., Gonnermann, H., Hajimirza, S., Soto, G.J., Saballos, A.,
 Hauri, E., 2019. Magma decompression rate correlates with explosivity at basaltic volcanoes constraints from water diffusion in olivine. J. Volcanol. Geotherm. Res., 387, 106664.
 https://doi.org/10.1016/j.jvolgeores.2019.106664.
- Behrens, H., Zhang, Y., Xu, Z., 2004. H₂O diffusion in dacitic and andesitic melts. Geochim.
 Cosmochim. Acta 68, 5139–5150. https://doi.org/10.1016/j.gca.2004.07.008.
- Bonaccorso, A., Currenti, G., Linde, A., Sacks, S., 2013. New data from borehole strainmeters to
 infer lava fountain sources (Etna 2011–2012). Geophys. Res. Lett., 40 (14), 3579–3584.
 https://doi.org/10.1002/grl.50692.
- Bonaccorso, A., Calvari, S., 2017. A new approach to investigate an eruptive paroxysmal sequence
 using camera and strainmeter networks: Lessons from the 3–5 December 2015 activity at Etna
 volcano. Earth Planet. Sci. Lett., 475, 231–241. <u>https://doi.org/10.1016/j.epsl.2017.07.020</u>.
- Borzi, A.M., Giuffrida, M., Zuccarello, F., Palano, M., Viccaro, M., 2020. The Christmas 2018
 eruption at Mount Etna: Enlightening how the volcano factory works through a multiparametric
 inspection. Geochem. Geophys. Geosyst., 21, e2020GC009226.
 https://doi.org/10.1029/2020GC009226.

- Calvari, S., Coltelli, M., Neri, M., Pompilio, M., Scribano, V., 1994. The 1991-1993 Etna eruption: 532 chronology and lava flow-field evolution. Acta Vulcanol., 4, 1–14. 533
- Calvari, S., Cannavò, F., Bonaccorso, A., Spampinato, L., Pellegrino, A.G., 2018. Paroxysmal 534 Explosions, Lava Fountains and Ash Plumes at Etna Volcano: Eruptive Processes and Hazard 535 Implications. Front. Earth Sci., 2018, 6, 107. https://doi.org/10.3389/feart.2018.00107. 536
- Cannata, A., Di Grazia, G., Giuffrida, M., Gresta, S., Palano, M., Sciotto, M., Viccaro, M. Zuccarello, 537
- 538 F., 2018. Space-time evolution of magma storage and transfer at Mt. Etna volcano (Italy): The 2015–2016 reawakening of Voragine crater. Geochem. Geophys. Geosyst., 19, 471–495. 539 https://doi.org/10.1002/2017GC007296.

- Coltelli, M., Del Carlo, P., Vezzoli, L., 2000. Stratigraphic constraints for explosive activity in the 541 100 ka at Etna Volcano, Italy. Int. J. Earth Sci., 89, 665-677. 542 past https://doi.org/10.1007/s005310000117. 543
- Corradini, S., Guerrieri, L., Stelitano, D., Salerno, G., Scollo, S., Merucci, L., Prestifilippo, M., 544 Musacchio, M., Silvestri, M., Lombardo, V., Caltabiano, T., 2020. Near real-time monitoring 545 of the Christmas 2018 Etna eruption using SEVIRI and products validation. Rem. Sens., 12 (8), 546 1336. https://doi.org/10.3390/rs12081336. 547
- deGraffenried R., Shea, T., 2020. Modeling diffusion in 1D within melt embayments: correcting for 548 3D geometry. AGU Fall Meeting 2020. 549
- Ferguson, D.J., Gonnermann, H.M., Ruprecht, P., Plank, T., Hauri, E.H., Houghton, B.F., Swanson, 550 D.A., 2016. Magma decompression rates during explosive eruptions of Kilauea volcano, 551 Volcanol., Hawaii, recorded by melt embayments. Bull. 78 (10). 552 https://doi.org/10.1007/s00445-016-1064-x. 553
- Freda, C., Baker, D. R., Romano, C., Scarlato, P., 2003. Water diffusion in natural potassic melts. 554 Geol. Soc. Spec. Publ., 213, 53-62. https://doi.org/10.1144/GSL.SP.2003.213.01.04. 555
- Freret-Lorgeril, V., Donnadieu, F., Scollo, S., Provost, A., Fréville, P., Guéhenneux, Y., Hervier, C., 556 Prestifilippo, M., Coltelli, M., 2018. Mass Eruption Rates of Tephra Plumes During the 2011-557
- 2015 Lava Fountain Paroxysms at Mt. Etna From Doppler Radar Retrievals. Front. Earth Sci., 558 6-73. https://doi.org/10.3389/feart.2018.00073. 559
- Geschwind, C.-H., Rutherford, M.J., 1995. Crystallization of microlites during magma ascent: the 560 fluid mechanics of 1980-86 eruptions at Mount St. Helens. Bull. Volcanol., 57,356-370. 561 https://doi.org/10.1007/BF00301293. 562
- Giuffrida, M., Viccaro, M., 2017. Three years (2011–2013) of eruptive activity at Mt. Etna: Working 563 modes and timescales of the modern volcano plumbing system from micro-analytical studies 564 565 of crystals. Earth Sci. Rev., 171, 289-322. https://doi.org/10.1016/j.earscirev.2017.06.003.

- Giuffrida, M., Viccaro, M. Ottolini, L., 2018. Ultrafast syn-eruptive degassing and ascent trigger
 high-energy basic eruptions. Sci. Rep., 8, 147. <u>https://doi.org/10.1038/s41598-017-18580-8</u>.
- Giuffrida, G., Scandura, M., Costa, G., Zuccarello, F., Sciotto, M., Cannata, A., Viccaro, M., 2021.
 Tracking the summit activity of Mt. Etna volcano between July 2019 and January 2020 by
 integrating petrological and geophysical data. J. Volcanol. Geotherm. Res., 418, article 107350.
 https://doi.org/10.1016/j.jvolgeores.2021.107350.
- Gonnermann, H.M., Manga, M., 2007. The fluid mechanics of volcanic eruptions. Annu. Rev. Fluid
 Mech. 39, 321–356. https://doi.org/10.1146/annurev.fluid.39.050905.110207.
- Gualda, G.A.R., Ghiorso, M.S., Lemons, R. and Carley, T.L., 2012. Rhyolite-MELTS: a modified
 calibration of MELTS optimized for silica-rich, fluid-bearing magmatic systems. J. Petrol., 53,
 875–890. <u>https://doi.org/10.1093/petrology/egr080</u>.
- Houghton, B.F., Wilson, C.J.N., Del Carlo, P., Coltelli, M., Sable, J.E., Carey, R.J., 2004. The 577 influence of conduit processes on changes in style of basaltic Plinian eruptions: Tarawera 1886 578 and Etna 122 BC. J. Volcanol. Geotherm. 137. 1 - 14.579 Res. 580 https://doi.org/10.1016/j.jvolgeores.2004.05.009.
- Humphreys, M.C.S., Menand, T., Blundy, J.D., Klimm, K., 2008. Magma ascent rates inexplosive
 eruptions: Constraints from H₂O diffusion in melt inclusions. Earth Planet. Sci. Lett., 270 (12), 25-40. <u>https://doi.org/10.1016/j.epsl.2008.02.041</u>.
- La Spina, G., Arzilli, F., Llewellin, E.W., Burton, M.K., Clarke, A.B., de' Michieli Vitturi, M.,
 Polacci, M., Hartley, M.E., Di Genova, D., Mader, H.M, 2021. Explosivity of basaltic lava
 fountains is controlled by magma rheology, ascent rate and outgassing. Earth Planet. Sci. Lett.,
 553, 116658. https://doi.org/10.1016/j.epsl.2020.116658.
- Liu, Y., Anderson, A.T., Wilson, C.J.N., 2007. Melt pockets in phenocrysts and decompression rates
 of silicic magmas before fragmentation. J. Geophys. Res. Solid Earth, 112 (B6), 12.
 https://doi.org/10.1029/2006JB004500.
- Lloyd, A., Ruprecht, P., Hauri, E., Rose, W., Gonnermann, H.H., Plank, T., 2014. NanoSIMS results
 from olivine-hosted melt embayments: Magma ascent rate during explosive basaltic eruptions.
 J. Volcanol. Geotherm. Res., 283, 1–18. https://doi.org/10.1016/j.jvolgeores.2014.06.002.
- Lloyd, A.S., Ferriss, E., Ruprecht, P., Hauri, E., Brian, R.J., Plank, T., 2016. An assessment of
 clinopyroxene as a recorder of magmatic water and magma ascent rate. J. Petrol., 57(10), 18651886. https://doi.org/10.1093/petrology/egw058.
- Mastin, L. G., Guffanti, M., Servranckx, R., Webley, P., Barsotti, S., Dean, K., Durant, A., Ewert,
 W., Neri, A., Rose, W. I., Schneider, D., Siebert, L., Stunder, B., Swanson, G., Tupper, A.,
 Volentik, A., and Waythomas, C. F., 2009. A multidisciplinary effort to assign realistic source

- parameters to models of volcanic ash-cloud transport and dispersion during eruptions. J.
 Volcan. Geoth. Res., 186, 10–21. <u>https://doi.org/10.1016/j.jvolgeores.2009.10.013</u>.
- Mangan, M.T., Cashman, K.V., 1996. The structure of basaltic scoria and reticulite and inferences
 for vesiculation, foam formation, and fragmentation in lava fountains. J. Volcanol. Geotherm.
 Res., 73, 1–18. https://doi.org/10.1016/0377-0273(96)00018-2.
- Moitra, P., Gonnermann, H. M., Houghton, B. F., Tiwary, C. S., 2018. Fragmentation and Plinian
 eruption of crystallizing basaltic magma. Earth Planet. Sci. Lett., 500, 97–104.
 https://doi.org/10.1016/j.epsl.2018.08.003.
- Moretti, R., Métrich, N., Arienzo, I., Di Renzo, V., Aiuppa, A., Allard, P., 2018. Degassing vs.
 eruptive styles at Mt. Etna volcano (Sicily, Italy). Part I: Volatile stocking, gas fluxing, and the
 shift from low-energy to highly explosive basaltic eruptions. Chem. Geol., 482, 1-17.
 https://doi.org/10.1016/j.chemgeo.2017.09.017.
- Moussallam, Y., Rose-Koga, E. F., Koga, K. T., Médard, E., Bani, P., Devidal, J.-L., Tari, D.,
 2019. Fast ascent rate during the 2017–2018 Plinian eruption of Ambae (Aoba) volcano: A
 petrological investigation. Contrib. Mineral. Petrol., 174, 90. https://doi.org/10.1007/s00410019-1625-z.
- Mourtada-Bonnefoi C.C., Laporte D., 2004. Kinetics of bubble nucleation in a rhyolitic melt: an
 experimental study of the effect of ascent rate. Earth Planet. Sci. Lett., 218, 521-537.
 https://doi.org/10.1016/S0012-821X(03)00684-8.
- Myers M.L., Druitt T.H., Schiavi F., Gurioli L., Flaherty T., 2021. Evolution of magma decompression and discharge during a Plinian event (Late Bronze-Age eruption, Santorini)
 from multiple eruption-intensity proxies. Bull. Volcanol., 83, 3.
 https://doi.org/10.1007/s00445-021-01438-3.
- Newman, S., Lowenstern, J.B., 2002. VolatileCalc: a silicate melt–H₂O–CO₂ solution model written
 in Visual Basic for excel. Comput. Geosci., 28, 597–604. <u>https://doi.org/10.1016/S0098-</u>
 <u>3004(01)00081-4</u>.
- Parfitt, E.A., Wilson, L., 1995. Explosive volcanic eruptions IX: The transition between Hawaiianstyle lava fountaining and Strombolian explosive activity. Geophys. J. Int., 121, 226–232.
 https://doi.org/10.1111/j.1365-246X.1995.tb03523.x.
- Pérez, W., Freundt, A., Kutterolf, S., Schmincke, H.U., 2009. The Masaya Triple Layer: A 2100 year
 old basaltic multi-episodic Plinian eruption from the Masaya Caldera Complex (Nicaragua). J.
- 631
 Volcanol.
 Geotherm.
 Res.,
 179
 (3-4),
 191-205.

 632
 https://doi.org/10.1016/j.jvolgeores.2008.10.015.

- Polacci, M., Corsaro, R.A., Andronico, D., 2006. Coupled textural and compositional characterization
 of basaltic scoria: Insights into the transition from Strombolian to fire fountain activity at Mount
 Etna, Italy. Geology, 34 (3), 201–204. <u>https://doi.org/10.1130/G22318.1</u>.
- Pompilio, M., Bertagnini, A., Del Carlo, P., Di Roberto, A., 2017. Magma dynamics within a basaltic
 conduit revealed by textural and compositional features of erupted ash: the December 2015 Mt.
 Etna paroxysms. Sci. Rep., 7, 4805. https://doi.org/10.1038/s41598-017-05065-x.
- Ripepe, M., Marchetti, E., Delle Donne, D., Genco, R., Innocenti, L., Lacanna, G., Valade, S., 2018.
 Infrasonic early warning system for explosive eruptions. J. Geophys. Res. Solid Earth, 123,
 9570–9585. https://doi.org/10.1029/2018JB015561.
- Roeder, P.L., Emslie, R.F., 1970. Olivine-Liquid Equilibrium. Contr. Mineral. and Petrol. 29, 275289. https://doi.org/10.1007/BF00371276.
- Rutherford, M.J., Hill, P.M., 1993. Magma ascent rates from amphibole breakdown: Experiments and
 the 1980-1986 Mount St. Helens eruptions. J. Geophys. Res., 98, 19667-19685.
 https://doi.org/10.1029/93JB01613.
- Rutherford, M. J., 2008. Magma ascent rates. Rev. Mineral. Geochem., 69, 241–271.
 https://doi.org/10.2138/rmg.2008.69.7.
- Scandone, R.S., Malone, S., 1985. Magma supply, magma discharge and readjustment of the feeding
 system of Mount St. Helens during 1980. J. Volcanol. Geotherm. Res., 23, 239-262.
 https://doi.org/10.1016/0377-0273(85)90036-8.
- Schiavi, F., Bolfan-Casanova, N., Withers, A.C., Médard, E., Laumonier, M., Laporte, D., Flaherty,
 T., Gómez-Ulla, A., 2018. Water quantification in silicate glasses by Raman spectroscopy:
 correcting for the effects of confocality, density and ferric iron. Chem. Geol., 483, 312–331.
 https://doi.org/10.1016/j.chemgeo.2018.02.036.
- Shea, T., 2017. Bubble nucleation in magmas: A dominantly heterogeneous process? J. Volcanol
 Geotherm. Res., 343, 155-170.
- Spilliaert, N., Allard, P., Métrich, N., Sobolev. A.V., 2006. Melt inclusion record of the conditions of 658 ascent, degassing and extrusion of volatile-rich alkali basalt during the powerful 2002 flank 659 660 eruption of Mount Etna (Italy). J. Geophys. Res., 111, B04203. https://doi.org/10.1029/2005JB003934. 661
- Taddeucci, J., Pompilio, M., Scarlato, P., 2004. Conduit processes during the July-August 2001 662 explosive activity of Mt. Etna (Italy): Inferences from glass chemistry and crystal size 663 664 distribution of ash particles. J. Volcanol Geotherm. Res., 137. 33-54. https://doi.org/10.1016/j.jvolgeores.2004.05.011. 665

- Toplis, M.J., 2005. The thermodynamics of iron and magnesium partitioning between olivine and
 liquid: criteria for assessing and predicting equilibrium in natural and experimental systems.
 Contrib. Mineral. Petrol., 149, 22–39. https://doi.org/10.1007/s00410-004-0629-4.
- Vergniolle, S., Jaupart, C., 1986. Separated twophase flow and basaltic eruptions. J. Geophys. Res.,
 91, 12842–12860. https://doi.org/10.1029/JB091iB12p12842.
- Viccaro, M., Giuffrida, M., Zuccarello, F., Scandura, M., Palano, M., Gresta, S., 2019. Violent
 paroxysmal activity drives self-feeding magma replenishment at Mt. Etna. Sci. Rep., 9, 6717,
 https://doi.org/10.1038/s41598-019-43211-9.
- Viccaro, M., Cannata, A., Cannavò, F., De Rosa, R., Giuffrida, M., Nicotra, E., Petrelli, M., Sacco,
 G., 2021. Shallow conduit dynamics fuel the unexpected paroxysms of Stromboli volcano
 during the summer 2019. Sci. Rep. 11, 266. <u>https://doi.org/10.1038/s41598-020-79558-7</u>.
- Walker, G.P.L., Self, S., Wilson, L., 1984. Tarawera 1886, New Zealand a basaltic plinian fissure
 eruption. J. Volcanol. Geotherm. Res. 21, 61–78. <u>https://doi.org/10.1016/0377-0273(84)90016-</u>
 <u>7</u>.
- Williams, S.N., 1983. Plinian airfall deposits of basaltic composition. Geology 11, 211–214.
 https://doi.org/10.1130/0091-7613(1983)11<211:PADOBC>2.0.CO;2.
- Zhang, Y., Xu, Z., Zhu, M., Wang, H., 2007. Silicate melt properties and volcanic eruptions.
 Rev. Geophys., 72, 171–225. <u>https://doi.org/10.1029/2006RG000216</u>.
- Zuccarello, F., Schiavi, F., Viccaro, M., 2021. Magma dehydration controls the energy of recent
 eruptions at Mt. Etna volcano. Terra Nova, 33, 423-429. https://doi.org/10.1111/ter.12527.





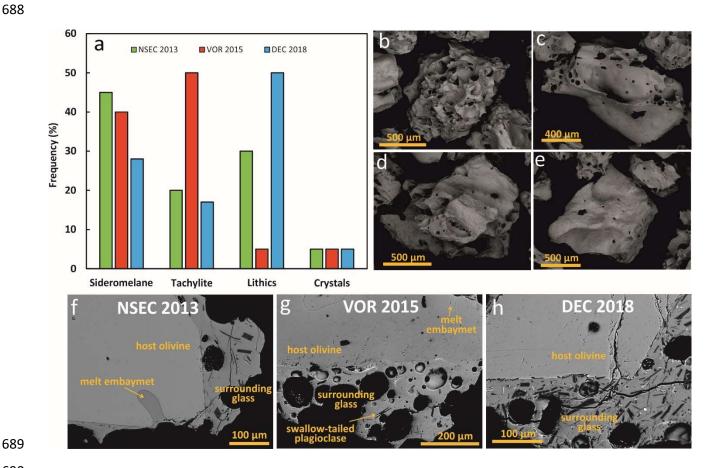


Figure 1. (a) Histograms showing the percentages of clasts found in tephra emitted during the 691 selected eruptive episodes from the post-2011 activity, and BSE images showing: (b) sideromelane 692 clast with spongy morphology; (c) sideromelane clast with fluidal morphology; (d) tachylite clast 693 with blocky morphology; (e) lithic clast; (f) host-olivine and surrounding glass from NSEC 2013 694 samples; (g) host olivine and surrounding glass from VOR 2015 samples; (h) host-olivine and 695 surrounding glass from DEC 2018 samples. 696

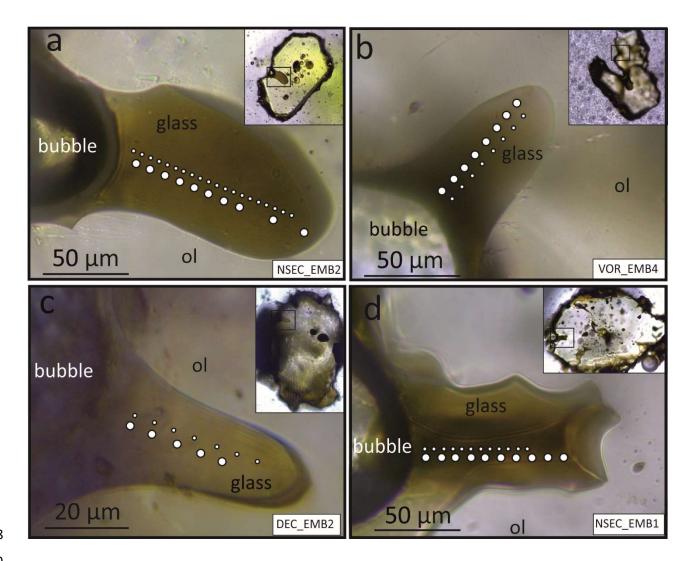


Figure 2. Images taken under optical microscope of representative melt-embayments analyzed using
Raman spectroscopy (small white spots) and EMP (great white spots). Most of samples are tubularshaped (a-c), few samples show more irregular shape (d). Inserts show the whole olivine crystals
hosting melt embayments.

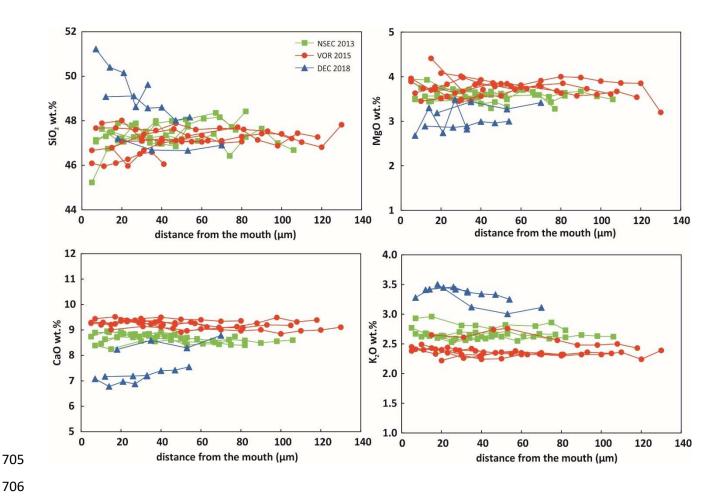


Figure 3. Selected major elements variations in melt-embayments found in products of the post-2011 activity at Mt. Etna as a function of the distance from the mouth of the embayment.

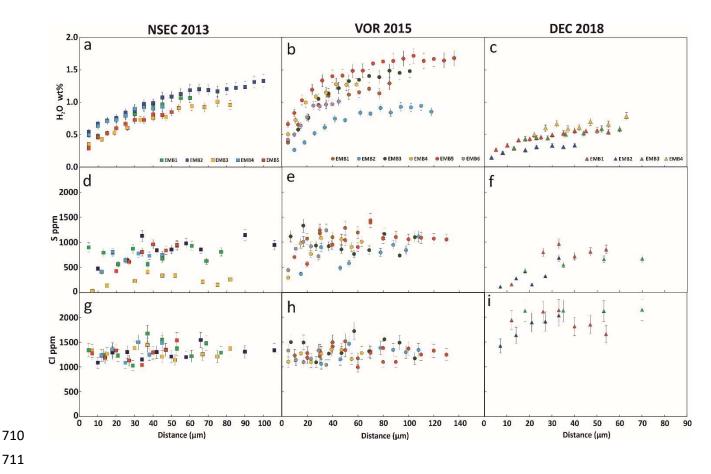




Figure 4. Compositional profiles for H₂O (a-c), S (d-f) and Cl (g-i) measured along each embayment 712 of NSEC 2013 (a, d, g), VOR 2015 (b, e, h) and DEC 2018 (c, f, i) eruptions. S and Cl compositions 713 in DEC2018_emb4 sample were collected by analyzing two spots in the embayment interior (see 714 Table S2). 715

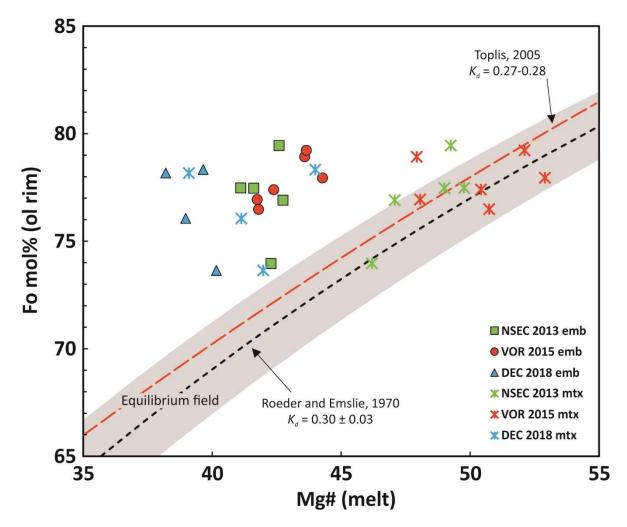


Figure 5. Diagrams showing the measured Fo mol% at the rim of host-olivine crystals vs. Mg# estimated in external melt (stars) and melt embayments for NSEC 2013 (green squares), VOR 2015 (red circles) and DEC 2018 (blue triangles). The equilibrium field (gray area) is calculated by using the $K_D^{\text{ol}(Mg/Fe)/\text{melt}(Mg/Fe)} = 0.30 \pm 0.03$ (the dashed black line represents the central value) by Roeder and Emslie (1970), similar to K_D values predicted using the model by Toplis (2005) (dashed red line).

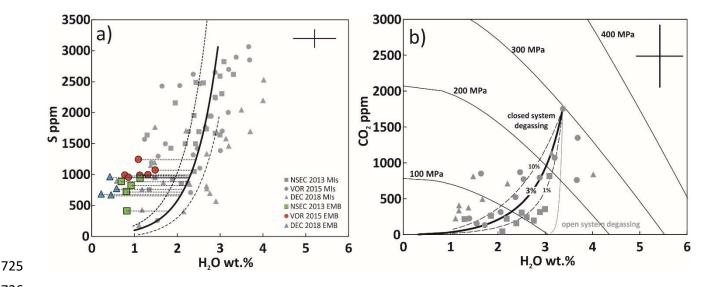
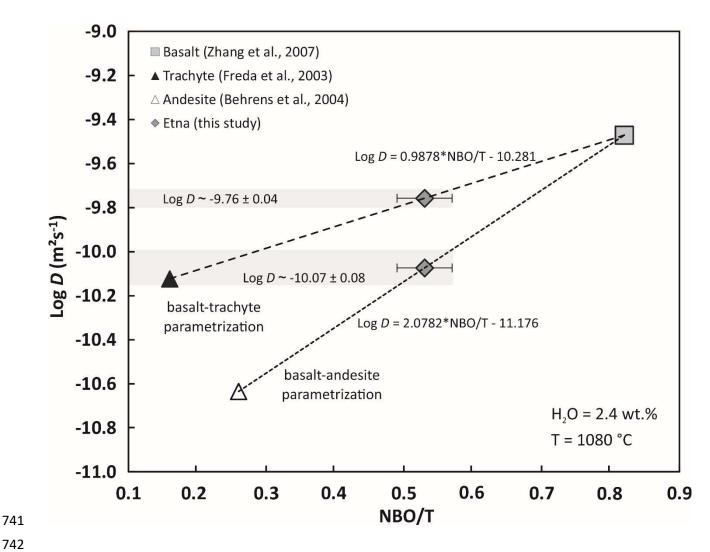




Figure 6. (a) Diagram reporting the average values of H₂O-S compositions in the interior of the 727 embayments and the degassing model for the H₂O-S system on MIs from the same eruptive episodes 728 (restored compositions for volatile loss; data from Zuccarello et al., 2021). Sulfur degassing has been 729 modeled by using the mass balance approach described in Spilliaert et al. (2006) (See supplementary 730 data for details). The thick black line is the best fit model, which is obtained starting from pressure 731 of 180 MPa, S = 3065 ppm and H₂O = 2.9 wt.%, at melt fraction f = 85% and initial gas phase $\alpha_0 =$ 732 3%. Initial H₂O concentrations before diffusive loss during ascent can be inferred from S degassing, 733 734 as well as the corresponding initial pressures are derived from the H₂O-CO₂ system. The dashed lines represent the $\pm 1\sigma$ of S degassing model, while the thin dotted lines indicate the potential initial H₂O 735 contents for each embayment (green squares: NSEC 2013; red circles: VOR 2015; blue triangles: 736 DEC 2018). (b) H₂O-CO₂ closed degassing model on MIs from the same eruptive episodes 737 (Zuccarello et al., 2021). The thick black line is the intermediate path (with initial gas phase = 3%) 738 739 used for constraining initial pressures in the diffusion modeling for each embayment.



742

743 **Figure 7.** Parametrization on H₂O diffusivity (m²/s) for Mt. Etna K-trachybasalt compositions, using a basalt from Zhang et al. (2007) and two different end members: an andesite from Behrens et al. 744 (2004) and a trachyte from Freda et al. (2003). Two distinct linear relationships are defined at $H_2O =$ 745 2.3 wt.% and T = 1080 °C between estimated LogD and NBO/T values. 746

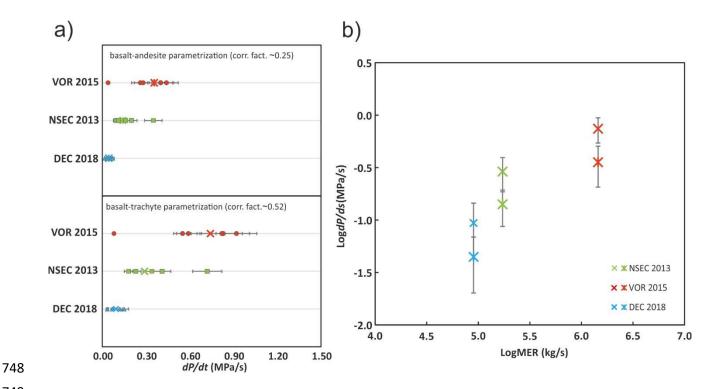




Figure 8. a) Results of decompression rates estimated using the two distinct basalt-andesite and basalt-trachyte parametrizations. Symbols are as shown in Figure 6; stars are averaged decompression rates calculated for the basalt-andesite, while crosses are averaged decompression rates calculated for the basalt-trachyte (outliers are not taken into account in the estimation of averaged values); b) MER vs. dP/dt, expressed as logarithm, showing the relationship between explosivity and decompression rate (symbols as in panel a).

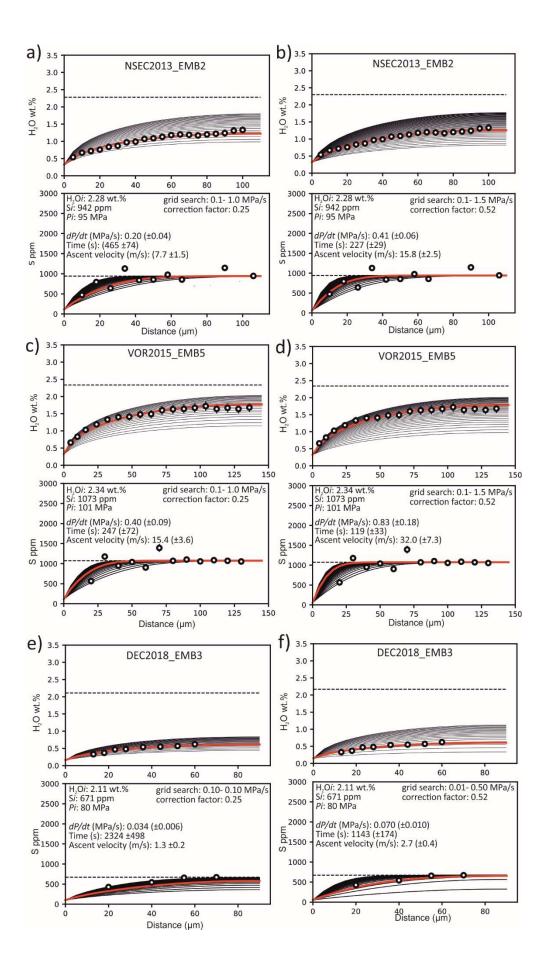


Figure 9. Representative results from volatile diffusion modeling along melt-embayments found in the selected post-2011 eruptions, using the basalt-andesite (a,c,d) and basalt-trachyte (b,d,f) parametrizations for D_{H2O} correction. Red line represents the best fit model corresponding to the estimated dP/dt by χ^2 minimization, starting from fixed initial P_i , S_i and H_2O_i (black dashed line) at constant $T_i = 1080$ °C; thin black lines represent individual simulations for different dP/dt values defined in the search grid used to minimize χ^2 at the same fixed conditions.

	H_2O_i	Si	P_i	P_f	dP/dt (MPa/s ±2 σ)	v (m/s ±2σ)	$\frac{dP/dt}{(MPa/s \pm 2\sigma)}$	v (m/s ±2σ)	t	χ^2
	(wt.%)	(ppm)*	(MPa)	(MPa)	$k_l = 0.25^{**}$	$k_l = 0.25$	$k_2 = 0.52$	$k_2 = 0.52$	(min)	(k_1 / k_2)
NSEC_emb1	2.21	825	89	1	0.16 (±0.03)	6.2 (±1.1)	0.34 (±0.06)	13.1 (±2.4)	4.3-9.2	21.4 / 13.1
NSEC_emb2	2.28	942	95	2	0.20 (±0.04)	7.7 (±1.5)	0.41 (±0.06)	15.8 (±2.5)	3.8-7.8	4.2 / 4.2
NSEC_emb3	1.88	412	62	1	0.094 (±0.018)	3.5 (±0.6)	0.18 (±0.03)	6.9 (±1.2)	5.6-11.3	10.0 / 10.0
NSEC_emb4	2.15	724	83	2	0.35 (±0.06)	13.5 (±2.4)	0.72 (±0.10)	27.7 (±4.1)	1.9-3.9	1.6 / 1.6
NSEC_emb5	2.25	888	92	1	0.11 (±0.02)	4.2 (±1.0)	0.23 (±0.036)	8.8 (±1.5)	6.6-13.8	9.3 / 9.3
Average***					0.14 (±0.10)	7.9 (±4.0)	0.29 (±0.10)	11.2 (±4.0)	4.4-9.2	
VOR_emb1	2.41	1245	108	1	0.26 (±0.06)	10.0 (±2.4)	0.55 (±0.06)	22.0 (±3.3)	3.1-6.9	14.3 / 14.2
VOR_emb2	2.30	992	97	1	0.039 (±0.008)	1.5 (±0.3)	0.08 (±0.01)	3.1 (±0.4)	19.8-41.0	54.9 / 55.0
VOR_emb3	2.31	999	98	1	0.28 (±0.06)	10.8 (±2.4)	0.59 (±0.08)	23.2 (±3.8)	2.7-5.8	22.9 / 22.6
VOR_emb4	2.30	989	97	2	0.44 (±0.08)	16.9 (±3.2)	0.92 (±0.14)	35.4 (±5.7)	1.7-3.6	5.4 / 5.5
VOR_emb5	2.34	1073	101	2	0.40 (±0.09)	15.4 (±3.6)	0.83 (±0.18)	32.0 (±7.3)	2.0-4.1	8.4 / 8.5
VOR_emb6	2.28	956	95	1	0.40 (±0.08)	15.4 (±3.2)	0.82 (±0.14)	31.5 (±5.7)	1.9-3.9	14.7 / 14.6
Average****					0.36 (±0.08)	13.7 (±3.1)	0.74 (±0.16)	28.8 (±5.9)	2.3-4.9	
DEC_emb1	2.29	965	96	1	0.057 (±0.012)	2.2 (±0.5)	0.12 (±0.018)	4.7 (±0.8)	13.1-27.8	1.9 / 1.9
DEC_emb2	2.12	686	81	1	0.016 (±0.002)	0.6 (±0.1)	0.035 (±0.008)	1.3 (±0.3)	38.6-83.3	4.0 / 4.0
DEC_emb3	2.11	671	80	1	0.034 (±0.006)	1.3 (±0.2)	0.07 (±0.01)	2.7 (±0.4)	19.1-38.7	4.3 / 4.3
DEC_emb4	2.18	773	86	1	0.071 (±0.010)	2.7 (±0.4)	0.15 (±0.03)	5.6 (±1.0)	9.8-20.0	5.4 / 5.4
Average					0.045 (±0.024)	1.7 (±0.9)	0.094 (±0.051)	3.6 (±1.9)	20.1-42.4	

Table 1. Results from volatile diffusion modeling on melt embayments of NSEC 2013, VOR 2015
and DEC 2018 eruptions at Mt. Etna.

* S initial concentrations are average values considering the first 1-4 spots measured in the embayments

771 interior.

** k_1 , k_2 are correction factors linked respectively to the basalt-andesite and basalt-trachyte parametrizations.

*** Sample NSEC 2013_emb3 is not considered in the calculation of average values.

**** Sample VOR2015_emb2 is not considered in the calculation of average values.