

# Magma mixing during conduit flow is reflected in melt-inclusion data from persistently degassing volcanoes

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## Abstract

Persistent volcanic activity is thought to be linked to degassing, but volatile transport at depth cannot be observed directly. Instead, we rely on indirect constraints such as CO<sub>2</sub>-H<sub>2</sub>O concentrations in melt inclusions trapped at different depth, but this data is rarely straight-forward to interpret. In this study, we develop a multiscale model of conduit flow during passive degassing to identify how flow behavior in the conduit is reflected in melt-inclusion data and surface gas flux. During the approximately steady flow likely characteristic of passive-degassing episodes, variability in degassing arises primarily from two processes, the mixing of volatile-poor and volatile-rich magma and variations in CO<sub>2</sub> influx from depth. To quantify how conduit-flow conditions alter mixing efficiency, we first model bidirectional flow in a conduit segment at the scale of tens of meters while fully resolving the ascent dynamics of intermediate-size bubbles at the scale of centimeters. We focus specifically on intermediate-size bubbles, because these are small enough not to generate explosive behavior, but large enough to alter the degree of magma mixing. We then use a system-scale volatile-concentration model to evaluate the joint effect of magma mixing and CO<sub>2</sub> influx on volatile concentrations profiles against observations for Stromboli and Mount Erebus. We find that the two processes have distinct observational signatures, suggesting that tracking them jointly could help identify changes in conduit flow and advance our understanding of eruptive regimes.

1 **Magma mixing during conduit flow is reflected in**  
2 **melt-inclusion data from persistently degassing**  
3 **volcanoes**

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7 **Key Points:**

- 8 • Magma mixing occurs commonly at the interface of up-welling and down-welling  
9 magma in persistently degassing volcanoes  
10 • Bubble speed, magma viscosities, bubble volume fraction, and the shear stress at  
11 the interface control magma mixing  
12 • Magma mixing and carbon dioxide influx have distinct observational signatures  
13 in melt-inclusion data

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**Abstract**

Persistent volcanic activity is thought to be linked to degassing, but volatile transport at depth cannot be observed directly. Instead, we rely on indirect constraints such as CO<sub>2</sub>-H<sub>2</sub>O concentrations in melt inclusions trapped at different depth, but this data is rarely straight-forward to interpret. In this study, we develop a multiscale model of conduit flow during passive degassing to identify how flow behavior in the conduit is reflected in melt-inclusion data and surface gas flux. During the approximately steady flow likely characteristic of passive-degassing episodes, variability in degassing arises primarily from two processes, the mixing of volatile-poor and volatile-rich magma and variations in CO<sub>2</sub> influx from depth. To quantify how conduit-flow conditions alter mixing efficiency, we first model bidirectional flow in a conduit segment at the scale of tens of meters while fully resolving the ascent dynamics of intermediate-size bubbles at the scale of centimeters. We focus specifically on intermediate-size bubbles, because these are small enough not to generate explosive behavior, but large enough to alter the degree of magma mixing. We then use a system-scale volatile-concentration model to evaluate the joint effect of magma mixing and CO<sub>2</sub> influx on volatile concentrations profiles against observations for Stromboli and Mount Erebus. We find that the two processes have distinct observational signatures, suggesting that tracking them jointly could help identify changes in conduit flow and advance our understanding of eruptive regimes.

**Plain Language Summary**

Some volcanoes like Stromboli or Mount Erebus, named persistently degassing volcanoes, erupt multiple times a day, emitting copious gas and thermal energy with little magma. Direct measurements of these volcanoes provide rich datasets for understanding how these volcanic systems work. Without the ability to observe processes at depth before magma reaches the surface, we rely on erupted samples to interpret these processes. Some of these samples seal magma droplets named melt inclusions during ascent, which thus represent valuable snapshots of magma composition. Here we study how the magma flow in the conduit connecting the surface to the source of magma contribute to the compositions of melt inclusions using numerical simulations. We demonstrate that the gas-rich, up-welling magma will mix with the down-welling magma, which loses its gas at the surface. The degree of mixing depends on the physical properties of magma and gas bubbles. This magma mixing, together with the influx of carbon dioxide into the system, significantly shift the concentrations of water and carbon dioxide in melt inclusions. Our study shows that magma mixing is almost inevitable in persistently degassing volcanoes. We suggest that melt inclusion data could potentially help us track the evolving flow conditions in volcanic conduits.

**1 Introduction**

Not all volcanic activity is rare: Persistently degassing volcanoes like Stromboli, Italy, or Mount Erebus, Antarctica, typically erupt multiple times a day (Dibble et al., 1988; Burton, Allard, et al., 2007). While eruptions are frequent, they are mild by volcanic standards and can be monitored directly, providing rich datasets for constraining how these volcanic systems work (Burton, Allard, et al., 2007; Oppenheimer et al., 2009; Johnson et al., 2008; Ilanko et al., 2015; Ripepe et al., 2015).

Measurements of surface gas fluxes show that persistently degassing volcanoes continually emit copious quantities of gas and thermal energy, but rarely erupt magma (Stoiber & Williams, 1986; Allard et al., 1994; Kazahaya et al., 1994; Palma et al., 2008; Oppenheimer et al., 2009; Woitischek et al., 2020). This imbalance suggests that more magma is being degassed than erupted, which leads to bidirectional flow of volatile-rich, less viscous magma ascending in the center of the conduit and volatile-poor, more viscous magma

63 descending along the sides (Francis et al., 1993; Kazahaya et al., 1994; Stevenson & Blake,  
64 1998).

65 The concept of bidirectional flow is appealing from a theoretical point of view, be-  
66 cause it provides the significant thermal energy flux required to maintain open-system  
67 conditions in persistently degassing volcanoes. Evaluating it from an observational point  
68 of view, has proven more challenging. One exception is the 1959 eruption at Kilauea Iki,  
69 Hawaii, where recent work suggests that the predominance of certain misalignment an-  
70 gles in olivine glomerocrysts emerges naturally only when the pre-eruptive conduit flow  
71 field was bidirectional (DiBenedetto et al., 2020). However, the majority of degassing  
72 observations refer to non-eruptive conditions (e.g., Burton, Allard, et al., 2007; Oppen-  
73 heimer et al., 2009; Ruth et al., 2018), emphasizing the need to link flow conditions and  
74 degassing processes during approximately steady conditions.

75 Some erupted samples can be used to reconstruct the degassing processes prior to  
76 eruption, because they contain host crystals that have entrapped small droplets of melts  
77 during their growth (e.g., Métrich et al., 2001, 2010; Oppenheimer et al., 2011; Rasmussen  
78 et al., 2017). These melt inclusions are sealed in at various depth and thus represent valu-  
79 able snapshots of evolving melt compositions (Ruth et al., 2018). Patching together these  
80 snapshots to obtain a consistent picture of degassing at depth, however, is hindered by  
81 the limited fidelity with which melt-inclusion seal in pre-eruptive conditions at depth (Bucholz  
82 et al., 2013; Aster et al., 2016; Barth et al., 2019) and measurement uncertainty (Oppenheimer  
83 et al., 2011). Another important observable that helps to constrain steady degassing is  
84 the surface-gas flux (Burton, Allard, et al., 2007; Oppenheimer et al., 2009; Ilanko et al.,  
85 2015). Surface gas flux measurements provide an important complement to melt inclu-  
86 sion data, because melt inclusions only seal melt and are unsuitable for estimating the  
87 total budgets of volatiles with low solubility, such as CO<sub>2</sub> (e.g., Wallace, 2005; Burton,  
88 Mader, & Polacci, 2007)

89 The goal of this study is to quantify how different rates of magma mixing during  
90 conduit flow and variations in CO<sub>2</sub> influx alter the volatile concentrations recorded by  
91 melt-inclusions during passive degassing. We hypothesize that CO<sub>2</sub> influx (Burton, Mader,  
92 & Polacci, 2007; Blundy et al., 2010; Métrich et al., 2010; Rasmussen et al., 2017) and  
93 magma mixing (Witham, 2011a; Moussallam et al., 2016) leave distinct observational  
94 signatures in melt-inclusion data. Identifying these distinct observational signatures would  
95 allow distinguishing between the relative importance of the two processes during con-  
96 duct flow and potentially afford new insights into their relationship with eruptive behav-  
97 ior. Spilliaert et al. (2006) provide a proof-of-concept of this idea, but without linking  
98 in a magma dynamics model.

99 To connect conduit flow to melt-inclusion data, we link a multiscale model of bidi-  
100 rectional conduit flow to a volatile-concentration model. The conduit-flow model is mul-  
101 tiscale in the sense that it resolves both the flow dynamics of a conduit segment at the  
102 tens-of-meter scale and the ascent dynamics of centimeter-scale bubbles through a di-  
103 rect numerical approach (Qin & Suckale, 2017; Suckale et al., 2018; Qin et al., 2020). We  
104 focus on resolving intermediate-size bubbles at the scale of centimeters that are buoy-  
105 ant enough to decouple from the magmatic liquid and ascend, but not so large that they  
106 might be related to eruptive behavior (e.g., Jaupart & Vergnolle, 1988). Smaller crys-  
107 tals and or bubbles at the millimeter scale have much smaller ascent speeds and hence  
108 remain largely entrained (Tryggvason et al., 2013). As a consequence, their main effect  
109 is to alter the effective material properties of the bubble-crystal-melt mixture (Bowen,  
110 1976).

111 We test our hypothesis by comparing model results against the volatile concentra-  
112 tions recorded in melt inclusions. We first quantify magma mixing with the conduit-flow  
113 model and then use the volatile-concentration model based on Witham (2011a) to cal-  
114 culate the associated system-scale concentration profiles. We focus specifically on Strom-

115 boli and Mount Erebus, because of their abundance of melt inclusion data (Métrich et  
 116 al., 2010; Oppenheimer et al., 2011; Rasmussen et al., 2017), the availability of contin-  
 117 uous measurements of surface gas fluxes (Burton, Allard, et al., 2007; Oppenheimer et  
 118 al., 2009; Ilanko et al., 2015), and the relatively steady patterns of their degassing and  
 119 eruption activities (Allard et al., 1994; Burton, Allard, et al., 2007; Oppenheimer et al.,  
 120 2009; Métrich et al., 2010; Oppenheimer et al., 2011; Rasmussen et al., 2017).

121 A particularly puzzling observation is that melt inclusions from many persistently  
 122 degassing volcanoes consistently indicate higher CO<sub>2</sub> content than predicted by either  
 123 closed-system or open-system degassing path (Métrich & Wallace, 2008; Métrich et al.,  
 124 2010; Blundy et al., 2010; Oppenheimer et al., 2011; Yoshimura, 2015; Rasmussen et al.,  
 125 2017; Barth et al., 2019). In contrast, melt inclusions from more silicic volcanoes appear  
 126 to match the expected trends more closely (e.g., Schmitt, 2001; Liu et al., 2006), sug-  
 127 gesting that melt inclusions may at least partially reflect systematic differences in con-  
 128 duct flow between different volcanic systems. While CO<sub>2</sub> influx (Burton, Mader, & Po-  
 129 lacci, 2007; Shinohara, 2008; Blundy et al., 2010; Métrich et al., 2010; Rasmussen et al.,  
 130 2017) and magma mixing (Dixon et al., 1991; Witham, 2011a; Sides et al., 2014) are of-  
 131 ten presented as alternative explanations (Métrich et al., 2011; Witham, 2011b), we ar-  
 132 gue here that they both contribute to the observed variability in volatile concentrations,  
 133 but do so in distinct ways.

## 134 2 Method

135 From individual bubbles and crystals to transcrustal plumbing systems (Cashman  
 136 et al., 2017), volcanic systems bridge ten orders of magnitude in spatial scales or more  
 137 (e.g., fig. 1). Fully resolving all physical and chemical processes over this vast spectrum  
 138 of spatial scales at the accuracy necessary to understand the nonlinear dynamics of a highly  
 139 coupled system is not possible. Instead, we develop a customized multiscale model that  
 140 focuses on the key elements required for linking bidirectional conduit flow and observa-  
 141 tions of melt-inclusions and surface-gas flux. Our model consists of two main components,  
 142 the conduit-flow model and the volatile-concentration model, described in more detail  
 143 in the next two sections.

### 144 2.1 Conduit-flow Model

145 Transcrustal plumbing system (Cashman et al., 2017; Magee et al., 2018) consists  
 146 of vertically stacked melt-rich tabular lenses and vertical conduit-like segments transiently  
 147 connecting these lenses (see fig. 1D). While magma properties, such as gas volume frac-  
 148 tion and melt viscosity, can vary significantly over the entirety of this system, we assume  
 149 that they are approximately constant at the scale of the vertical, conduit-like segments  
 150 (fig. 1C). This assumption implies that exsolution and dissolution are negligible within  
 151 the segments. Volatiles exsolved at depth provide the buoyancy required for the ascent  
 152 of volatile-rich magma. Upon degassing at the free surface, volatile-poor magma remains  
 153 and sinks back to depth, creating a bidirectional flow field (Blake & Campbell, 1986; Fran-  
 154 cis et al., 1993; Kazahaya et al., 1994; Stevenson & Blake, 1998; Molina et al., 2012). More  
 155 specifically, we assume core-annular flow here, because this particular bidirectional flow  
 156 field is most commonly observed in vertical pipes at moderate to high viscosity contrasts  
 157 (Stevenson & Blake, 1998; Beckett et al., 2011; Suckale et al., 2018).

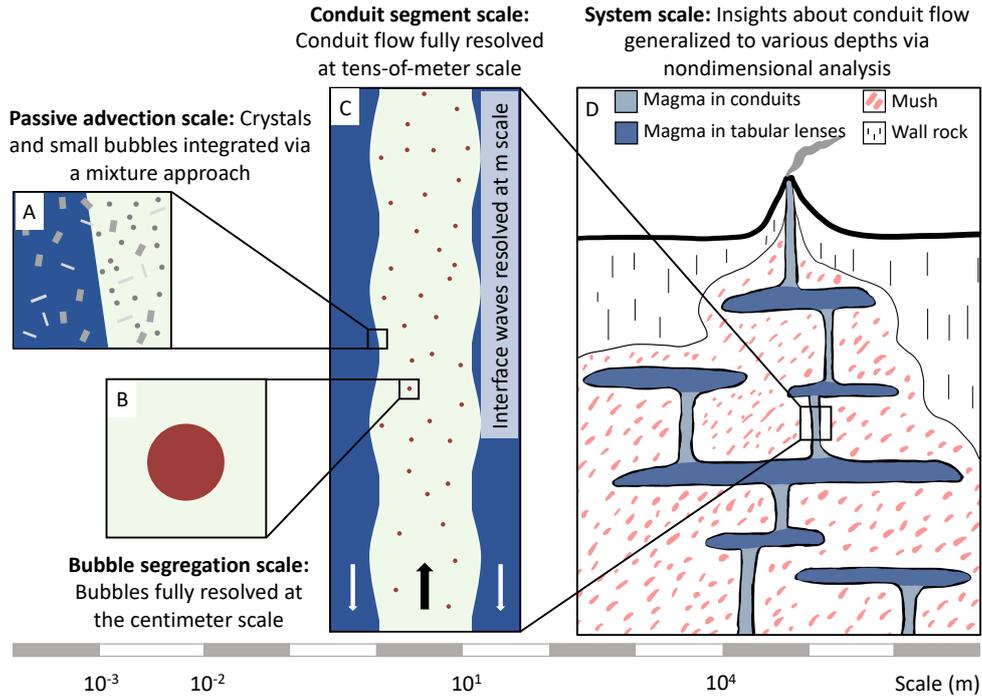
158 In the conduit segments, centimeter-scale gas bubbles segregate from the ambient  
 159 magma flow and rise towards the surface to degas. We capture these bubbles explicitly  
 160 using direct numerical simulations (fig. 1B). Crystals and millimeter-scale bubbles, how-  
 161 ever, have much smaller segregation speeds and hence largely remain entrained in the  
 162 ambient magma flow. We represent these implicitly through a mixture approximation  
 163 (Bowen, 1976) by reducing their effect to changes in the effective density and viscosity  
 164 of the crystal- and bubble-bearing magma (fig. 1A). For the rest of this manuscript, we

**Table 1.** Definition of symbols

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$\rho(kg/m^3)$ :	magma density
$\mu(kg/m^3)$ :	magma viscosity
$\rho_c(kg/m^3)$ :	density of volatile-rich magma
$\rho_a(kg/m^3)$ :	density of volatile-poor magma
$\rho_b(kg/m^3)$ :	density of bubbles
$M_b(kg)$ :	mass of a bubble
$\mathbf{F}_b(N)$ :	hydrodynamic force exerted onto the bubble by the surrounding magma
$\mathbf{X}_b(m)$ :	bubble location
$\mu_c(Pa \cdot s)$ :	viscosity of volatile-rich magma
$\mu_a(Pa \cdot s)$ :	viscosity of volatile-poor magma
$\mathcal{S}$ :	speed ratio
$\mathcal{I}$ :	interface stability number
$\Gamma(\%/MPa)$ :	mixing factor
$\sigma(\%/MPa)$ :	error of mixing factor
$R(m)$ :	conduit radius
$L(m)$ :	conduit length
$r(m)$ :	bubble radius
$\phi$ :	volume fraction of resolved bubbles in volatile-rich magma
$\phi_{tot}$ :	total volume fraction of resolved and subgrid bubbles in volatile-rich magma
$c$ :	concentration variable in conduit-flow simulations
$D(m^2/s)$ :	diffusion coefficient
$i_u$ :	weight percent of dissolved volatiles in up-welling magma
$i_d$ :	weight percent of dissolved volatiles in down-welling magma
$i_g$ :	weight percent of exsolved volatiles in up-welling magma
$i_*$ :	effective up-welling volatile content
$p(Pa)$ :	pressure in the conduit-flow model
$P(Pa)$ :	pressure in the calculation of volatile concentration profiles
$\Delta p(Pa)$ :	pressure step size
$P_{min}(Pa)$ :	minimum pressure in the calculation of volatile concentration profiles
$P_{max}(Pa)$ :	maximum pressure in the calculation of volatile concentration profiles
$\mathbf{v}(m/s)$ :	velocity
$\mathbf{V}_b(m/s)$ :	bubble velocity
$U(m/s)$ :	characteristic speed of the analytical solution of core-annular flow
$v_c(m/s)$ :	vertical speed at the center line of the analytical solution
$v_b(m/s)$ :	analytical bubble rise speed
$\tau_{xy}(Pa)$ :	simulated shear stress
$\tau(Pa)$ :	analytical interfacial shear stress
$t$ :	nondimensional time
$g(m/s^2)$ :	gravitational acceleration
$\lambda$ :	H <sub>2</sub> O/CO <sub>2</sub> in the gas phase at the surface

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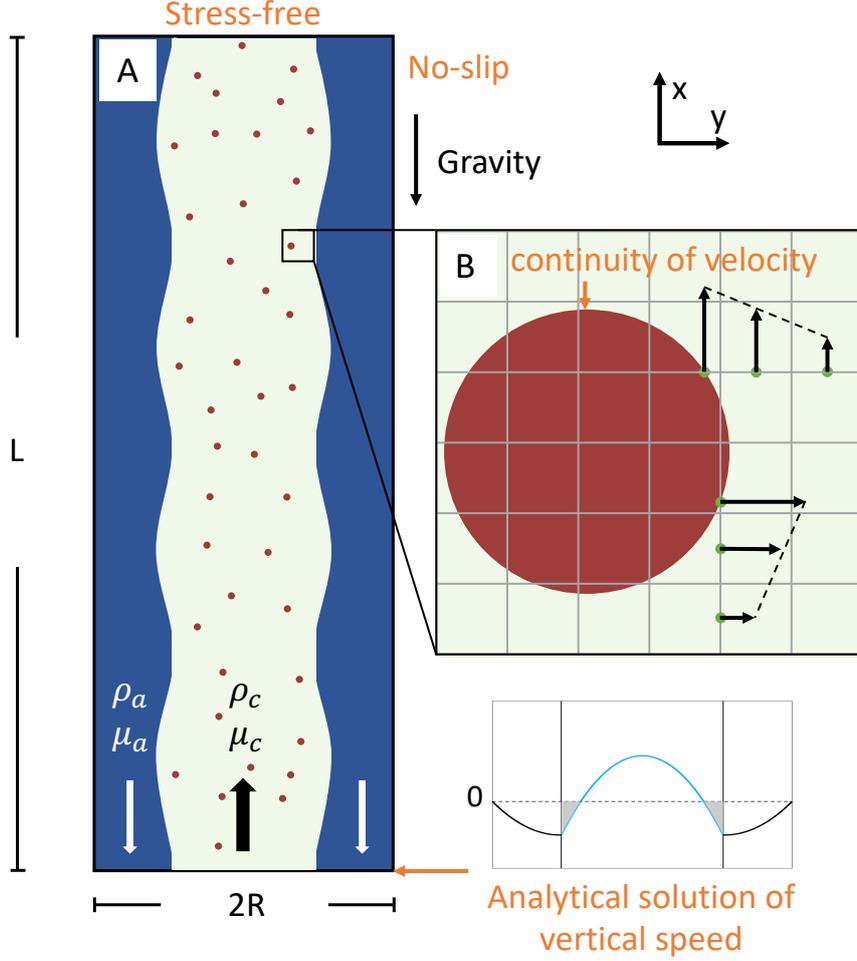


**Figure 1.** Overview of relevant spatial scales and their model representation.

165 use the term "magma" to refer to the mixture of melt and passively advected crystals  
 166 and bubbles. Resolving actively segregating bubbles while incorporating passively advecting  
 167 bubbles and crystals through a subgrid mixture model is commonly used in multi-  
 168 phase modeling as reviewed in Tryggvason et al. (2013).

169 The first step of our analysis is to quantify magma mixing via the multiscale conduit-  
 170 flow model (fig. 2). The multiscale approach described above reduces our model to three  
 171 distinct phases, the volatile-rich, up-welling magma, the volatile-poor, down-welling magma  
 172 and gas bubbles of intermediate size, contained mostly in the up-welling flow. All model  
 173 variables and parameter choices are summarized in table 2. The viscosities for both mag-  
 174 mas in our model are informed by previously estimated ranges for Stromboli (Burton,  
 175 Mader, & Polacci, 2007) and Mount Erebus (Sweeney et al., 2008). Since our model fo-  
 176 cuses on approximately steady flow during non-eruptive phases, we do not consider the  
 177 potential presence of large bubbles or slugs, because these are related to eruptive pro-  
 178 cesses (Jaupart & Vergnolle, 1988; Del Bello et al., 2012; Qin et al., 2018). Since our  
 179 bubbles are not large enough to deform significantly, we model them as spherical in the  
 180 interest of simplicity.

181 We define a 2D rectangular simulation domain (fig. 2A) to represent a conduit seg-  
 182 ment (fig. 1C). We apply a stress-free condition ( $p = const.$ ,  $\frac{\partial \mathbf{v}}{\partial x} = 0$ ) at the top bound-  
 183 ary to enable free outflow. At the base we impose the analytical solution of vertical speed  
 184 in core-annular flow (Suckale et al., 2018). The side walls are no-slip. We assume that  
 185 the two magmas are miscible Newtonian fluids differing in density and viscosity. The volatile-  
 186 rich magma has lower density because the entrained small bubbles reduce the effective  
 187 density of magma (fig. 1). The volatile-rich magma is less viscous by 1 to 2 orders of mag-  
 188 nitude because it contains higher concentration of dissolved  $H_2O$  and lower amount of  
 189 crystals (e.g., McBirney & Murase, 1984; Giordano et al., 2008).



**Figure 2.** Illustration of the simulation domain (not to scale). The orange text represents the boundary conditions. **(A):** The model domain for simulating the conduit flow. In this study,  $L = 21\text{m}$  and  $R = 1.5\text{m}$ . **(B):** We enforce the continuity of velocity as the boundary condition at the bubble-magma interface by linearly interpolating the bubble velocity and magma velocity for magma grid cells adjacent to bubbles, see Qin and Suckale (2017) for details. Vertical and horizontal arrows represent vertical and horizontal velocity components, respectively. Figure (B) modified from Qin and Suckale (2017).

190 Our model solves for the mass and momentum balance in an incompressible core-  
 191 annular flow at low Reynolds number (Qin & Suckale, 2017; Suckale et al., 2018; Qin et  
 192 al., 2020). The governing equations are conservation of mass and momentum

193 
$$0 = -\nabla p + \nabla \cdot (\mu \nabla \mathbf{v}) + \rho \mathbf{g}, \tag{1}$$

194 and advection-diffusion equation for concentration to capture magma mixing

195 
$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = D \nabla^2 c, \tag{2}$$

196 where density,  $\rho$ , and viscosity,  $\mu$ , are defined as

$$197 \quad \rho = \begin{cases} \rho_a - c(\rho_a - \rho_c), & \text{in magma} \\ \rho_b, & \text{in bubbles} \end{cases}, \quad (3)$$

$$198 \quad \mu = \mu_a - c(\mu_a - \mu_c), \quad (4)$$

199  $p$  is pressure,  $\mathbf{v}$  is velocity, and  $g$  is the gravitational acceleration. We solve the flow field  
 200 on a Cartesian staggered grid with the finite difference method as described in detail by  
 201 Qin and Suckale (2017). The concentration variable,  $c$ , in eq. 2 represents the content  
 202 of dissolved volatile and subgrid bubbles and ranges from  $c \in [0, 1]$ . The diffusion co-  
 203 efficient  $D = 10^{-10} \text{m}^2/\text{s}$  refers to the diffusion of water in basaltic magma (Zhang &  
 204 Stolper, 1991; Witham, 2011a). Initially,  $c = 1$  in the volatile-rich magma and  $c = 0$   
 205 in the volatile-poor magma. For the purpose of analyzing the flow regime stability, we  
 206 define the contour of  $c = 0.5$  as the interface between the two magmas. We assume that  
 207 the density and viscosity of magma depend linearly on  $c$ , as shown in eqs. 3 and 4, where  
 208  $\rho_c, \rho_a, \mu_c, \mu_a$  are the density and viscosity of the volatile-rich and volatile-poor magmas,  
 209 respectively.

210 Following Qin and Suckale (2017), Qin et al. (2020), and Qin and Suckale (2020),  
 211 we describe intermediate-size bubbles by the Newton's Laws of Motion

$$212 \quad M_b \frac{d\mathbf{V}_b}{dt} = \mathbf{F}_b + M_b \mathbf{g}, \quad (5)$$

$$213 \quad \frac{d\mathbf{X}_b}{dt} = \mathbf{V}_b, \quad (6)$$

214 where  $M_b$  is the mass of a bubble,  $\mathbf{V}_b$  the bubble velocity,  $\mathbf{F}_b$  the hydrodynamic force  
 215 exerted onto the bubble by the surrounding magma, and  $\mathbf{X}_b$  the bubble location. As shown  
 216 in fig. 2B, we enforce continuity of velocity at the bubble-magma interface by linearly  
 217 interpolating the bubble velocity and magma velocity for magma grid cells adjacent to  
 218 bubbles.

219 The numerical implementation (Wei et al., 2021) consists of three steps. The first  
 220 step is solving eq. 1. In this step, we modify the numerical implementation of Qin and  
 221 Suckale (2017), Qin et al. (2020), and Qin and Suckale (2020) by using the actual den-  
 222 sity of each phase to reduce the convergence steps. These previous studies use liquid den-  
 223 sity for the entire domain in the scenario where different phases have similar densities,  
 224 which is inconsistent with this study. The second step is solving eq. 2 following Suckale  
 225 et al. (2018). The third step is solving bubble motion following Qin and Suckale (2017),  
 226 Qin et al. (2020), and Qin and Suckale (2020).

227 In our model setup, magma mixing occurs at the interface (fig. 2A) between volatile-  
 228 rich and volatile-poor magma. Previous studies demonstrate that in the absence of small  
 229 bubbles or crystals in the flow, the interface is stable for two miscible magmas with low  
 230 diffusivity,  $D$  (Stevenson & Blake, 1998; Suckale et al., 2018). The presence of bubbles  
 231 and crystals, however, might lead to significantly more mixing than observed in the purely  
 232 fluid limit, because interactions between both bubbles and crystals act over a very long  
 233 spatial range at low Reynolds number (Segre et al., 1997). Even at very low phase frac-  
 234 tions of a few percent of solids or bubbles in the flow, multiphase interactions create spa-  
 235 tial correlations in velocity that are reminiscent of turbulence at high Reynolds number  
 236 (Xue et al., 1992; Tong & Ackerson, 1998; Levine et al., 1998). In volcanic systems, mix-  
 237 ing is hence dominated by multiphase processes rather than turbulence. In that aspect,  
 238 our model differs from Witham (2011a), who assumed turbulent mixing.

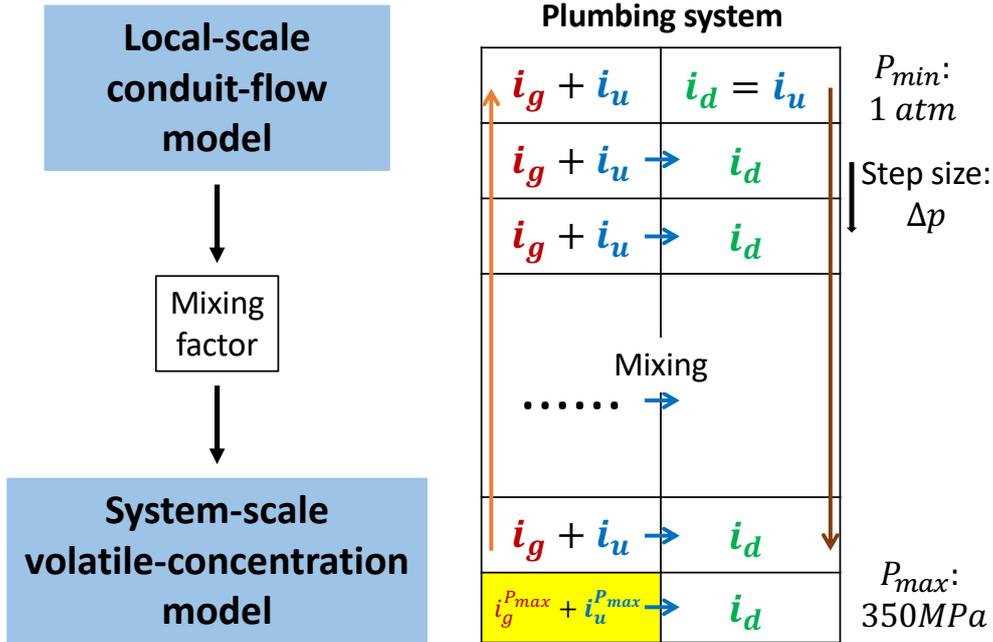
239 To quantify the magma mixing that occurs at the scale of a conduit segment, we  
 240 define the mixing factor  $\Gamma$  as the mixing associated with a pressure drop of  $\Delta p = 1 \text{MPa}$ .  
 241 We calculate  $\Gamma$  from the concentration in the magma entering the domain from the bot-  
 242 tom ( $c_b$ ) and leaving the domain from the top ( $c_t$ ) by averaging  $c$  in the up-welling magma

laterally. We use the median value of  $\frac{c_b - c_t}{c_b}$  over time as the estimated amount of mixing after the up-welling magma moves through the domain. The pressure drop in this process is  $\frac{L(\rho_a + \rho_c)g}{2}$ . For each conduit flow simulation, we compute  $\Gamma$  and its associated error  $\sigma$  as

$$\left\{ \begin{array}{l} \Gamma \\ \sigma \end{array} \right\} = 1 - \left[ 1 - \left\{ \begin{array}{l} median \\ std \end{array} \right\} \left( \frac{c_b - c_t}{c_b} \right) \right]^{\frac{2\Delta p}{L(\rho_a + \rho_c)g}}. \quad (7)$$

## 2.2 Volatile-concentration Model

As a consequence of mixing, the up-welling magma is gradually diluted as it ascends, while the down-welling magma becomes more volatile-rich as it descends. Using the estimated mixing factors from our simulations, we compute CO<sub>2</sub>-H<sub>2</sub>O concentration profiles at a system scale following Witham (2011a) with some modifications (Wei et al., 2021). For both CO<sub>2</sub> and H<sub>2</sub>O, we calculate the steady-state concentration profiles  $i_u$ ,  $i_d$  and  $i_g$  that represent the weight percent of dissolved volatiles in the up-welling magma, dissolved volatiles in the down-welling magma, and exsolved, up-welling volatiles, respectively. Although some bubbles enter the down-welling magma in our simulations, most of these bubbles return to the up-welling magma relatively quickly or continue ascending in down-welling magma because of their own buoyancy (fig. 4G), introducing only a minor and transient disruption. Therefore, we assume that no exsolved volatiles descend.



**Figure 3.** Left: workflow of our analysis. We summarize the simulation result of the conduit-flow model as a mixing factor, which is an input parameter for the system-scale volatile-concentration model. Right: Illustration of the volatile-concentration model. The yellow cell represents the fixed input set as the composition of the most volatile-rich melt inclusions.

We illustrate the calculation of CO<sub>2</sub>-H<sub>2</sub>O concentration profiles in fig. 3. The pressure  $P$  ranges from  $P_{min} = 0.1$  MPa to  $P_{max}$  with a step size  $\Delta p$ . We set  $i_u^{P_{max}} + i_g^{P_{max}}$  as the composition of the most volatile-rich melt inclusions (Métrich et al., 2010; Op-

penheimer et al., 2011), and set  $P_{max} = 350\text{MPa}$  based on the volatile solubility model MagmaSat (Gualda et al., 2012; Ghiorso & Gualda, 2015). We assume a constant magma temperature  $1180^\circ\text{C}$  and  $1000^\circ\text{C}$  for Stromboli (Bertagnini et al., 2003; Métrich et al., 2010) and Mount Erebus (Kyle, 1977), respectively.

Following Witham (2011a), we initialize  $i_u$  and  $i_g$  according to closed-system degassing. Then, we initialize the down-welling concentration profile by calculating

$$i_d^P = (1 - \Gamma) i_d^{P-\Delta p} + \Gamma i_u^P \quad (8)$$

for the entire pressure range. The superscripts indicate the pressure corresponding to the concentrations. We assume  $i_d^{P_{min}} = i_u^{P_{min}}$ , because up-welling magma starts to sink at the surface. Witham (2011a) defines the effective up-welling concentration  $i_*^P$  as

$$\phi_u i_*^P = \phi_u (i_u^P + i_g^P) - \phi_d i_d^P, \quad (9)$$

where  $\phi_u$  and  $\phi_d$  are the up-welling and down-welling mass flux, respectively. We assume negligible magma extrusion and approximately steady degassing such that  $\phi_u = \phi_d$  and  $i_*$  is constant throughout the domain, yielding

$$i_* = i_u^P + i_g^P - i_d^P. \quad (10)$$

Once  $i_*$  is known, we can compute  $i_u + i_g$  at each depth using eq. 10 and  $i_d$ . We then update  $i_u$  and  $i_g$  by partitioning  $i_u^P$  and  $i_g^P$  for the entire pressure range using MagmaSat (Gualda et al., 2012; Ghiorso & Gualda, 2015).

To compute  $i_*$ , we fix  $i_u^{P_{max}} + i_g^{P_{max}}$ , rather than fix  $i_g^{P_{min}}$  as Witham (2011a) does. We can also vary  $i_u^{P_{max}} + i_g^{P_{max}}$  to test the effect of variable volatile influx. We make this adjustment because current measurements only constrain surface gas flux (Burton, Allard, et al., 2007; Oppenheimer et al., 2009). Using surface gas flux to compute  $i_g^{P_{min}}$  requires the knowledge of  $\phi_u$ , which is unavailable from data. We hence compute  $i_*$  by

$$i_* = i_u^{P_{max}} + i_g^{P_{max}} - i_d^{P_{max}}. \quad (11)$$

After updating  $i_u$  and  $i_g$ , we iterate eqs.(8), (11), and (10) until reaching a steady state.

### 3 Results

#### 3.1 Bubbles Can Lead to Substantial Magma Mixing in Volcanic Conduits

To understand how intermediate-size gas bubbles create magma mixing during bidirectional conduit flow, we perform a series of simulations summarized in Table 2 with selected snapshots shown in fig. 4. We find that bubble speed (figs. 4A-C), the viscosities of both magmas (figs. 4A and D), and the volume fraction of resolved bubbles control the stability of the flow regime. The resolved bubbles, together with the subgrid bubbles contributing to the density difference between the volatile-rich and volatile-poor magma, correspond to total bubble fractions ranged from 2.1% to 12.0% in our simulations (table 2). To account for the different flow speeds in the simulations, we compare them at the same non-dimensional time  $t$ . We use  $R$  as the characteristic length and the vertical speed at the center line,  $v_c$ , of the analytical solution enforced at the bottom boundary as the characteristic speed in our nondimensionalization (Suckale et al., 2018).

For constant magma properties, bubble speed depends on both bubble radius and bubble density. Using fig. 4A as the baseline, we reduce the bubble size by 30% in simulation No. 2 shown in fig. 4B and reduce the density contrast between bubble and upwelling magma by 49% in simulation No. 3 shown in fig. 4C. All other parameters are constant. We select these particular values, including the unrealistically high bubble density in simulation No. 3, to keep the analytical bubble rise speed  $v_b = (\rho_c - \rho_b)gr^2/\mu_c$

**Table 2.** Values of variables in simulations.

Simulation No.	$\rho_c(\text{kg}/\text{m}^3)$	$\rho_a(\text{kg}/\text{m}^3)$	$\rho_b(\text{kg}/\text{m}^3)$	$\mu_c(\text{Pa}\cdot\text{s})$	$\mu_a(\text{Pa}\cdot\text{s})$	$r(\text{m})$	$\phi$	$\phi_{tot}$	$\mathcal{S}$	$\mathcal{I}$	$\Gamma(\%/MPa)$	$\sigma(\%/MPa)$
1	2400	2500	600	$3 \times 10^4$	$9 \times 10^4$	$4.3 \times 10^{-2}$	2%	7.16%	$4.05 \times 10^{-1}$	$7.36 \times 10^{-8}$	10.16	19.09
2	2400	2500	600	$3 \times 10^4$	$9 \times 10^4$	$3 \times 10^{-2}$	2%	7.16%	$2.83 \times 10^{-1}$	$7.36 \times 10^{-8}$	2.97	2.97
3	2400	2500	1518	$3 \times 10^4$	$9 \times 10^4$	$4.3 \times 10^{-2}$	2%	11.98%	$2.29 \times 10^{-1}$	$5.51 \times 10^{-8}$	2.93	3.90
4	2400	2500	600	$1.85 \times 10^4$	$5.55 \times 10^4$	$4.3 \times 10^{-2}$	2%	7.16%	$4.05 \times 10^{-1}$	$1.94 \times 10^{-7}$	22.55	38.07
5	2400	2500	76.21	$5 \times 10^4$	$1.5 \times 10^5$	$4.3 \times 10^{-2}$	2%	6.04%	$4.85 \times 10^{-1}$	$3.07 \times 10^{-8}$	5.57	3.43
6	2450	2452	300	$6 \times 10^3$	$1.8 \times 10^4$	$4.3 \times 10^{-2}$	2%	2.09%	$1.46 \times 10^0$	$2.01 \times 10^{-7}$	82.07	23.07
7	2400	2500	600	$2 \times 10^4$	$2 \times 10^5$	$4.3 \times 10^{-2}$	2%	7.16%	$7.90 \times 10^{-1}$	$3.33 \times 10^{-8}$	3.34	5.32
8	2400	2500	600	$9 \times 10^3$	$9 \times 10^4$	$4.3 \times 10^4$	2%	7.16%	$7.90 \times 10^{-1}$	$1.65 \times 10^{-7}$	13.00	13.26
9	2400	2500	600	$9 \times 10^3$	$9 \times 10^4$	$6 \times 10^{-2}$	2%	7.16%	$1.11 \times 10^0$	$1.65 \times 10^{-7}$	34.41	26.79
10	2450	2452	300	$5 \times 10^3$	$5 \times 10^4$	$4.3 \times 10^{-2}$	2%	2.09%	$2.85 \times 10^0$	$5.84 \times 10^{-8}$	84.34	28.8
12	2400	2500	600	$5 \times 10^3$	$5 \times 10^5$	$6 \times 10^{-2}$	2%	7.16%	$7.77 \times 10^0$	$9.55 \times 10^{-9}$	3.26	30.67
11	2400	2500	600	$2 \times 10^4$	$1 \times 10^6$	$4.3 \times 10^{-2}$	2%	7.16%	$2.91 \times 10^0$	$2.21 \times 10^{-9}$	0.54	3.15
13	2400	2500	600	$3 \times 10^4$	$9 \times 10^4$	$4.3 \times 10^{-2}$	1%	6.21%	$2.33 \times 10^{-1}$	$2.77 \times 10^{-8}$	3.57	7.41
14	2400	2500	600	$3 \times 10^4$	$9 \times 10^4$	$4.3 \times 10^{-2}$	3%	8.11%	$5.36 \times 10^{-1}$	$1.42 \times 10^{-7}$	19.31	27.87
15	2400	2500	600	$9 \times 10^3$	$9 \times 10^4$	$6 \times 10^{-2}$	1.5%	6.68%	$8.88 \times 10^{-1}$	$1.08 \times 10^{-7}$	17.60	10.44
16	2400	2500	700	$5 \times 10^3$	$2.5 \times 10^5$	$6 \times 10^{-2}$	0.5%	6.03%	$1.20 \times 10^0$	$5.61 \times 10^{-9}$	0.17	0.70
17	2400	2500	300	$3 \times 10^4$	$3 \times 10^5$	$4.3 \times 10^{-2}$	1%	5.50%	$5.18 \times 10^{-1}$	$5.87 \times 10^{-9}$	0.06	0.07
18	2520	2550	100	$5 \times 10^3$	$1 \times 10^6$	$6 \times 10^{-2}$	1.3%	2.80%	$3.58 \times 10^0$	$2.37 \times 10^{-8}$	57.88	42.80
19	2450	2550	300	$8 \times 10^3$	$1.6 \times 10^5$	$6 \times 10^{-2}$	1.5%	6.36%	$1.70 \times 10^0$	$4.94 \times 10^{-8}$	7.13	24.74
20	2366	2500	-	$5 \times 10^3$	$2.5 \times 10^5$	-	0%	7.44%	0	0	0.13	0.08
Example magma properties at deep and shallow conduit:												
Deep	2400	2500	700	$5 \times 10^3$	$2.5 \times 10^5$	$1 \times 10^{-2}$	0.5%	6.03%	$2.01 \times 10^{-1}$	$5.61 \times 10^{-9}$		
Shallow	2400	2500	100	$2 \times 10^4$	$1 \times 10^6$	$8 \times 10^{-2}$	3%	7.04%	$1.49 \times 10^0$	$2.02 \times 10^{-7}$		

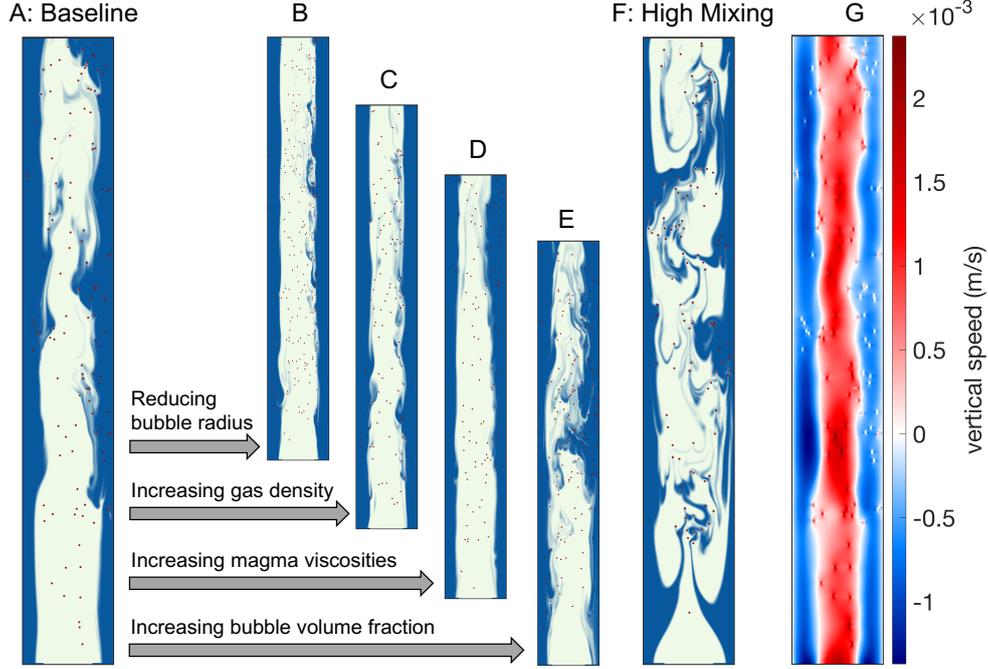
309 the same in both simulations, so that the bubble speed is approximately the same. Here  
310  $\rho_b$  is the bubble density,  $r$  the bubble radius, and  $\mu_c$  the viscosity of the volatile-rich magma.

311 Fig. 4A shows a flow field with significant mixing ( $\Gamma=10.16\%$ ). The oscillatory inter-  
312 face separating the two magmas entraps some of the volatile-poor magma into the volatile-  
313 rich magma. In contrast, both figs. 4B and C show a flow field with a much smaller and  
314 similar degree of mixing ( $\Gamma=2.97\%$  and  $2.93\%$ , respectively) and a stabler core-annular  
315 geometry. As compared to fig. 4A, the entrapment of volatile-poor magma into the volatile-  
316 rich magma is less frequent and entails smaller batches of magma.

317 Figs. 4A and D highlight the importance of both magma viscosities,  $\mu_c$  and  $\mu_a$  in  
318 governing mixing. With both viscosities equally increased by  $\frac{2}{3}$ , the flow field in fig. 4D  
319 becomes more stable and exhibits less mixing ( $\Gamma=5.57\%$ ) than in fig. 4A. In addition to  
320 increasing both magma viscosities, we decrease bubble density in fig. 4D to ensure that  
321 the bubble speed is the same in both simulations. We maintain a constant viscosity contrast  
322 between the magmas to isolate the effect of individual magma viscosities from that  
323 of a varying viscosity contrast, which also affects the bidirectional flow regime (Stevenson  
324 & Blake, 1998).

325 Figs. 4A and E highlight the importance of the volume fraction of centimeter-scale  
326 bubbles. With the resolved bubble volume fraction increased to 3%, the flow field in fig. 4E  
327 becomes less stable and exhibits more mixing ( $\Gamma=19.31\%$ ) than in fig. 4A. Comparing  
328 simulation No. 1 with 13 and 9 with 15 also demonstrates that decreasing the resolved  
329 bubble volume fraction decreases the degree of mixing (see table 2).

330 Fig. 4F illustrates the compound effect of increasing bubble speed and decreasing  
331 magma viscosities. In this simulation,  $v_b$  is 6 times higher than in fig. 4A and the magma  
332 viscosities are a fifth of those in fig. 4A. The consequence is extensive mixing and a com-  
333 plete collapse of core-annular flow. It may seem surprising that bubbles with radii much  
334 smaller than the conduit width can have such a profound effect on conduit flow at bub-  
335 ble fractions as low as 2%. To understand the physical mechanism, we quantify the stress  
336 disruptions created by bubbles stirring the bidirectional interface (fig. 5). Fig. 5A shows  
337 the interfacial stress deviation,  $\tau_{xy} - \tau$ , where  $\tau_{xy}$  is the simulated shear stress and  $\tau$   
338 is the analytical interfacial shear stress (Suckale et al., 2018). The interfacial stress de-  
339 viations lead to localized interface deformation, and, if pronounced enough, to interfa-  
340 cial wave build-up and mixing.



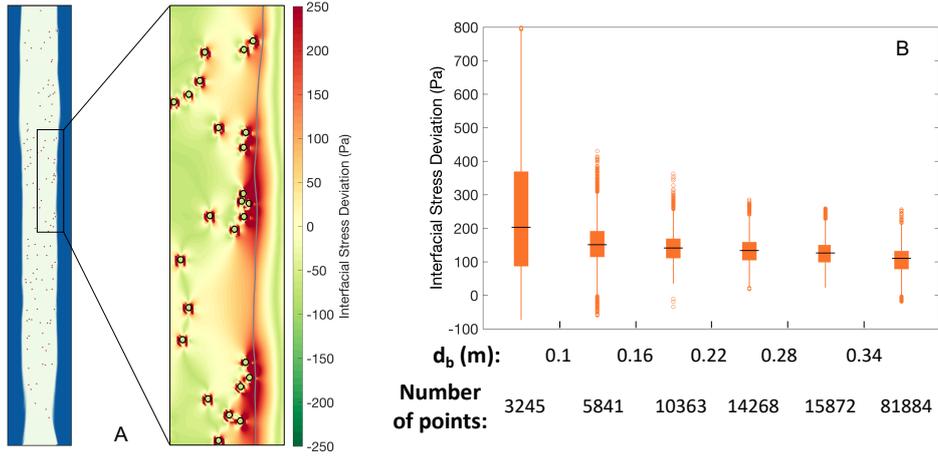
**Figure 4.** (A)-(E): Snapshots taken at nondimensional time  $t=40$  from simulations No. 1 (A), No. 2 (B), No. 3 (C), No. 5 (D), and No. 14 (E). (F): Snapshot taken at  $t=6.5$  from simulation No. 6, which has highest bubble speed and lowest magma viscosities among simulations in (A)-(F). (G): Corresponding vertical speed field of (A).

341 We conduct a statistical analysis (fig. 5B) of the simulation results in fig. 5A. Within  
 342 a period of time  $t \in [0, 15]$ , where the core-annular flow is stable, we sample points on  
 343 the interface. At each point, we compute the interfacial stress deviation and the distance  
 344 to the nearest bubble. We exclude bubble clusters from this analysis, because the hydrodynamic  
 345 stress field around a bubble cluster is dominated by the diverging interaction forces between  
 346 bubbles. Fig. 5B shows that the interfacial stress deviation increases as the distance to the  
 347 nearest bubble decreases, highlighting the significant stress deviation introduced at the interface  
 348 by nearby bubbles.

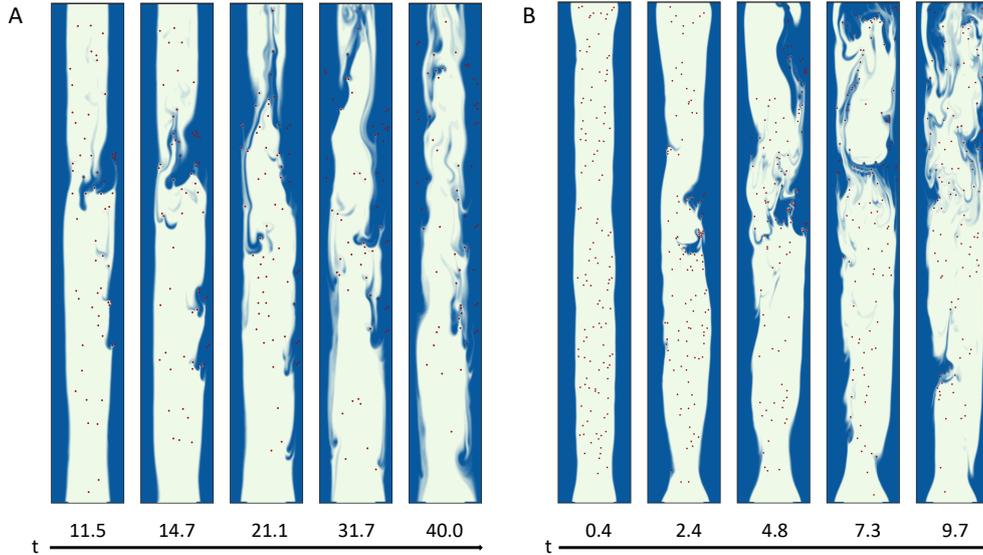
349 As shown in fig. 4F, the presence of bubbles can trigger the collapse of core-annular  
 350 flow. More specifically, we find two types of collapse in our simulations. Simulation No. 9  
 351 shown in fig. 6A demonstrates the type-1 collapse, where a large batch of down-welling,  
 352 degassed magma drips into the up-welling, volatile-rich magma, disrupting the initially  
 353 stable core-annular flow. The consequence is a significant amount of mixing, but the flow  
 354 field itself recovers eventually (fig. 6A). Simulation No. 10 shown in fig. 6B demonstrates  
 355 the type-2 collapse, where pronounced interfacial waves build up at the beginning of the  
 356 simulation and quickly lead to seemingly chaotic mixing. In this case, the disrupted core-  
 357 annular flow never recovers.

### 358 3.2 Generalizing Simulation Results through Nondimensional Analysis

360 To generalize our insights into the physical processes controlling mixing and flow-  
 361 regime stability in bubble-bearing core-annular flow to various depths within volcanic  
 362 systems (fig.7B), we identify two nondimensional numbers - the speed ratio  $\mathcal{S}$  and the  
 363 interface stability number  $\mathcal{I}$ . The speed ratio  $\mathcal{S}$  describes the effect of bubble speed by



**Figure 5.** (A): Interfacial stress deviation caused by bubbles (black circles) in the marked subregion of simulation No. 4 at  $t=6.2$ . The grey curve marks the interface ( $c=0.5$ ). (B): Statistical analysis of the relationship between the interfacial shear stress and the vicinity of bubbles for simulation No. 4. Each sample is a point on the interface at  $t \in [0, 15]$ .  $d_b$  is the distance between the sample point and its nearest bubble. The black line segments mark the median ( $q_2$ ) of each group. The bottom and top of the boxes mark the 25% ( $q_1$ ) and 75% ( $q_3$ ) quantiles, respectively. The whiskers mark the range  $[q_1 - 1.5 \times (q_3 - q_1), q_3 + 1.5 \times (q_3 - q_1)]$ . The circles mark the outliers.



**Figure 6.** Snapshots from simulations No. 9 (A) and No. 10 (B) showing the collapse of core-annular flow.

364 comparing  $v_b$  with  $v_c$ . The interface stability number  $\mathcal{I}$  captures the competition of the  
 365 interfacial shear stress and the magma viscosities. Both nondimensional numbers also  
 366 incorporate the number of bubbles in the domain. We emphasize that these two num-  
 367 bers are in addition to the more commonly used non-dimensional numbers that charac-  
 368 terize the force balance in the flow (e.g., Reynolds number), the bidirectional flow (e.g.,

369 Transport number), the domain geometry (e.g., the aspect ratio of the conduit), and the  
 370 material contrasts between the phases in the flow (e.g., the viscosity contrast).

371 To estimate the speed ratio and interface stability number, we dimensionalize the  
 372 non-dimensional, analytical solution of core-annular flow by Suckale et al. (2018). The  
 373 characteristic speed is

$$374 \quad U = (\rho_a - \rho_c)gR^2/\mu_a, \quad (12)$$

375 The interfacial shear stress is

$$376 \quad \tau = \mu_c \left( \frac{\partial v}{\partial y} \right)_{ndc} \frac{U}{R} = \mu_a \left( \frac{\partial v}{\partial y} \right)_{nda} \frac{U}{R}, \quad (13)$$

377 where  $\left( \frac{\partial v}{\partial y} \right)_{ndc}$  and  $\left( \frac{\partial v}{\partial y} \right)_{nda}$  is the nondimensional lateral component of the vertical speed  
 378 gradient at the volatile-rich and volatile-poor side of the interface, respectively.

379 We compute  $\mathcal{S}$  by

$$380 \quad \mathcal{S} = \frac{v_b \phi R}{v_c r}. \quad (14)$$

381 Here  $\frac{\phi R}{r}$  characterizes the frequency of bubble-interface interaction, which is controlled  
 382 by the density of bubbles in the domain and thus determined by the domain size ( $R$ ),  
 383 bubble volume fraction ( $\phi$ ) and bubble size ( $r$ ).

384 We compute  $\mathcal{I}$  by

$$385 \quad \mathcal{I} = \frac{\tau^2 \phi R}{\tau_v^2 r} = \frac{\tau^2 r \phi R}{\mu_c^2 g r} = \frac{\left[ \left( \frac{\partial v}{\partial y} \right)_{ndc} (\rho_a - \rho_c) \right]^2 \phi g R^3}{\mu_a^2}, \quad (15)$$

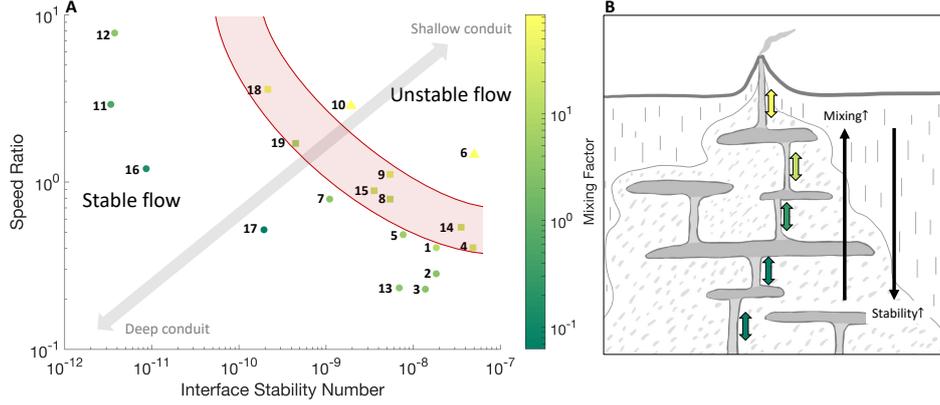
386 which represents the ratio of the interfacial shear stress and the viscous stress multiplied  
 387 with the frequency of bubble-interface interaction.

388 We summarize the effect of both non-dimensional numbers on mixing and the sta-  
 389 bility of the core-annular flow in Fig. 7A. Increasing  $\mathcal{S}$  and  $\mathcal{I}$  destabilizes the core-annular  
 390 flow and increases the degree of magma mixing. The decrease of magma mixing and the  
 391 change from the wavy to stable core-annular flow from simulation No. 1 (fig. 4A) to No. 2  
 392 and 3 (figs. 4B-C) are associated with the decrease of  $\mathcal{S}$  and  $\mathcal{I}$ . The decrease of magma  
 393 mixing from simulation No. 1 (fig. 4A) to No. 5 (fig. 4D) is consistent with the decrease  
 394 of  $\mathcal{I}$ . The increase of magma mixing from simulation No. 1 (fig. 4A) to No. 14 (fig. 4E)  
 395 is consistent with the increase of  $\mathcal{S}$  and  $\mathcal{I}$ . Among simulations shown in fig. 4, simula-  
 396 tion No. 6 (fig. 4F) has the largest  $\mathcal{S}$  and  $\mathcal{I}$  and shows the highest degree of mixing and  
 397 the most unstable flow regime. The transition zone in fig. 7A shows that the type-1 un-  
 398 stable flow is a transitional scenario between the stable core-annular flow and the type-  
 399 2 unstable flow that collapses quickly and irreversibly.

400 The magma properties listed in table 2 demonstrate that shallower depth corre-  
 401 sponds to larger  $\mathcal{S}$  and  $\mathcal{I}$  (fig. 7). As magma ascends, the conduit flow transitions from  
 402 stable core-annular flow with relatively low mixing to unstable flow with high mixing.  
 403 The parameters for shallow magma are similar to simulations showing high mixing and  
 404 unstable flow regime (simulation No. 6 and 10). On the other hand, with limited volatile  
 405 exsolution, all bubbles in deep magma are likely subgrid-scale. Therefore, we run sim-  
 406 ulation No. 20 where we only simulate the two liquid phases to represent the flow in  
 407 the bottom left region of fig. 7A. The model produces a completely stable core-annular  
 408 flow regime with low mixing only generated by diffusion, suggesting that bottom left re-  
 409 gion of fig. 7A corresponds to low mixing and stable core-annular flow regime.

### 410 3.3 Magma Mixing Alters H<sub>2</sub>O-CO<sub>2</sub> Concentration Profiles

411 We test the effect of different mixing factors,  $\Gamma$ , and varying CO<sub>2</sub> influx on the H<sub>2</sub>O  
 412 and CO<sub>2</sub> concentrations in melt inclusions by computing the concentration profiles for



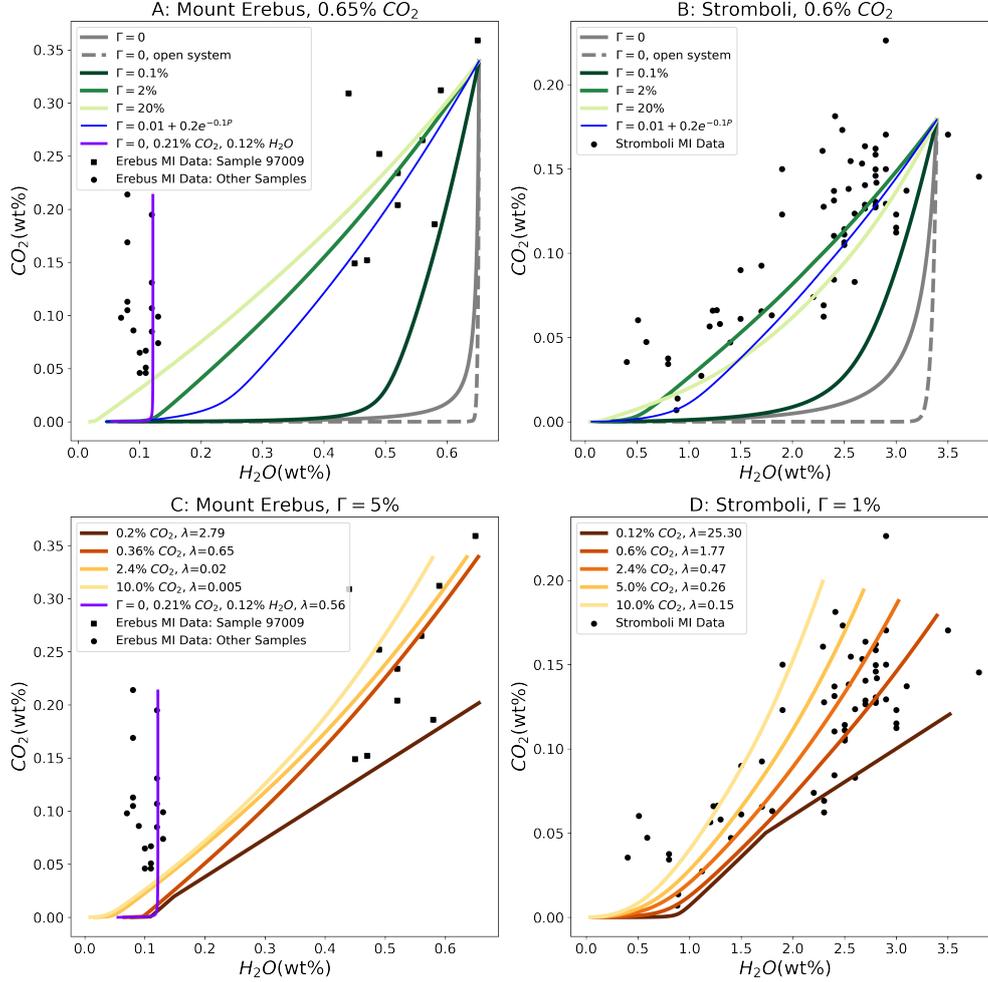
**Figure 7.** (A): Regime diagram for the stability of core-annular flow. The numbers identify individual simulations and the color scale represents the mixing factor  $\Gamma$ . Round, square and triangle markers highlight stable, type-1 unstable, and type-2 unstable core-annular flow during  $t \in [0, 90]$ , respectively. The inferred red transition zone covering the type-1 unstable flow separates the stable and unstable flow. (B): Our model indicates higher degree of mixing and less stable flow regime in the conduits towards shallower depth.

413 Stromboli and Mount Erebus. To compare the two processes, we conduct two suites of  
 414 calculations for each volcano. In each group, we fix one process and vary the other one  
 415 to test whether the two processes have distinct observational signatures. In both cases,  
 416 we fix the total amount of  $\text{H}_2\text{O}$  as the concentrations in the most volatile-rich melt in-  
 417 clusions, because at  $P_{max}$   $\text{H}_2\text{O}$  is unsaturated according to MagmaSat (Gualda et al.,  
 418 2012; Ghiorso & Gualda, 2015).

419 Fig. 8 compares the computed volatile concentration profiles to existing data (Métrich  
 420 et al., 2010; Oppenheimer et al., 2011). Even small degrees of mixing ( $\Gamma < 5\%$ ) sensitively  
 421 affect the functional relationship between the  $\text{CO}_2$  and the  $\text{H}_2\text{O}$  (figs. 8A-B). Increas-  
 422 ing mixing shifts the concentration profiles towards higher  $\text{CO}_2$  and lower  $\text{H}_2\text{O}$  concen-  
 423 tration relative to the closed-system profiles ( $\Gamma = 0$ ). However, the profiles quickly be-  
 424 come insensitive to further mixing as shown by the profiles with  $\Gamma = 20\%$  in figs. 8A-B.  
 425 With  $\Gamma > 30\%$ , stable core-annular flow no longer exists in our simulations (fig. 7A). There-  
 426 fore, we only compute profiles with mixing factors below this limit. While we have evalu-  
 427 ated both constant and depth-variable mixing factors, both results are consistent with  
 428 data, suggesting that the data does not currently afford the resolution necessary to iden-  
 429 tify potential depth-variability in mixing (figs. 8A-B).

430 Accounting for magma mixing results in concentration profiles that are more consis-  
 431 tent with the Stromboli data and sample 97009 from Mount Erebus than open- or closed-  
 432 system degassing alone (figs. 8A-B). Samples other than 97009 from Mount Erebus match  
 433 a closed-system profile (black dots in fig. 8A) and clearly distinct sample 97009. How-  
 434 ever, we are unable to constrain the mixing factor through the melt inclusion data ex-  
 435 actly due to data scatter. When analyzing the effect of variable  $\text{CO}_2$  influx, we there-  
 436 fore only consider minimal mixing,  $\Gamma = 1\%$ . Figs. 8C and D show that for a fixed  $\Gamma = 1\%$ ,  
 437 varying  $\text{CO}_2$  influx also significantly alters the volatile concentration profiles and fur-  
 438 ther improves the fit between model and data. Increasing  $\text{CO}_2$  influx shifts the profiles  
 439 towards higher  $\text{CO}_2$  and lower  $\text{H}_2\text{O}$  concentration, especially at high pressures. This ef-  
 440 fect is distinct from the effect of magma mixing.

441 Varying  $\text{CO}_2$  influx also changes the ratio of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in the gas phase in our  
 442 calculations. In the legend of figs. 8C and D, we include the values of  $\lambda = \text{H}_2\text{O}/\text{CO}_2$  in  
 443 the gas phase at the surface in each calculation. According to the surface gas flux data,  
 444  $\lambda$  ranges from 0.82 to 2.49 and 0.56 to 0.79 at Stromboli and Mount Erebus, respectively  
 445 (Burton, Allard, et al., 2007; Oppenheimer et al., 2009).



**Figure 8.**  $\text{H}_2\text{O}$ - $\text{CO}_2$  concentration profiles with varied mixing factors (A and B) and total amount of  $\text{CO}_2$  (C and D). The blue curves in A and B represents profiles with mixing factors varied with pressure.

#### 446 4 Discussion

447 Analogue laboratory models illustrate the basic physical processes that govern bidi-  
 448 directional flow (Stevenson & Blake, 1998; Beckett et al., 2011), but are highly idealized  
 449 representations of actual volcanic systems. Conduit models can help bridge the gap (Suckale  
 450 et al., 2018; Fowler & Robinson, 2018), but are difficult to test against observational data.  
 451 The challenge arises because observational data, such as melt inclusion compositions and  
 452 surface gas flux (Métrich et al., 2001; Burton, Allard, et al., 2007; Oppenheimer et al.,  
 453 2009; Métrich et al., 2010; Oppenheimer et al., 2011; Ilanko et al., 2015; Rasmussen et  
 454 al., 2017), are the product of multi-scale processes while most existing conduit models

455 are single-scale and do not entail testable model predictions at the scale of individual  
456 bubbles or crystals.

457 In this study, we integrate numerical simulations of bidirectional conduit flow at  
458 the scale of individual bubbles with a system-scale calculation of H<sub>2</sub>O-CO<sub>2</sub> concentra-  
459 tion profiles. We analyze how the presence of bubbles affects the degree of magma mix-  
460 ing in a conduit segment (fig. 4). Previous experimental and numerical studies show that  
461 the viscosity contrast ( $\mu_c/\mu_a$ ) governs the stability of the flow regime (Stevenson & Blake,  
462 1998; Suckale et al., 2018). Here, we demonstrate that the properties of the gas phase  
463 are important, too. Bubbles with a sufficient rise speed can trigger significant mixing and  
464 even flow-regime collapse at viscosity contrasts that are stable in the absence of bubbles  
465 (Stevenson & Blake, 1998; Suckale et al., 2018).

466 While there is not doubt that viscosity contrast is important for the stability of core-  
467 annular flow as suggested by previous studies (Stevenson & Blake, 1998; Suckale et al.,  
468 2018), our results indicate that two nondimensional numbers,  $\mathcal{S}$  and  $\mathcal{I}$ , are valuable ad-  
469 ditions to consider. Simulations with the same viscosity contrast ( $\frac{\mu_a}{\mu_c} = 3$  for simula-  
470 tions No. 1-6, 13, 14,  $\frac{\mu_a}{\mu_c} = 10$  for simulations No. 7-10, 15, 17,  $\frac{\mu_a}{\mu_c} = 20$  for simula-  
471 tions No. 18-19) show significantly varied mixing and stability. This variance is well cap-  
472 tured by  $\mathcal{S}$  and  $\mathcal{I}$  (fig. 7).

473 We argue that bubbles locally increase the interfacial stress (fig. 5). This interfa-  
474 cial stress deviation disrupts the linearly unstable (Selvam et al., 2007; Martin et al., 2009;  
475 Selvam et al., 2009) but nonlinearly stable interface (Ullmann & Brauner, 2004; Suckale  
476 et al., 2018). In the absence of bubbles, linear growth of instability is suppressed by the  
477 nonlinear interaction between the growing interface wave and viscous damping in the two  
478 magmas (Ullmann & Brauner, 2004; Suckale et al., 2018). We show that the presence  
479 of bubbles introduces additional perturbations into this metastable flow configuration  
480 (e.g., fig. 4B) that can trigger wave breaking (e.g., fig. 4A) and mixing (e.g., fig. 4F).

481 The finding that bubbles with radii much smaller than the conduit width can have  
482 such a significant effect may appear surprising. However, flow-regime stability at the con-  
483 duit scale ultimately hinges on interface stability, which in turn hinges on the disrup-  
484 tions introduced by the bubbles. The relevant scale comparison is thus not between bub-  
485 ble radius and conduit width, but between bubble radius and the amplitude of the in-  
486 terfacial wave. So long as a well-defined interface exists, these scales are comparable (Ullmann  
487 & Brauner, 2004; Suckale et al., 2018). We emphasize that we only simulate magmas with  
488 low diffusivities here, similar to Stevenson and Blake (1998).

489 Our simulations suggest that some degree of mixing is almost inevitable in core-  
490 annular flow unless bubbles remain very small, which could occur particularly for very  
491 low H<sub>2</sub>O contents. Magma mixing tend to increase at shallow depth, potentially to the  
492 point of core-annular flow collapse (fig. 7). The reason is that the gas phase plays an in-  
493 creasingly important role in the system dynamics at decreasing depth below the surface,  
494 because of continued exsolution, bubble growth, and gas decompression (e.g., Gonner-  
495 mann & Manga, 2013).

496 If magma mixing is as common as our simulations suggest, it would be reflected  
497 in observational data. To test the compatibility of our model results with observations,  
498 we compute the H<sub>2</sub>O-CO<sub>2</sub> concentration profiles associated with different mixing fac-  
499 tors building on Witham (2011a). The fit between modeled and measured volatile con-  
500 centrations increases notably when accounting for magma mixing, even for low mixing  
501 factors (figs. 8A-B).

502 Figs. 8C-D show that varying CO<sub>2</sub> influx also improves the match between mod-  
503 eled and measured volatile concentrations, as also argued by previous studies (e.g., Bur-  
504 ton, Mader, & Polacci, 2007; Métrich et al., 2010; Rasmussen et al., 2017). Both Burton,  
505 Mader, and Polacci (2007) and Métrich et al. (2010) estimate that the amount of CO<sub>2</sub>

506 influx at Stromboli is 2.4%. In our simulations, this CO<sub>2</sub> influx results in a  $\lambda=0.47$  (H<sub>2</sub>O/CO<sub>2</sub>  
 507 in the gas phase at the surface) as shown in fig. 8D. Even with a low degree of mixing,  
 508 this resultant  $\lambda$  is outside the range 0.82-2.49 observed at Stromboli (Burton, Allard, et  
 509 al., 2007). Increased mixing further decreases  $\lambda$  due to more loss of H<sub>2</sub>O to the down-  
 510 welling magma. We argue here that when accounting for magma mixing, it is unneces-  
 511 sary to invoke a large amount of CO<sub>2</sub> for reproducing melt inclusion data (Métrich et  
 512 al., 2010; Rasmussen et al., 2017). Fig. 8D shows that a CO<sub>2</sub> influx of 0.6% results in  
 513 a  $\lambda=1.77$ , which is in the observed range (Burton, Allard, et al., 2007).

514 For Erebus, most samples are H<sub>2</sub>O-poor except sample 97009 (Oppenheimer et al.,  
 515 2011). Oppenheimer et al. (2011) propose that Mount Erebus is occasionally fed by volatile-  
 516 rich magma but continuously flushed by CO<sub>2</sub>-rich fluid. The resultant dry magma leads  
 517 to high magma viscosity and thus low mixing. This idea is compatible with our model  
 518 results: The purple curve in figs. 8A and C shows that the closed-system profile matches  
 519 the data. Assuming complete degassing of CO<sub>2</sub> and H<sub>2</sub>O, the calculated  $\lambda$  matches the  
 520 surface gas flux measurements. Sample 97009 may have formed shortly after the injec-  
 521 tion of volatile-rich magma, which decreases magma viscosity and increases mixing.

522 We emphasize that apart from magma mixing and variable CO<sub>2</sub> influx, several other  
 523 processes not considered in our study contribute to the pronounced scatter in melt in-  
 524 clusion data. These include uncertainties in measurements (Métrich & Wallace, 2008;  
 525 Métrich et al., 2010; Oppenheimer et al., 2011), disequilibrium degassing potentially gen-  
 526 erating CO<sub>2</sub>-oversaturated melt (Pichavant et al., 2013) and crystallization affecting volatile  
 527 solubility (Gualda et al., 2012; Ghiorso & Gualda, 2015). In addition, the complex ge-  
 528 ometry of some volcanic plumbing systems may introduce variability. At shallow depth,  
 529 some conduits flare out into lava lakes such as at Mount Erebus, altering both mixing  
 530 and surface gas flux (Oppenheimer et al., 2009). At deep depth, volcanic conduits are  
 531 thought to be connected to heterogeneous and largely crystalline transcrustal plumbing  
 532 systems (Cashman et al., 2017; Magee et al., 2018). Melt inclusions that form at con-  
 533 siderable depth (Métrich et al., 2001, 2010; Oppenheimer et al., 2011; Rasmussen et al.,  
 534 2017) might hence sample a different portion of the plumbing system and record pro-  
 535 cesses not considered here.

536 Despite these caveats, our analysis suggests that melt inclusions might offer the op-  
 537 portunity to constrain magma mixing in volcanic conduits and variations in CO<sub>2</sub> influx  
 538 over time. Both of these processes contribute to variability in the surface gas flux, which  
 539 is correlated with the eruptive cycles of persistently degassing volcanoes (Burton, Allard,  
 540 et al., 2007; Oppenheimer et al., 2009; Ilanko et al., 2015). Constraining their inherent  
 541 variability over multiple eruptive cycles hence has the potential for increasing the con-  
 542 straints we can bring to bear in conduit-flow models. We hence suggest that with im-  
 543 proved measurement accuracy and reduced uncertainty, disaggregating the scattered melt  
 544 inclusion data could help us track and better understand the evolving flow conditions  
 545 in volcanic conduits, as already attempted in Spilliaert et al. (2006) and Sides et al. (2014).

## 546 5 Conclusions

547 Observables such as melt inclusions provide important testimony on degassing pro-  
 548 cesses at persistently active volcanoes, but their testimony is rarely straight-forward to  
 549 interpret. Models such as bidirectional conduit flow, on the other hand, account for im-  
 550 portant physical processes, but are difficult to connect to and evaluate against observa-  
 551 tional data. This study contributes towards forging a closer link between a commonly  
 552 used and theoretically well-motivated conduit model for persistent degassing, core-annular  
 553 flow, and the volatile concentration observed in melt-inclusion data. We find that bub-  
 554 bles that are large enough to decouple from the ambient flow field and ascend individ-  
 555 ually can destabilize the bidirectional flow and can lead to significant mixing between  
 556 volatile-rich and volatile-poor magma. This finding suggests that magma mixing is com-

557 mon in core-annular flow in the conduits of persistently degassing volcanoes, but vari-  
 558 ations in CO<sub>2</sub> influx may occur simultaneously. Being able to identify the relative im-  
 559 portance of these two processes in observational data is valuable to track and better un-  
 560 derstand the evolving flow conditions in volcanic systems. Our study shows that while  
 561 both magma mixing and increasing CO<sub>2</sub> influx shifts the profiles towards higher CO<sub>2</sub>  
 562 and lower H<sub>2</sub>O concentration, the observational signature of increasing CO<sub>2</sub> influx is dis-  
 563 tinct from that of magma mixing by being most prominent at high pressures. Disaggre-  
 564 gating scattered melt inclusion data for different volcanic centers or eruptive episodes  
 565 may hence help to identify variability in degassing.

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### 572 Open Research

573 v1.0.7 of the code used for the conduit-flow model and the volatile-concentration model  
 574 is preserved at <https://doi.org/10.5281/zenodo.5090109> with open access. The us-  
 575 age instructions are provided in the README file of the repository.

### 576 Author contributions

577 Z.W. performed the numerical simulations, computed the concentration profiles, produced  
 578 the figures and wrote most of the text. Z.Q. developed the numerical technique. J.S. con-  
 579 ceptualized the study, advised Z.W. and contributed to the text.

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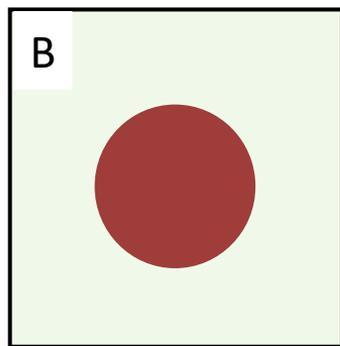
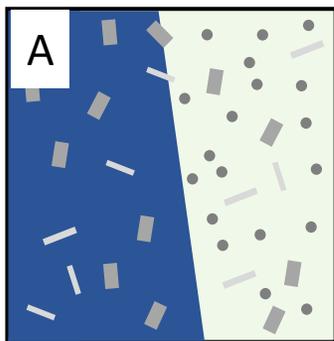
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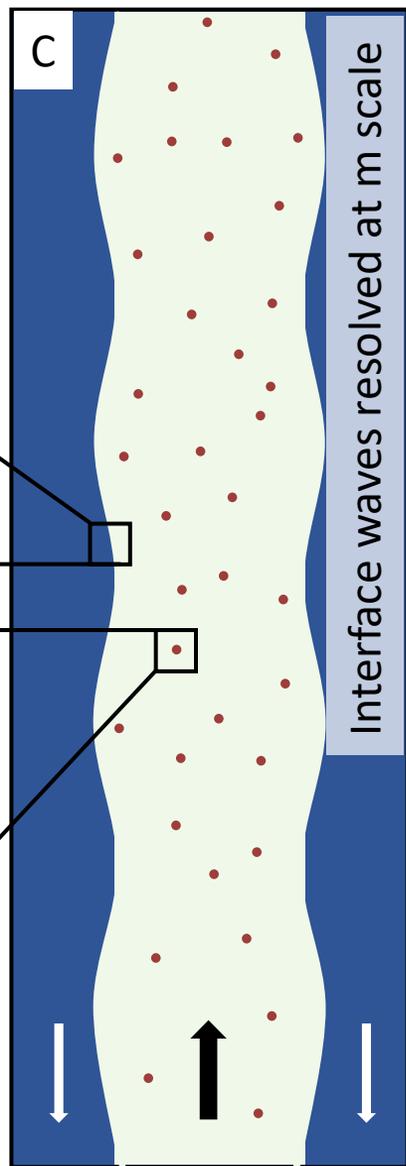
Figure 1.

**Passive advection scale:** Crystals and small bubbles integrated via a mixture approach

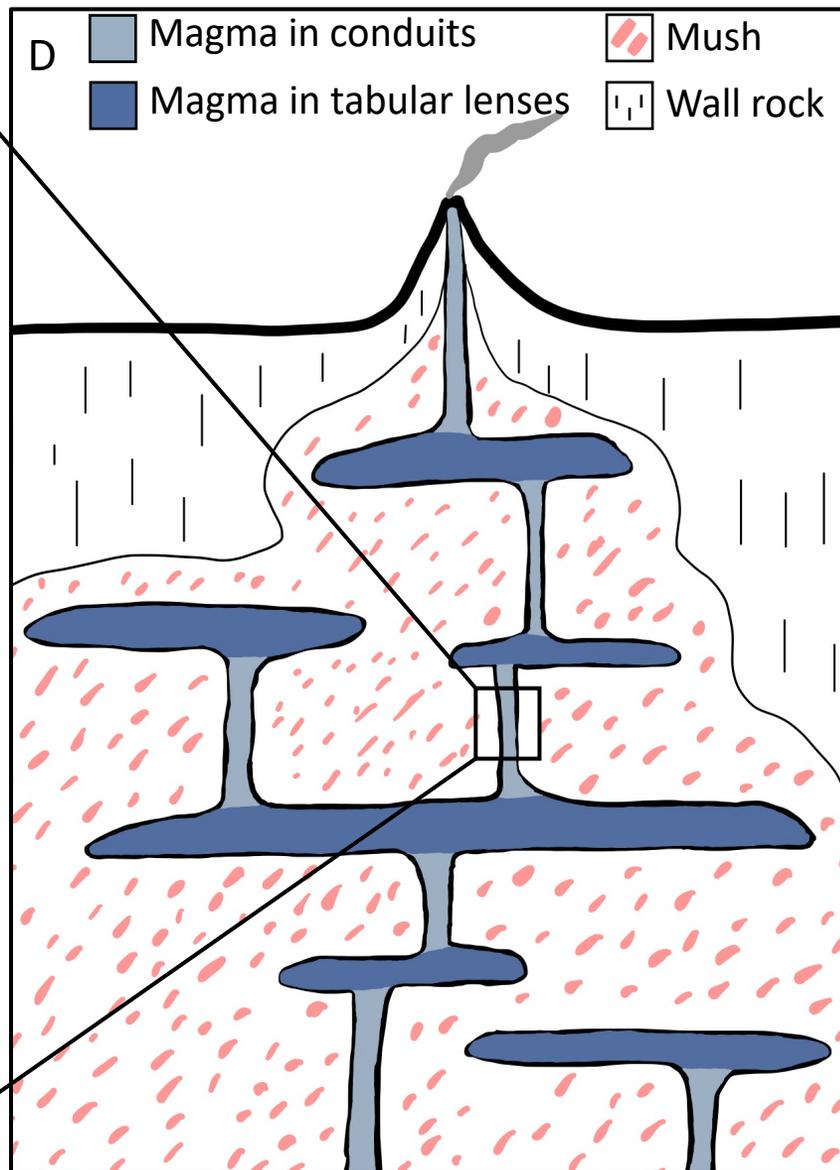


**Bubble segregation scale:** Bubbles fully resolved at the centimeter scale

**Conduit segment scale:** Conduit flow fully resolved at tens-of-meter scale



**System scale:** Insights about conduit flow generalized to various depths via nondimensional analysis



$10^{-3}$

$10^{-2}$

$10^1$

$10^4$

Scale (m)

Figure 2.

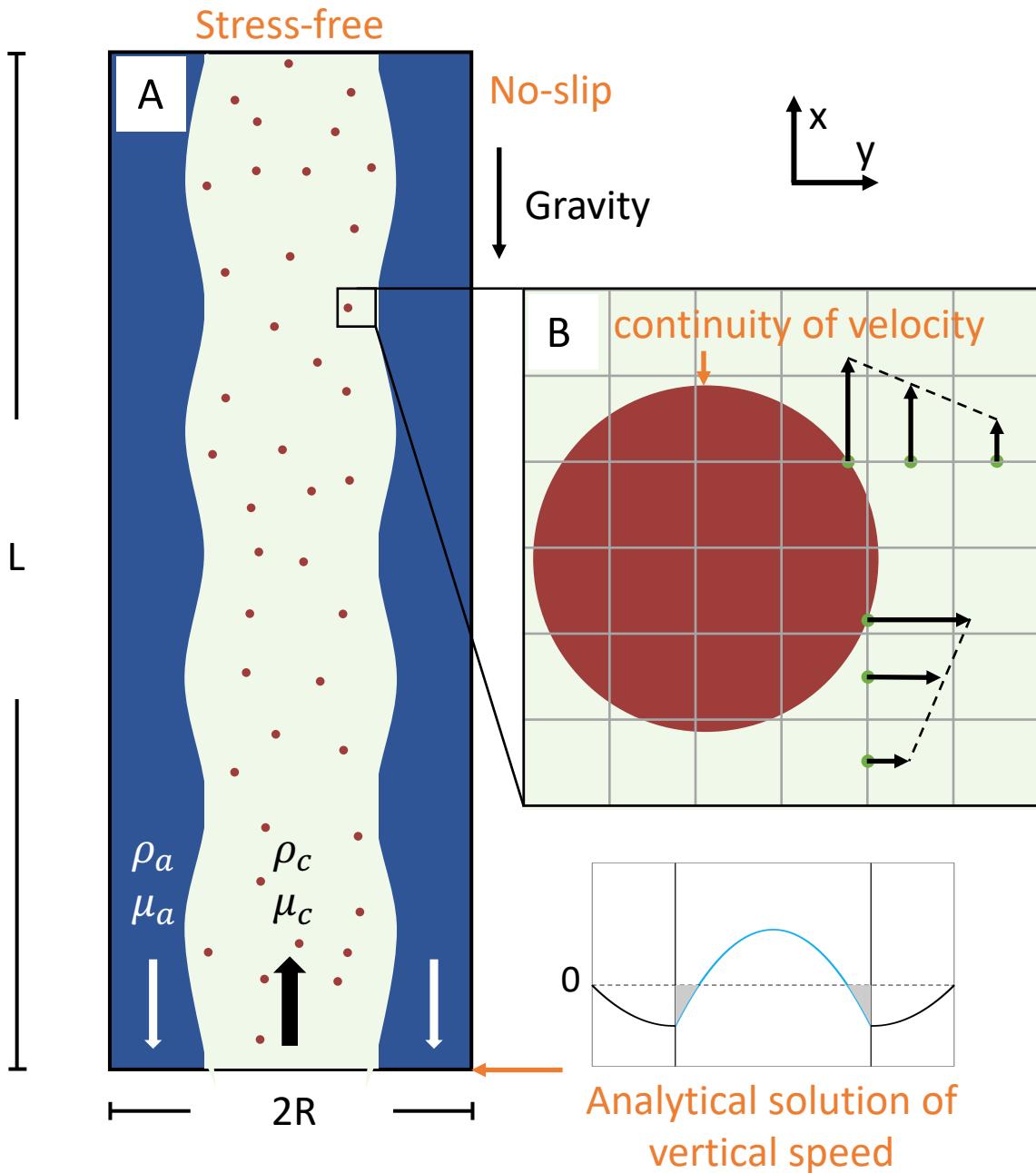


Figure 3.

Local-scale  
conduit-flow  
model

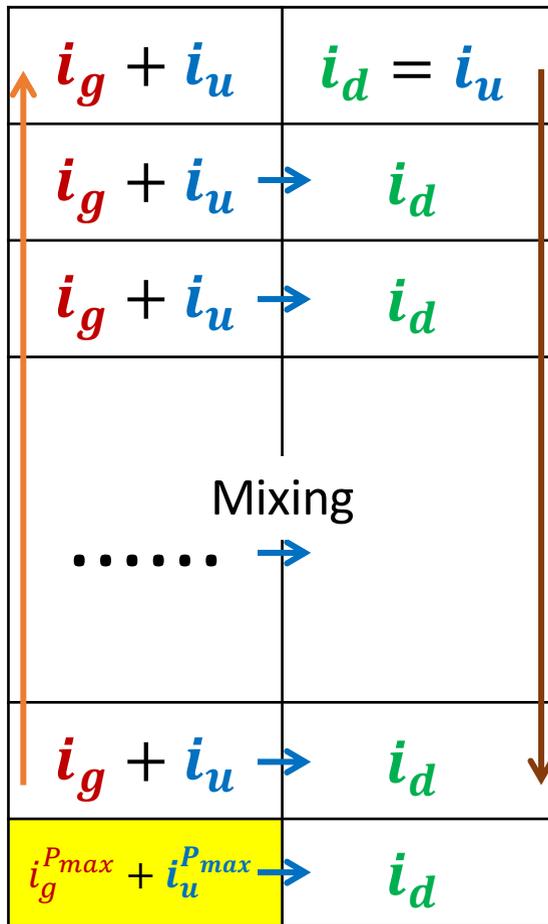


Mixing  
factor



System-scale  
volatile-concentration  
model

Plumbing system



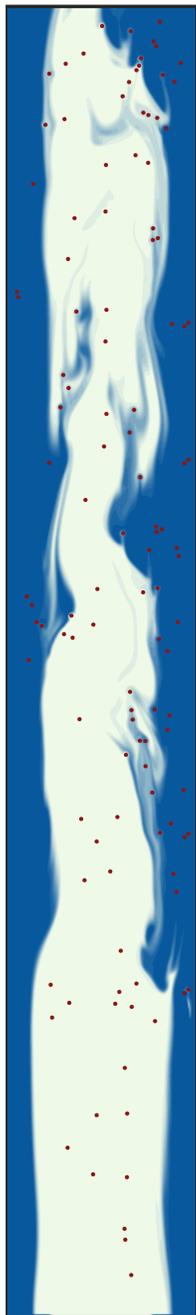
$P_{min}$ :  
1 atm

Step size:  
 $\Delta p$

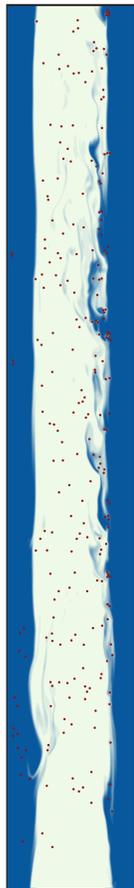
$P_{max}$ :  
350 MPa

**Figure 4.**

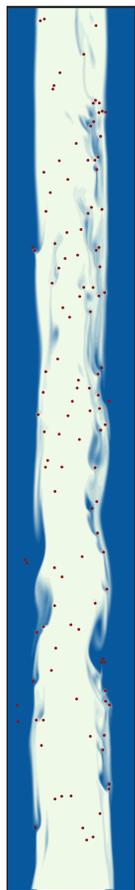
A: Baseline



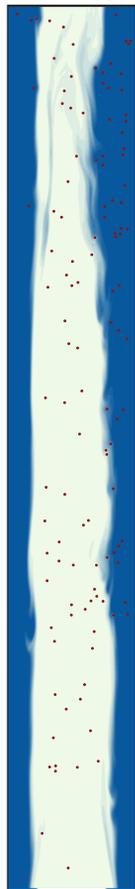
B



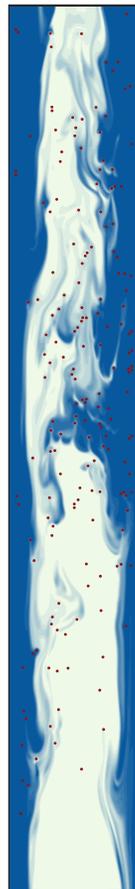
C



D



E



Reducing  
bubble radius



Increasing gas density



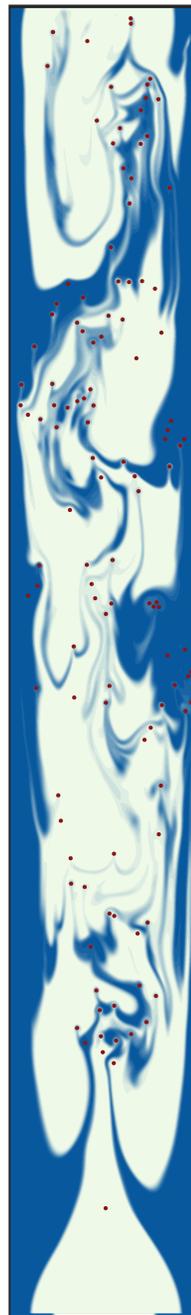
Increasing magma viscosities



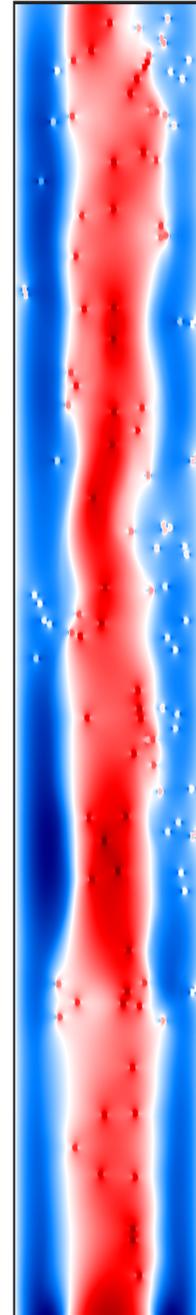
Increasing bubble volume fraction



F: High Mixing



G



$\times 10^{-3}$

2

1.5

1

0.5

0

-0.5

-1

vertical speed (m/s)

Figure 5.

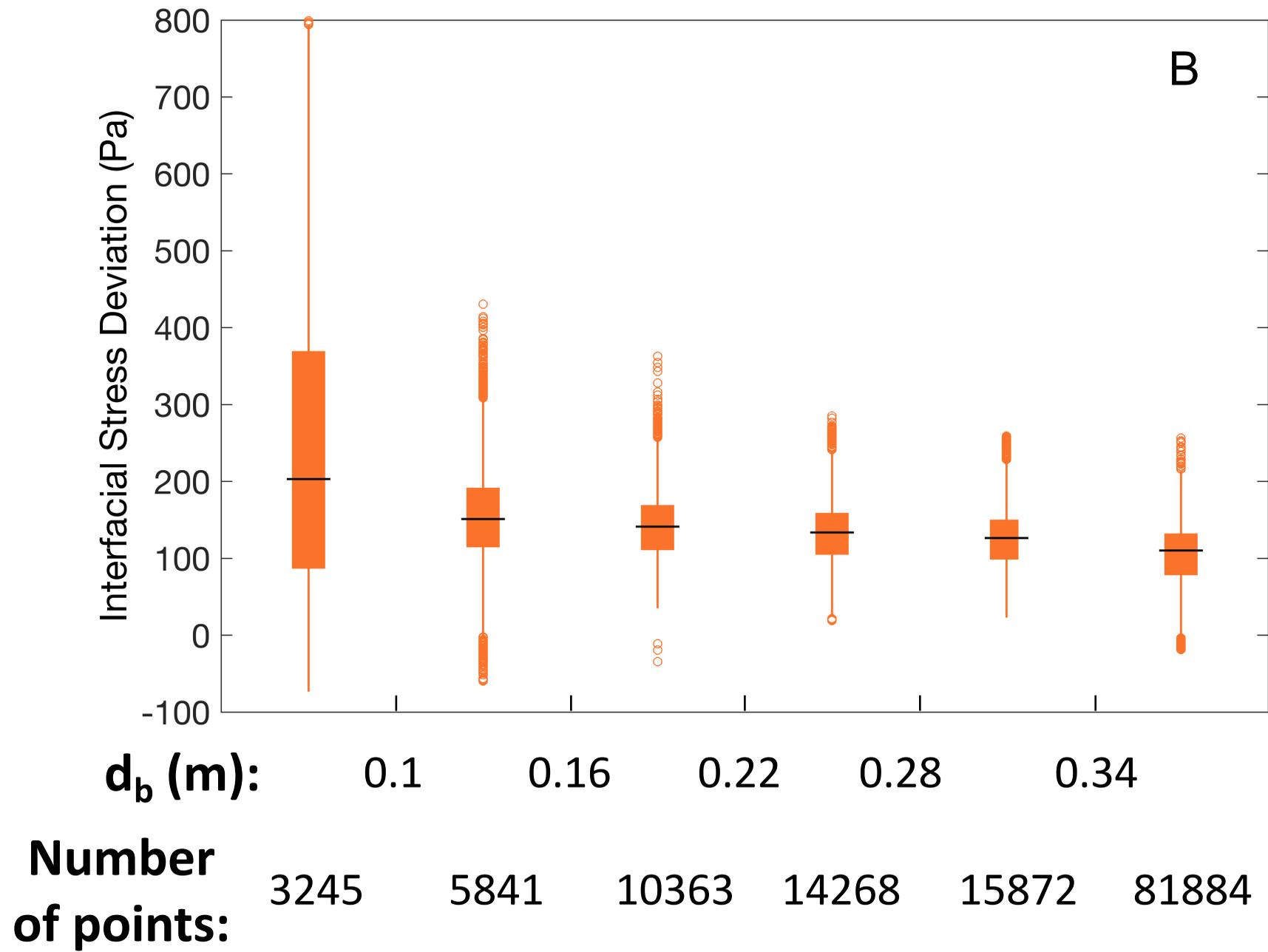
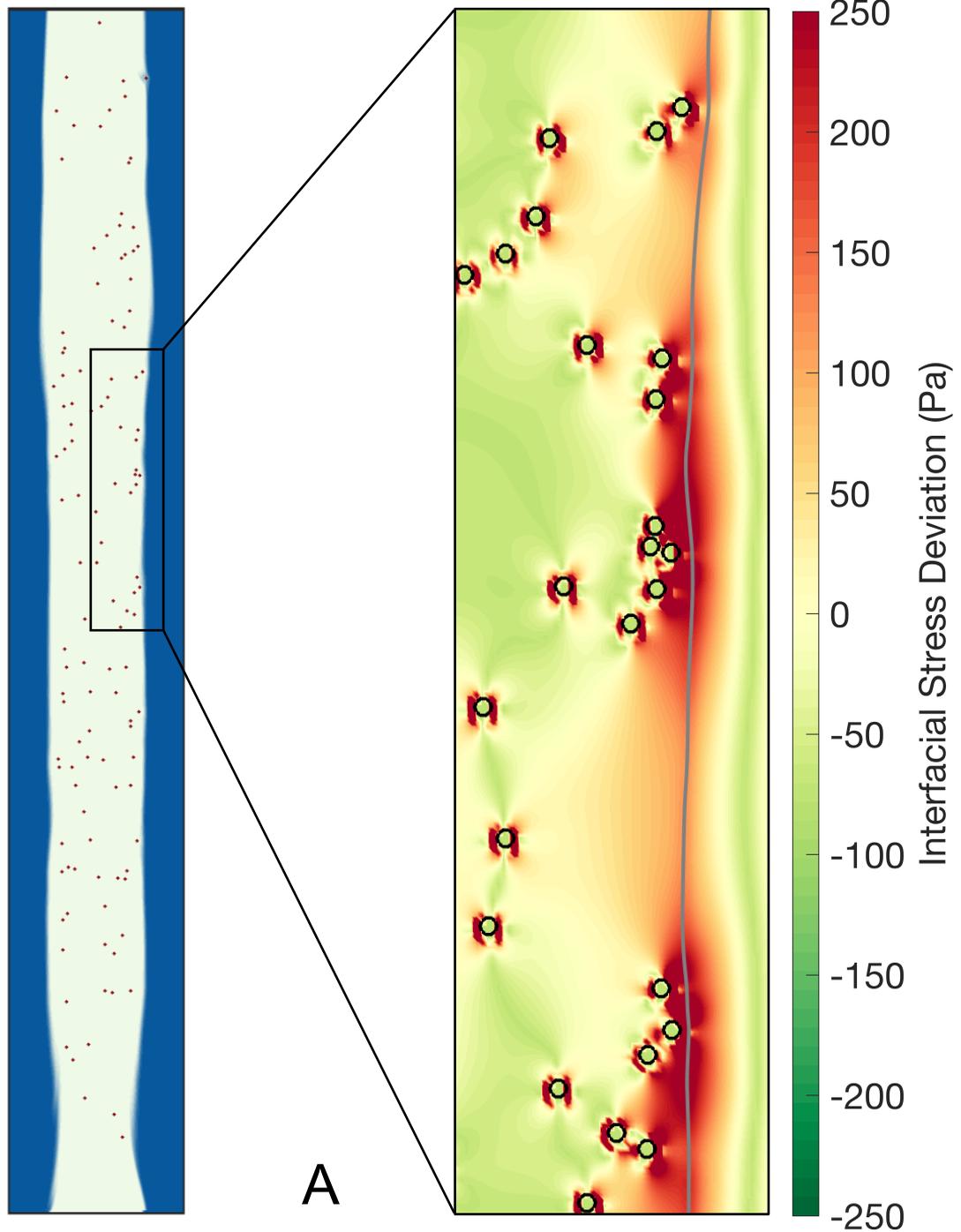
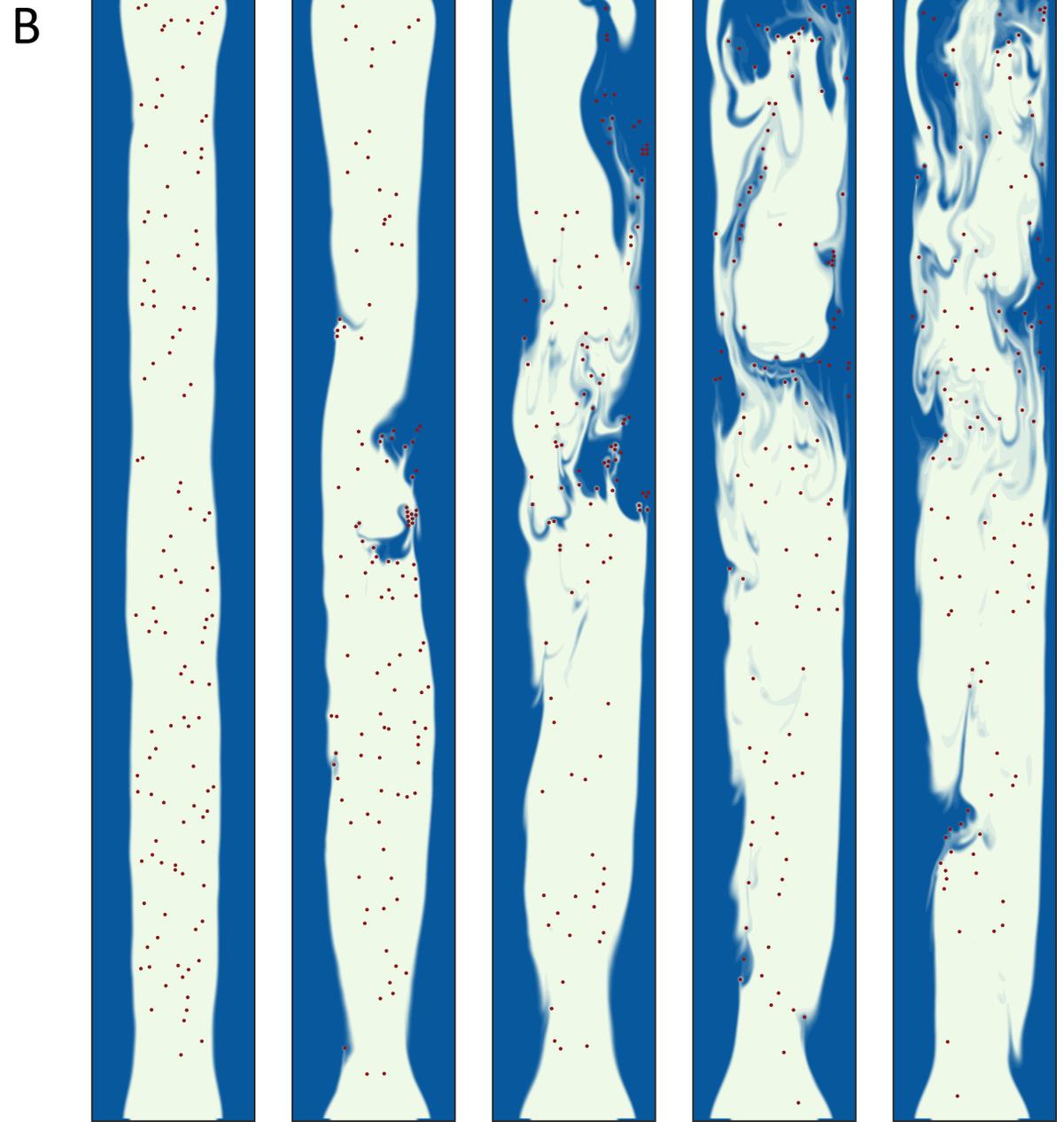
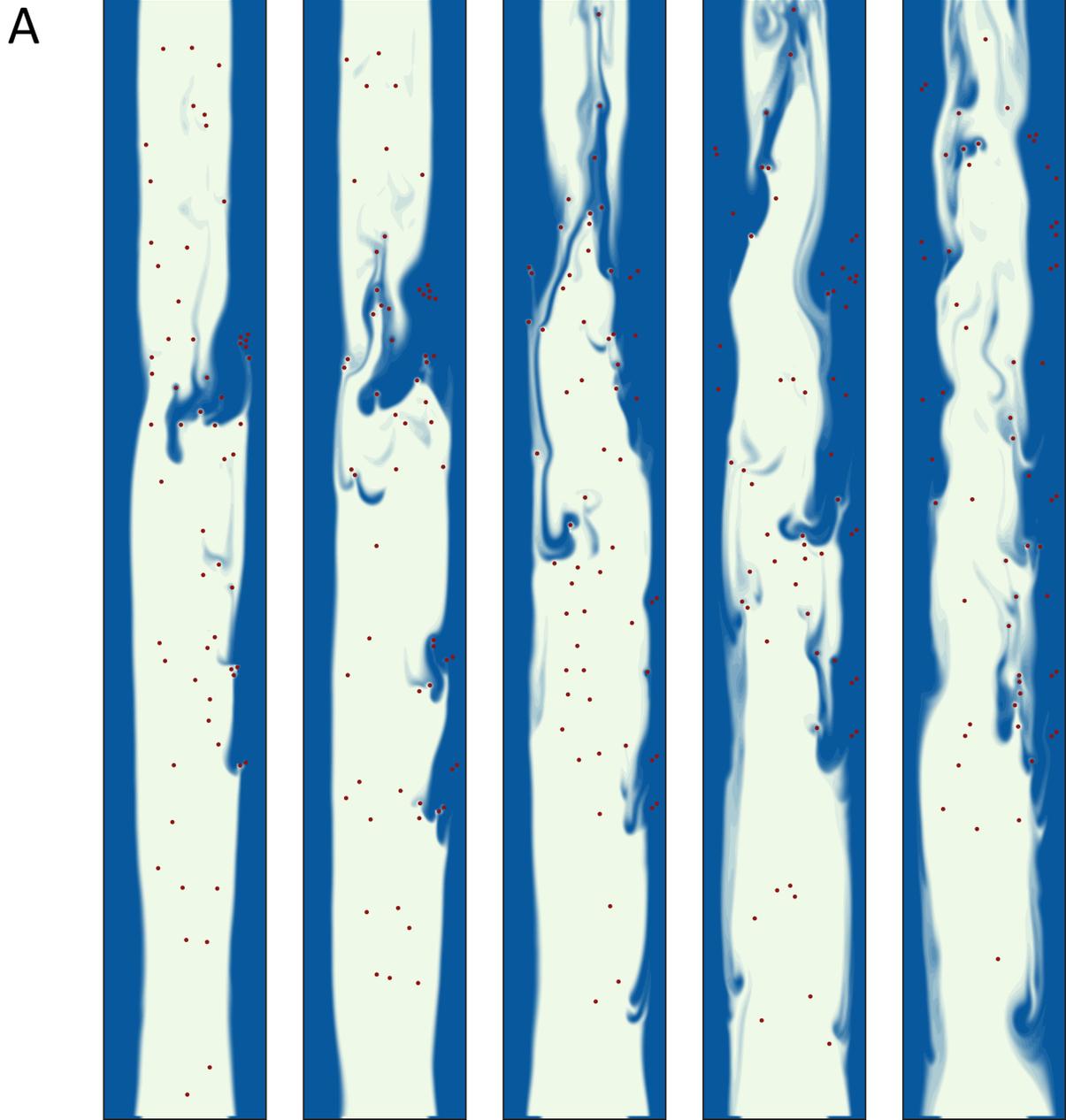


Figure 6.



t  $\xrightarrow{\hspace{10em}}$

t  $\xrightarrow{\hspace{10em}}$

**Figure 7.**

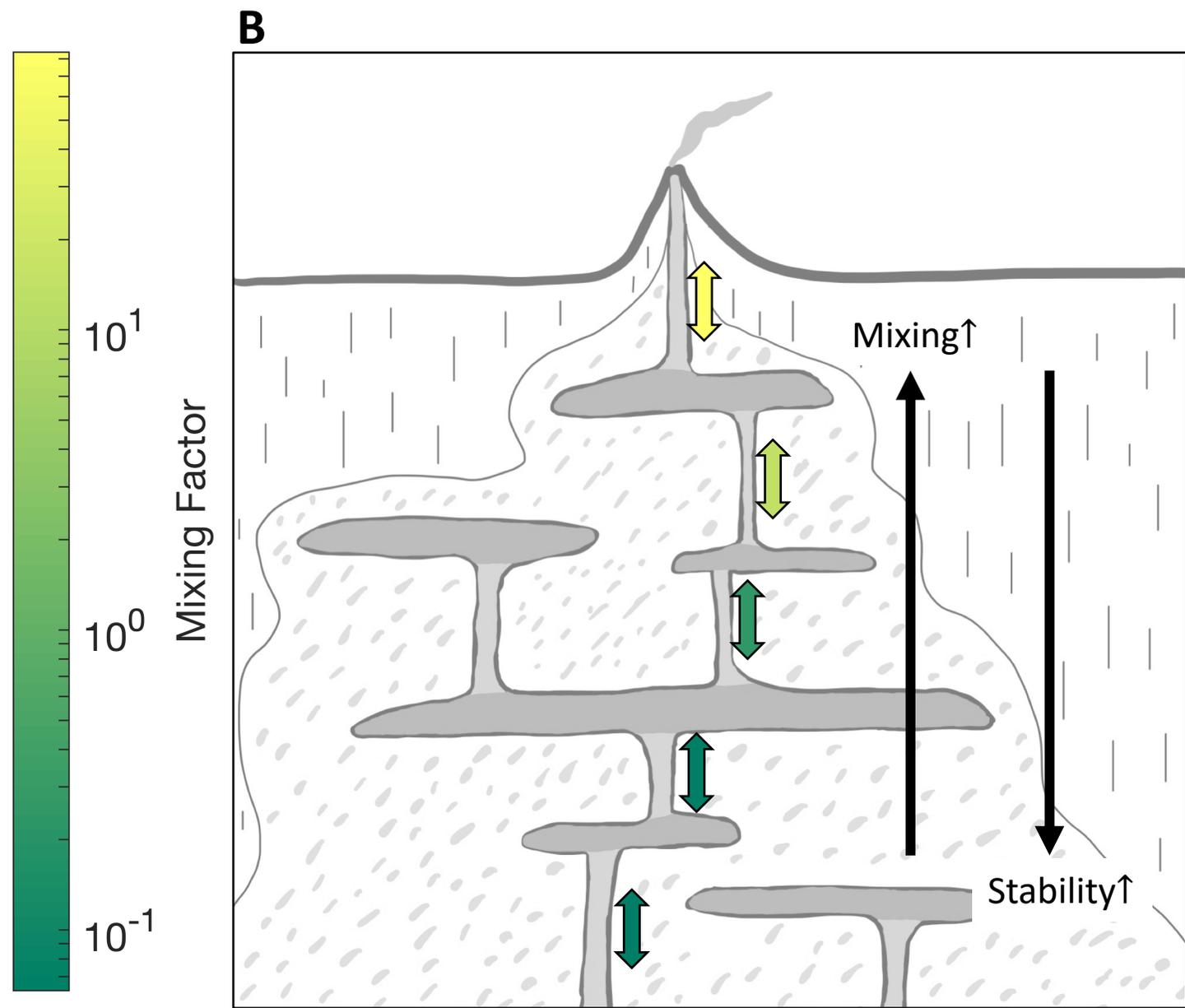
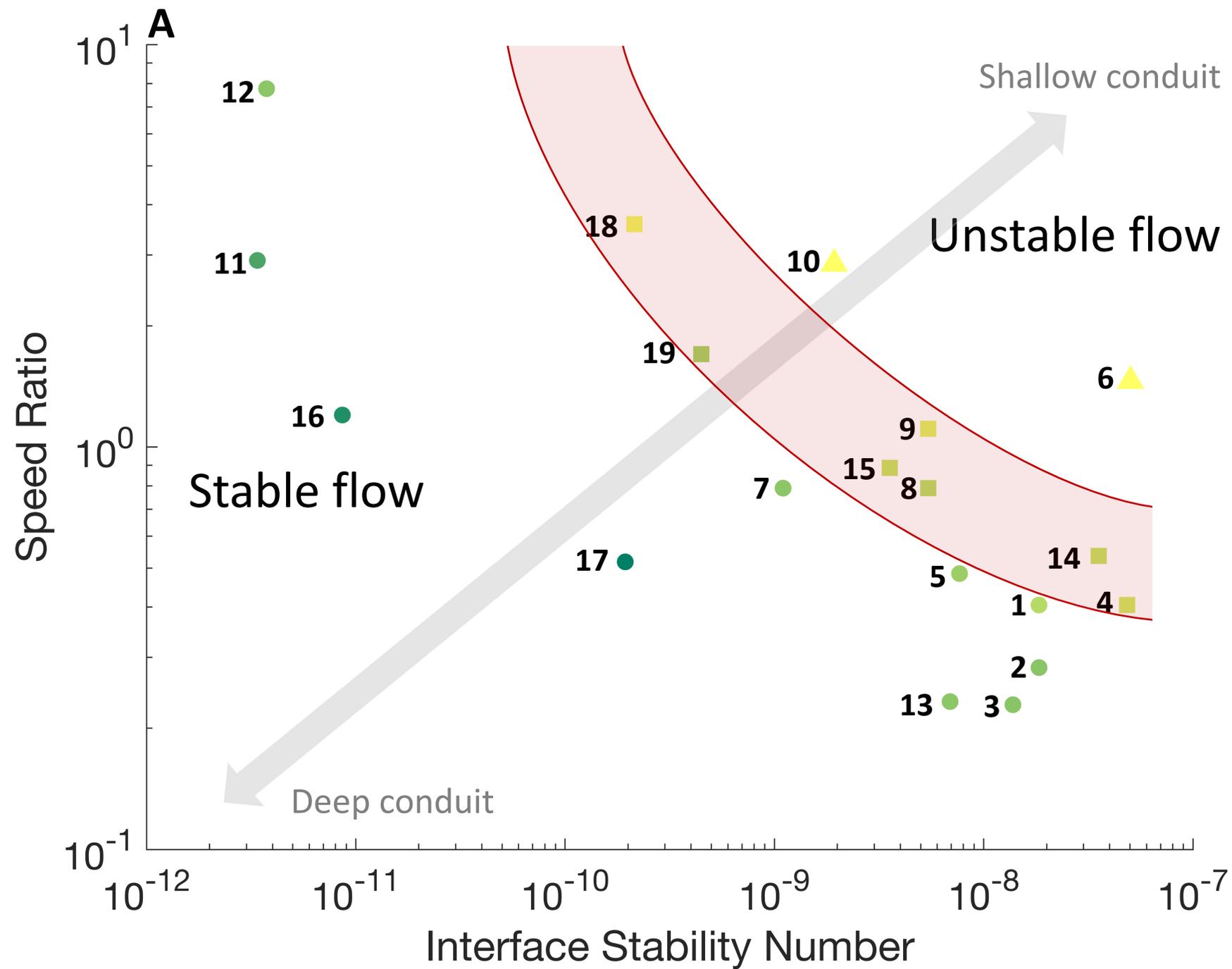
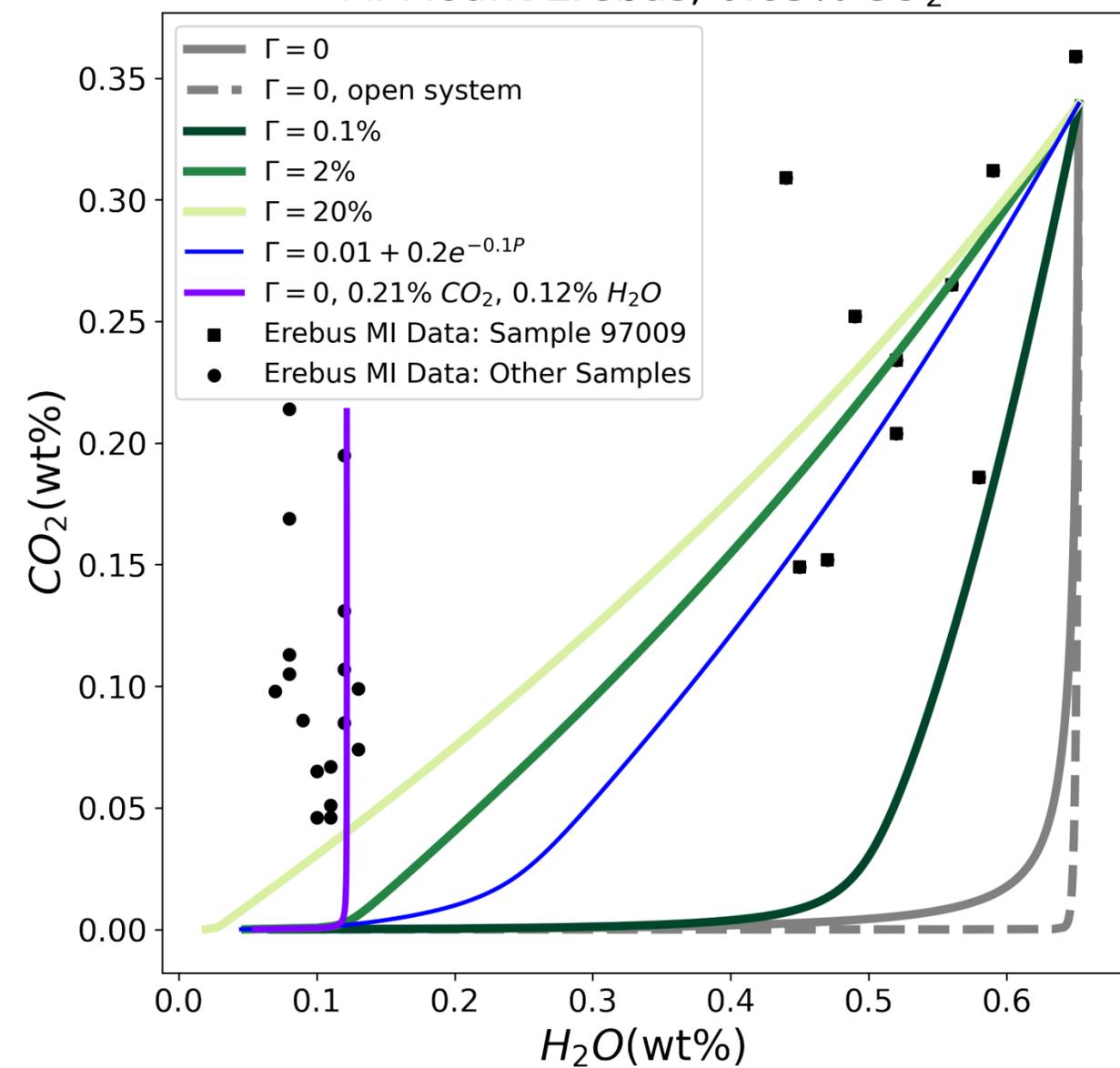
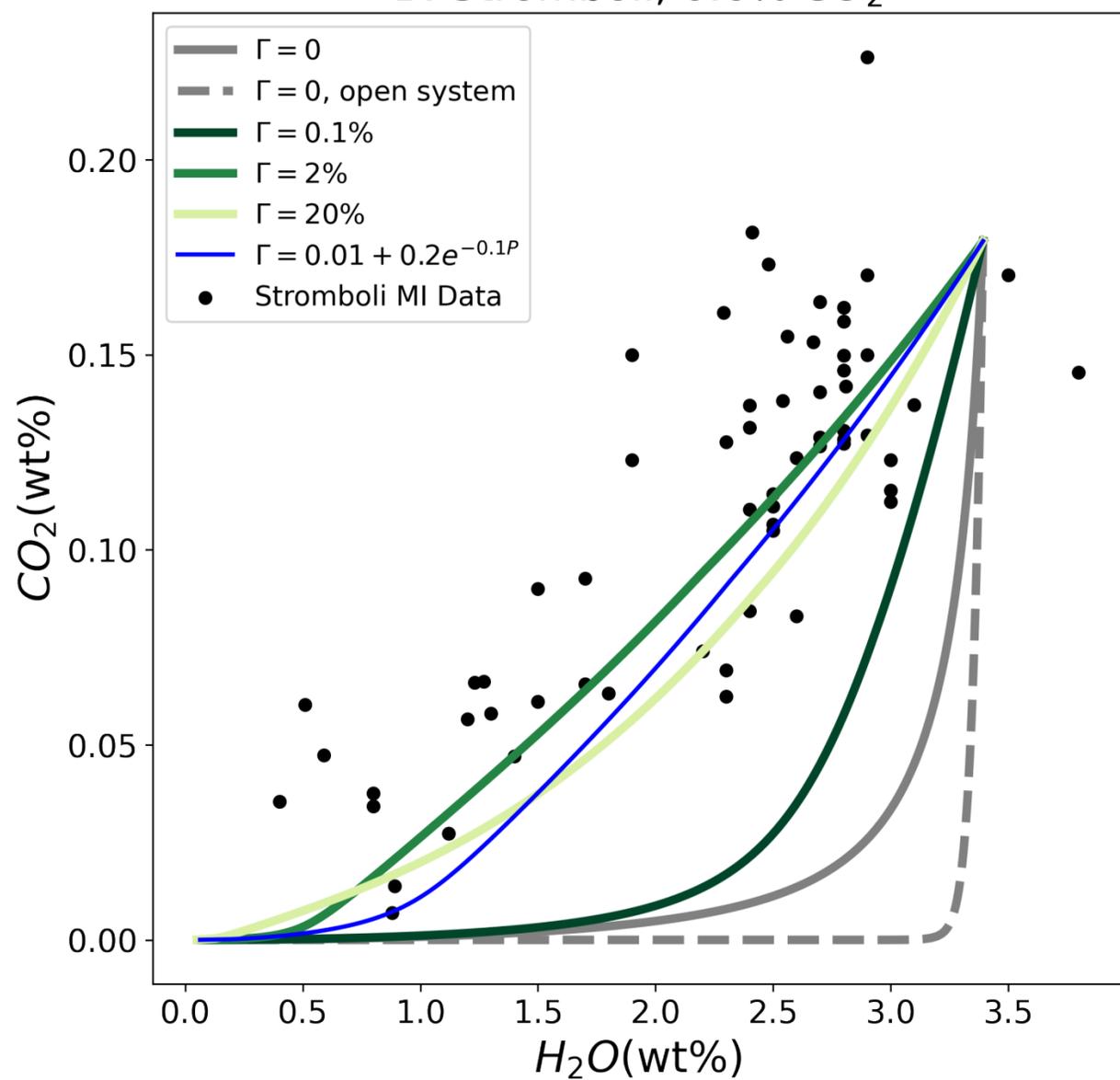
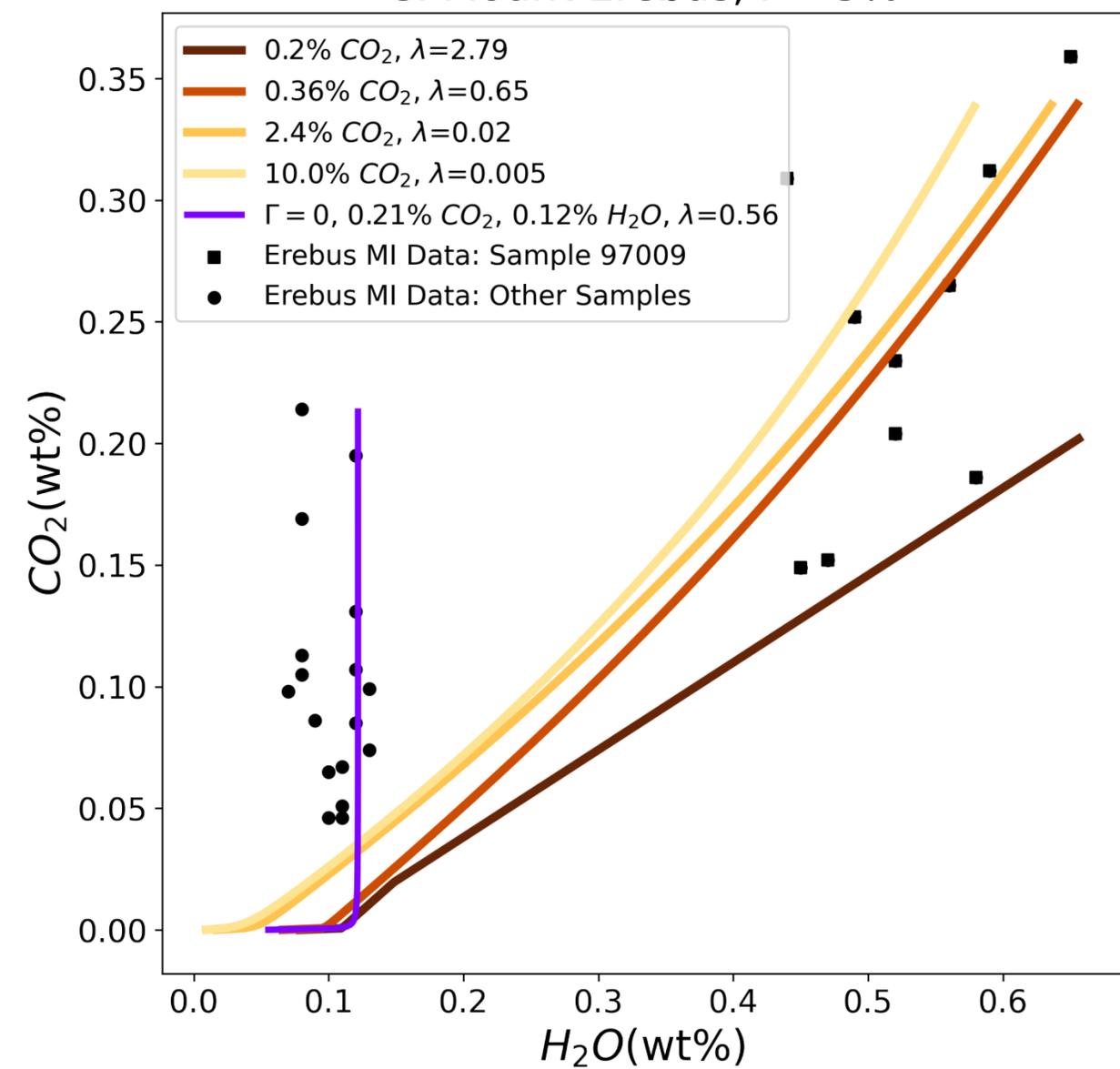


Figure 8.

A: Mount Erebus, 0.65% CO<sub>2</sub>B: Stromboli, 0.6% CO<sub>2</sub>C: Mount Erebus,  $\Gamma = 5\%$ D: Stromboli,  $\Gamma = 1\%$ 