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1	Alteration-induced volcano instability at La Soufrière de Guadeloupe
2	(Eastern Caribbean)
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22	
23	Abstract
24	Volcanoes are unstable structures that deform laterally and frequently experience mass
25	wasting events. Hydrothermal alteration is often invoked as a mechanism that contributes

26 significantly to volcano instability. We present a study that combines laboratory experiments, 27 geophysical data, and large-scale numerical modeling to better understand the influence of alteration on volcano stability, using La Soufrière de Guadeloupe (Eastern Caribbean) as a case 28 29 study. Laboratory experiments on variably-altered (advanced argillic alteration) blocks show 30 that uniaxial compressive strength, Young's modulus, and cohesion decrease as a function of 31 increasing alteration, but that the internal friction angle does not change systematically. 32 Simplified volcano cross sections were prepared (a homogenous volcano, a volcano containing 33 the alteration zone identified by a recent electrical survey, and a volcano with an artificially 34 enlarged area of alteration) and mechanical properties were assigned to zones corresponding to 35 unaltered and altered rock. Numerical modeling performed on these cross sections, using a hydro-thermo-mechanical modeling code, show (1) the importance of using upscaled values in 36 37 large-scale models and (2) that alteration significantly increases volcano deformation and 38 collapse volume. Finally, we combined published muon tomography data with our laboratory 39 data to create a 3D strength map, exposing a low-strength zone beneath the southern flank of 40 the volcano coincident with the hydrothermal system. We conclude that hydrothermal alteration 41 decreases volcano stability and thus expedites volcano spreading and increases the likelihood of mass wasting events and associated volcanic hazards. Hydrothermal alteration, and its 42 43 evolution, should therefore be monitored at active volcanoes worldwide.

44

45 Key points

- Laboratory experiments show that hydrothermal alteration reduces the strength of
 volcanic rock from La Soufrière.
- 48
- Numerical modeling shows that hydrothermal alteration significantly increases volcano
 deformation and collapse volume.

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• We provide a 3D strength map of La Soufrière that exposes a low-strength zone coincident with the hydrothermal system.

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53

55 Plain language summary

56 The rocks forming a volcanic edifice can be altered by circulating hydrothermal fluids. 57 This alteration can influence the physical and mechanical properties of these rocks, which could jeopardize volcano stability. The stability of a volcanic edifice is an important consideration in 58 59 volcanic hazards and risk assessments due to the potentially dire consequences of partial 60 volcanic flank collapse. Using a combination of experimental data, geophysical data, and 61 modeling, and La Soufrière de Guadeloupe (Eastern Caribbean, France) as a case study, we find that hydrothermal alteration decreases volcano stability and thus promotes volcano 62 63 instability and associated volcanic hazards. As a result, we conclude that hydrothermal 64 alteration, and its evolution, should be monitored at active volcanoes worldwide.

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Keywords: hydrothermal alteration; uniaxial compressive strength; Young's modulus;
upscaling; flank collapse

69 **1 Introduction**

70 Volcanoes are inherently unstable structures that are built haphazardly, in both space and time, from the products of successive effusive and explosive eruptions and endogenous 71 72 growth. These materials have highly variable physical and mechanical properties (Heap and Violay, 2021) and often form oversteepened and unstable slopes. As a result, volcano 73 74 deformation (such as volcano spreading; Borgia et al., 2000) and mass wasting events (such as 75 debris avalanches resulting from partial flank collapse, lahars, and rockfalls; Roverato et al., 76 2021) are commonplace at many volcanoes worldwide. Catastrophic collapse resulting from 77 volcano spreading (van Wyk de Vries and Francis, 1997) and mass wasting events present a 78 significant volcanic hazard, and can also trigger hazardous laterally-directed explosions and 79 devastating-both economically and in terms of loss of life-pyroclastic density currents (Cole et al., 2015; Glicken, 1996; Sparks et al., 2020; Voight et al., 1981). Indeed, partial flank 80 81 collapses at about 200 volcanoes have resulted in at least 20,000 fatalities in the last 10,000 82 years (Siebert et al., 2010). Although about 52% of these flank collapses are associated with 83 magmatic eruptions, about 22% are associated with non-magmatic phreatic and/or 84 hydrothermal eruptions (Siebert, 1984; Siebert et al., 1987, 2010). As a result, stability assessments at volcanoes are an essential component of volcano monitoring and volcanic 85 hazard mitigation. 86

Hydrothermal alteration, common to many volcanoes worldwide, is often invoked as a mechanism that contributes significantly to volcano instability. Indeed, studies using geological evidence, geochemical or geophysical data, numerical modeling, laboratory experiments, or a combination of these approaches have highlighted that hydrothermal alteration has or can weaken a volcanic slope sufficiently to promote collapse (Ball et al., 2013, 2015, 2018; Cecchi et al., 2004; del Potro and Hürlimann, 2009; Finn et al., 2018; John et al., 2008; López and Williams, 1993; Opfergelt et al., 2006; Reid et al., 2001; Rosas-Carbajal et al., 2016; Salaün et al., 2011; van Wyk de Vries et al., 2000; Voight et al., 2002; Watters et al., 2000). Fluid and
pore fluid pressure re-distributions caused by alteration are also thought to promote instability
(Ball et al., 2018; Day, 1996; Heap et al., 2021a; Reid, 2004) and erratic explosive behavior (de
Moor et al., 2019; Heap et al., 2019; Mick et al., 2021).

98 Geological evidence is provided by the abundance of hydrothermally altered materials 99 typically found in debris avalanche deposits resulting from partial flank collapse. For example, 100 in their analysis of the 26 December 1997 partial edifice collapse at Soufrière Hills volcano 101 (Montserrat, Eastern Caribbean), Voight et al. (2002) found varicolored, hydrothermally-102 altered materials within the avalanche deposits, highlighting that alteration could have 103 contributed to the collapse of the southern sector of the volcano. Avalanche deposits and 104 material ejected during phreatic explosions at La Soufrière de Guadeloupe (Eastern Caribbean; 105 France) were also found to contain various parts of the active and ancient hydrothermal systems 106 of the volcano (Salaün et al., 2011). Debris avalanche deposits at Tutupaca volcano (Peru) also 107 contain hydrothermally altered materials (Samaniego et al., 2015). Extensive fumarole activity 108 and hydrothermal alteration on the unstable flanks of Casita volcano (Nicaragua) was 109 documented by van Wyk de Vries et al. (2000), a volcano that experienced a partial flank 110 collapse in 1998 with associated debris avalanches that killed about 2,500 people (Kerle and 111 van Wyk de Vries, 2001; van Wyk de Vries et al., 2000). Opfergelt et al. (2006) further 112 suggested that the presence of abundant smectite (up to 50 wt.%) in the hydrothermally-altered 113 core of Casita volcano contributed to slope instability by acting as a barrier to water infiltration 114 thereby promoting fluid circulation along marked discontinuities, gradually decreasing the 115 shear strength of the host-rock. Finally, more than 55 debris flows have originated from Mt 116 Rainer (USA) in the Holocene, and hydrothermally derived clays have been found in some of 117 the most widespread lahar deposits (Reid et al., 2001).

118 Geophysical and geochemical evidence has been provided by a wide range of different 119 techniques. For example, electrical tomography data exposed the extent of the hydrothermally 120 altered zone, the location and geometry of low-strength detachment planes, and potential 121 collapse volumes at La Soufrière de Guadeloupe (Rosas-Carbajal et al., 2016). Hyperspectral 122 remote sensing (Kereszturi et al., 2021) and helicopter electromagnetic and magnetic 123 measurements (Finn et al., 2018) have been used to map hydrothermal alteration zones to 124 inform slope stability modeling at Mt Ruapehu (New Zealand) and Mount Baker (USA), 125 respectively, highlighting the most likely location of future collapses. Finally, geochemical data 126 and modeling provided by López and Williams (1993) suggested that hydrothermal alteration 127 along faults can create weak sliding planes that can facilitate slope failure.

128 Numerical and analog modeling has also provided key insights into how hydrothermal 129 alteration can influence volcano stability. For example, using analogue experiments in which 130 sand-plaster mixtures and silicone were used to represent fresh and altered rock, respectively, 131 van Wyk de Vries et al. (2000) showed that a weak and ductile (hydrothermally altered) core 132 can reproduce the deformation structures observed at Casita volcano, suggesting that 133 hydrothermal alteration destabilized the volcano. Motivated by intensive alteration on the 134 flanks and summit of Mt Rainer, Reid et al. (2001) examined the influence of alteration on 135 volcano stability using a geotechnical method of 3D column limit-equilibrium slope stability 136 analysis. Using a best-estimate distribution of the alteration, and typical values of cohesion, 137 internal friction angle, and rock unit weight for fresh, lightly altered, and highly altered rocks, 138 Reid et al. (2001) concluded that steep slopes and large volumes of altered and weak rock can 139 promote potentially large gravitational failures.

Although these aforementioned studies, and others, have improved our understanding of alteration-induced volcano instability, the shape and size of the alteration zones and/or the geomechanical properties used in large-scale stability modeling are often poorly constrained or

143 arbitrarily chosen. Indeed, there is a paucity of mechanical data for variably altered volcanic 144 rocks for use in large-scale numerical models designed to better understand flank stability. As 145 noted by Ball et al. (2018), the scarcity of rock physical and mechanical properties for altered 146 volcanic rocks limits the investigation of volcano stability using large-scale models. We present 147 here a multidisciplinary study in which we first provide physical and mechanical properties for 148 variably altered andesites from La Soufrière de Guadeloupe. We then assign laboratory-scale 149 and upscaled (i.e. modified to account for large-scale discontinuities that are not captured in 150 laboratory-scale samples) physical and mechanical properties to zones identified by a recent 151 electrical survey of the dome of La Soufrière de Guadeloupe (Rosas-Carbajal et al., 2016) and 152 perform large-scale numerical modeling to showcase the importance of hydrothermal alteration 153 on volcano stability. Finally, we combine recent muon tomography data (Rosas-Carbajal et al., 154 2017) with our laboratory data to create a 3D compressive strength map of the volcano. 155 Although we use La Soufrière de Guadeloupe as a case study, we consider our salient 156 conclusions relevant for andesitic stratovolcanoes worldwide.

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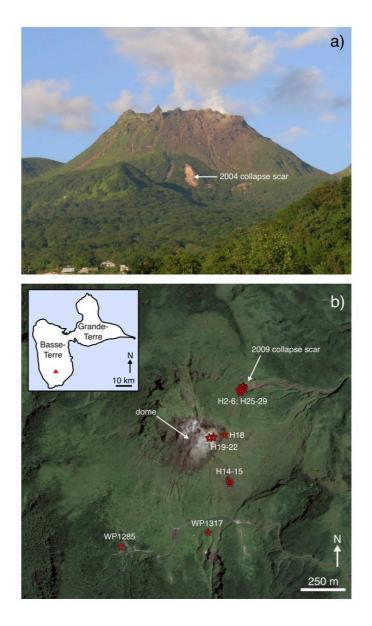
158 **2** La Soufrière de Guadeloupe (Eastern Caribbean, France)

159 La Soufrière de Guadeloupe (Figure 1a), henceforth called La Soufrière, is located on 160 the island of Guadeloupe in the Eastern Caribbean (Figure 1b) and is an archetype of the 161 hazardous andesitic volcanoes that occur in many subduction zones. The current lava dome 162 formed in 1530 CE and, since 1635 CE, there have been six non-magmatic phreatic or 163 hydrothermal eruptions (Komorowski et al., 2005). The largest and most-recent eruption 164 occurred in 1976–1977. After months of seismic unrest, the volcano exploded suddenly and 165 without precursory signs in 1976–1977, resulting in a volcanic crisis that led to the evacuation 166 of about 70,000 people and serious socioeconomic consequences for the island (Feuillard et al., 167 1983; Hincks et al., 2014; Komorowski et al., 2005, 2015).

168 Volcanic unrest has been increasing since the volcano reawakened in 1992, marked by 169 the restart of fumarolic and seismic activity (Komorowski et al., 2005) and, in April 2018, the 170 largest felt volcano-tectonic earthquake since the 1976–1977 crisis (M_L 4.1 or M_W 3.7; Moretti 171 et al., 2020). The 2018 episode of accelerated unrest was interpreted as a failed phreatic eruption 172 and raised serious concerns for the stability of the southwest flank of the dome, which shows 173 the largest displacements (up to 9 mm/year towards the southwest) over the last 20 years 174 (Moretti et al., 2020). The main features of the ongoing unrest include the continuous expansion 175 of the outgassing area on top of the lava dome and the appearance of new steam-dominated 176 fumaroles in July 2014 and February 2016 that are characterized by a deep magmatic gas 177 component (Brombach et al., 2000; Moretti et al., 2020; Villemant et al., 2014). Such expansion 178 was accompanied by an increase in the heat output from the entire dome, which evolved from 179 ~1.2 MW in 2010 to ~7.6 MW in 2020 (Jessop et al., 2021). This increase in heat output is of 180 great concern for La Soufrière as it has been recently demonstrated that volcanoes with a high 181 heat flux are more likely to experience magmatic or phreatic eruptions (Girona et al., 2021). 182 High-flow rate thermal acid sulfate-chloride springs, associated with degassing of the 183 underlying magmatic body, are observed on the slopes and at the base of the dome and supply 184 hot acid fluids and CO₂, but also SO₂, H₂S, HCl, and HF, to the above hydrothermal system 185 (Villemant et al., 2005; 2014). These acidic fluids are (1) transferred through the dome and feed 186 the steam-rich summit fumaroles and (2) circulate through the dome structure along listric, 187 concave-upward structural discontinuities that drain to the southwest (Brombach et al., 2000; 188 Rosas-Carbajal et al., 2016; Salaün et al., 2011; Villemant et al., 2014). Evidence for the 189 preferential circulation of acidic thermal fluids along these structures is provided by the 3D 190 geometry of thermal springs (Moretti et al., 2020; Villemant et al., 2005) and geophysical 191 imaging (Bouligand et al., 2016; Brothelande et al., 2014; Nicollin et al., 2006; Rosas-Carbajal 192 et al., 2016). The shallow hydrothermal system is considered responsible for the recent

193 observed shallow deformation and seismic activity (at depths at or above sea level, with 194 magnitudes less than one) (Moretti et al., 2020). Indeed, the monthly bulletins from the 195 observatory (Observatoire Volcanologique et Sismologique de Guadeloupe, Institut de 196 Physique du Globe de Paris (OVSG-IPGP), 1999-2021: available at 197 http://www.ipgp.fr/fr/ovsg/bulletins-mensuels-de-lovsg) document hundreds of shallow, low-198 magnitude (M < 1) earthquakes each month. Deeper and higher magnitude off-axis seismic 199 activity are thought to be related to increases in pore pressure in response to the pulsatory arrival 200 of hot magmatic fluids along the larger faults that cut the dome (Moretti et al., 2020).

201 The recent unrest, suggesting that La Soufrière is undergoing a renewed period of 202 structural and chemical weakening, raises concern for the stability of the volcano. Indeed, the 203 most-recent unrest and instability at La Soufrière was associated with hydrothermal activity. 204 For example, the materials ejected during the explosive hydrothermal eruption of 1976–1977 205 were largely hydrothermally altered (Feuillard et al., 1983) and geological studies have also 206 shown that La Soufrière has an exceptional record of partial edifice collapse (at least eight 207 collapses in the past 9,150 years) that have produced extensive debris avalanche deposits 208 dominantly composed of hydrothermally altered materials (Boudon et al., 1987; Komorowski 209 et al., 2005; Le Friant et al., 2006; Peruzzetto et al., 2019; Rosas-Carbajal et al., 2016; Salaün 210 et al., 2011). The simulations of Le Friant et al. (2006) and Peruzzetto et al. (2019) suggest that 211 the northern and eastern parts of the town of Saint Claude could be impacted by small-volume 212 partial dome collapses and that, in the worst-case scenario of a major dome collapse, a large 213 part of Basse-Terre could be affected. As a result, not only is La Soufrière an ideal natural 214 laboratory to study the influence of hydrothermal alteration on volcano stability, but such a 215 study is also timely because of the ongoing, and increasing, unrest at the volcano.



218 Figure 1. (a) Photograph of La Soufrière de Guadeloupe (Eastern Caribbean, France). This 219 photograph also shows the collapse scar (indicated by the white arrow) triggered by the 21 220 November 2004 Les Saintes magnitude Mw 6.3 regional earthquake (Feuillet et al., 2011). (b) 221 Map of La Soufrière de Guadeloupe (taken from Google Maps®) showing the sampling 222 locations for the 17 blocks acquired for this study. This image also shows the location of the 223 dome and the 2009 collapse scar (indicated by white arrows). Inset shows a map of 224 Guadeloupe in which the location of La Soufrière de Guadeloupe is indicated by a red 225 triangle.

227 **3 Materials and Methods**

A total of 17 blocks were collected from La Soufrière during a field campaign in 2019 (sampling locations shown as red stars in Figure 1b). Our aim was to sample a range of materials from different locations on the volcano that best represent the variability in porosity and hydrothermal alteration of the rocks forming the edifice. A large proportion of the blocks (eight out of 17) were collected from the collapse scar of the 19 November 2009 landslide, triggered by extreme rainfall (Moretti et al., 2020; Peruzzetto et al., 2019), which was seen as an opportunity to sample blocks that are more representative of the interior of the volcano.

235 Eight blocks were taken from the collapse scar of the 2009 landslide (H2A, H2B, H3, 236 H4A, H5A, H6, H25, and H29), one block was taken from the collapse scar of the landslide 237 triggered by the 21 November 2004 Les Saintes magnitude Mw 6.3 regional earthquake 238 (Feuillet et al., 2011; Figure 1b) (WP1285), two blocks were taken from the West wall of the 239 fault "Faille 30 août" (H14 and H15), and one block, a volcanic bomb from the 1976–1977 240 eruption, was taken from the roof of a small disused thermal bathhouse to the South of the dome 241 (WP1317). The remaining samples were taken from the dome: one block was taken from the 242 "Lacroix Supérieur" outgassing fracture (H18), and four blocks were taken from the lava spines of the 1530 CE dome: two blocks from "Cratère Sud Central" (H19 and H20) and two blocks 243 244 from an adjacent site (H21 and H22). Site descriptions and Global Positioning System (GPS) 245 coordinates are available for each block in the Supplementary Information.

Cylindrical samples were prepared from each of the blocks to a diameter of 20 mm and then cut and precision-ground to a nominal length of 40 mm. The samples were washed and then dried in a vacuum-oven at 40 °C for at least 48 h. The connected porosity of each sample was calculated using the bulk sample volume and the skeletal (solid) sample volume measured by a helium pycnometer. Dry and wet bulk sample densities were calculated using the bulk sample volume and the dry and wet mass, respectively. Polished thin sections were prepared from offcuts of the samples for microstructural analysis, performed using a Tescan Vega 2 XMU scanning electron microscope (SEM). The mineral content of the 17 blocks was analyzed and quantified using X-ray powder diffraction (XRPD) on powdered offcuts of the samples, assisted by Raman spectroscopy on the prepared thin sections (see Supplementary Information for more details).

257 Dry uniaxial compressive strength was measured on oven-dry samples from all 17 258 blocks in a uniaxial load frame. Samples were deformed under ambient laboratory pressure and temperature at a constant axial strain rate of 10⁻⁵ s⁻¹ until macroscopic failure. The static 259 Young's modulus was determined from the elastic portion of the uniaxial stress-strain curves 260 261 (Heap et al., 2020a). Triaxial compression experiments were performed on blocks selected to 262 best represent the variability in observed alteration (samples H2B, H3, and H18). These samples were vacuum-saturated in deionized water, inserted into a rubber jacket, placed inside a 263 264 pressure vessel, and then taken to the target confining (from 10.5 to 25 MPa) and pore fluid 265 pressures (10 MPa for all experiments) using servo-controlled pumps. Samples were deformed under ambient laboratory temperature at a constant axial strain rate of 10⁻⁵ s⁻¹ until either 266 267 macroscopic failure (for brittle experiments) or until 3% axial strain (for ductile experiments). The triaxial apparatus was also used to measure the water-saturated uniaxial compressive 268 269 strength of samples H2B, H3, and H18. For both uniaxial and triaxial experiments, axial 270 displacement and axial load were measured using a linear variable differential transducer and a 271 load cell, respectively. Axial displacement (minus the displacement accumulated within the 272 load chain) and axial load were converted to axial strain and axial stress using the sample dimensions. Here, we define the effective pressure, P_{eff} , as the confining pressure, P_c , minus 273 274 the pore fluid pressure, P_p . We adopt the convention that compressive stresses and strains are 275 positive.

4 Results

4.1 Microstructure, mineralogy, and alteration

279 The mineral contents for each of the 17 blocks are given in Table 1, and Figure 2 shows 280 backscattered SEM images of three blocks selected to represent the range of alteration intensity 281 (defined by their percentage of secondary minerals) observed in the rocks collected for this study (relatively unaltered - H18; moderately altered - H3; and highly altered - H2B). SEM 282 283 images of all the blocks are available in the Supplementary Information. All of the andesite 284 blocks are characterized by a porphyritic texture comprising phenocrysts of dominantly 285 plagioclase and pyroxene (and high-density oxides) within a crystallized groundmass (Figure 286 2). All of the samples also contain secondary minerals, including silica polymorphs (quartz, 287 cristobalite, tridymite, and opal-A), hematite, pyrite, alunite or natroalunite, gypsum, kaolinite, 288 and talc (Table 1). Plagioclase phenocrysts are often pervasively altered and partially replaced 289 by kaolinite (e.g., Figure 2c) or opal-A. Secondary minerals (natroalunite, alunite, cristobalite, 290 tridymite, pyrite, and kaolinite) are also found precipitated within pores and microcracks (see 291 Supplementary Information for additional SEM images).

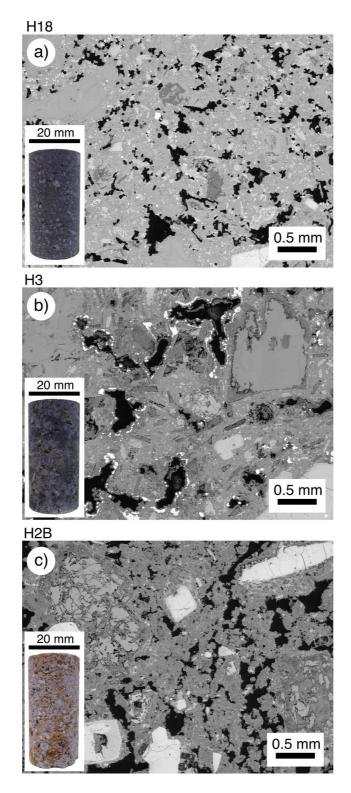


Figure 2. Backscattered scanning electron microscope images of three andesites from La
Soufrière de Guadeloupe (Eastern Caribbean, France) selected to represent the range of
observed alteration: (a) relatively unaltered – H18, (b) moderately altered – H3, and (c) highly

297 altered – H2B. Black – porosity; grayscale – rock groundmass. Insets show photographs of

the 20 mm-diameter cylindrical samples.

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Mineral	H2A	H2B	Н3	H4A	H5A	H6	H14	H15	H18	H19	H20	H21	H22	H25	H29	WP1285	WP1317
Plagioclase	56.7	12.3	46.6	23.3	41.3	30.0	60.7	22.5	61.2	22.0	28.7	24.2	59.5	38.7	62.4	64.7	61.6
Clinopyroxene	8.7	3.4	5.6	4.9	5.2	6.4	6.3	7.3	8.4	5.0	8.9	12.4	8.9	5.3	7.8	5.2	5.9
Orthopyroxene	10.8	9.5	11.8	11.8	11.1	10.8	8.6	9.2	12.2	10.2	15.0	19.3	13.6	10.2	11.2	13.2	15.6
(Ti-) Magnetite	0.7	-	0.8	-	-	-	0.8	-	2.9	-	2.4	3.1	0.8	-	2.7	3.5	0.7
Quartz*	1.0	0.5	0.6	0.6	0.5	0.5	1.7	0.7	0.7	1.7	0.3	0.2	0.6	0.3	0.4	0.2	0.7
Cristobalite*	11.3	12.8	10.6	11.8	13.0	11.1	13.5	10.2	11.7	9.5	11.4	11.7	10.6	9.8	12.4	-	-
Tridymite*	-	-	-	-	-	-	-	0.7	-	-	-	-	-	-	-	13.2	13.2
Hematite*	-	-	-	-	-	-	3.4	-	2.8	2.4	-	-	-	-	3.1	-	-
Pyrite*	3.5	-	3.8	2.3	-	-	-	-	-	-	-	0.4	3.1	0.6	-	-	-
Alunite*	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	2.4
Na-Alunite*	1.4	1.6	2.8	1.3	5.4	5.1	5.1	15.0	-	14.2	0.5	0.5	-	9.8	-	-	-
Gypsum*	-	-	-	0.7	-	-	-	-	-	-	0.8	1.2	-	-	-	-	-
Kaolinite*	6	59.7	17.4	43.3	23.5	36.0	< 1	34.3	-	2.0	2.0	2.0	< 1	25.3	-	-	-
Talc*	-	-	-	-	-	-	-	-	-	-	-	-	2.9	-	-	-	-
Opal-A*	-	-	-	-	-	-	-	-	-	33.0	30.0	25.0	-	-	10.0	-	-

300

301 Table 1. Mineral contents, measured by X-ray powder diffraction and refined using Raman 302 spectroscopy and optical microscopy, of the 17 blocks collected for this study (sampling 303 locations shown in Figure 1b). Values in wt.%. Asterisk denotes a secondary/alteration 304 mineral.

305

306 4.2 Mechanical data

307 Representative uniaxial stress-strain curves for selected samples are shown in Figure 308 3a. Stress is first a non-linearly increasing function of strain, often attributed to the closure of 309 pre-existing microcracks. This stage is followed by a quasi-linear elastic stage. In the next stage, 310 stress is a non-linearly decreasing function of strain, a result of the initiation, propagation, and 311 coalescence of microcracks. Finally, a stress drop signals the formation of a macroscopic shear 312 fracture. Uniaxial compressive strength and Young's modulus are plotted as a function of 313 connected porosity in Figures 3b and 3c, respectively (data available in Table 2). We first note

that the connected porosity of our samples varies from 0.12 to 0.43. The uniaxial compressive strength and Young's modulus of our sample suite varies from ~3.5 to ~150 MPa and from ~1 to ~43 GPa, respectively (Figure 3). Our data show that uniaxial compressive strength and Young's modulus both decrease as a function of increasing porosity (Figure 3).

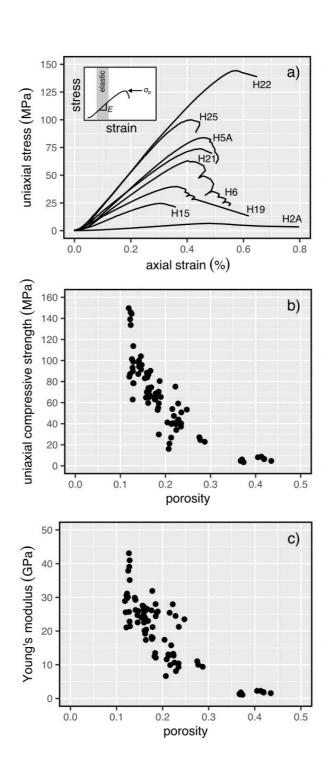


Figure 3. (a) Representative stress-strain curves for selected andesite samples from La Soufrière de Guadeloupe (Eastern Caribbean, France). Inset shows an isolated curve indicating from where the Young's modulus (the slope of the stress-strain curve), *E*, was determined (the quasi-linear elastic stage is indicated by the gray zone). Also labeled is the uniaxial compressive strength, σ_p , which is the maximum axial stress the sample attained prior to macroscopic failure (signaled by the stress drop). Uniaxial compressive strength (b) and Young's modulus (c) as a function of connected porosity (data available in Table 2).

Sample	Connected porosity	Uniaxial compressive strength (MPa)	Young's modulus (GPa)	Percentage of secondary minerals
H2A_2	0.18	63.7	31.9	23.2
H2A 5	0.19	80.5	25.5	23.2
H2A_6	0.19	65.4	25.8	23.2
H2A_11	0.19	59.3	24.4	23.2
H2A_12	0.18	70.4	28.0	23.2
H2B_3	0.42	6.6	1.8	74.6
H2B_10	0.42	6.4	1.9	74.6
H2B_11	0.41	8.7	2.3	74.6
H2B_12	0.41	8.1	2.3	74.6
H2B_15	0.43	4.6	1.6	74.6
H3_3	0.16	70.3	27.3	35.2
H3_7	0.16	69.9	24.9	35.2
H3_8	0.16	64.8	27.5	35.2
H3_11	0.16	59.7	24.0	35.2
H3_13	0.16	71.2	26.2	35.2
H4A_2	0.23	40.3	10.4	60.0
H4A_4	0.23	42.0	10.6	60.0
H4A_6	0.23	43.9	8.1	60.0
H4A_8	0.23	37.2	9.3	60.0
H4A_9	0.22	40.4	9.9	60.0
H5A_2	0.16	88.3	26.4	42.4
H5A_3	0.16	86.6	22.8	42.4
H5A_5	0.16	83.9	22.5	42.4
H5A_8	0.18	68.8	21.2	42.4
H5A_10	0.17	74.5	23.0	42.4
H6_6	0.18	55.1	13.5	52.7
H6_9	0.18	68.0	18.0	52.7
H6_9	0.18	53.0	12.5	52.7
H6_12	0.18	64.2	17.6	52.7
H6_13	0.18	63.0	18.3	52.7
H14_2	0.18	65.4	26.1	23.7

F	I		I	[
H14_3	0.21	21.1	11.6	23.7
H14_5	0.21	26.8	13.1	23.7
H14_6	0.19	29.8	12.1	23.7
H14_10	0.21	16.1	6.6	23.7
H15_3	0.28	24.6	10.0	60.9
H15_4	0.28	27.1	11.0	60.9
H15_5	0.29	22.8	9.4	60.9
H18_3	0.13	101.3	37.9	15.2
H18_4	0.13	89.8	39.1	15.2
H18_5	0.13	98.8	41.0	15.2
H18_6	0.13	93.2	43.1	15.2
H18_7	0.13	113.7	35.1	15.2
H19_3	0.21	39.7	12.7	62.8
H19_8	0.22	47.4	15.8	62.8
H19_9	0.22	34.1	12.7	62.8
H19_12	0.20	41.2	17.4	62.8
H19_15	0.22	39.6	13.2	62.8
H20_2	0.37	4.8	1.2	45.0
H20_3	0.37	4.4	1.3	45.0
 H20_7	0.37	4.6	1.4	45.0
H20 8	0.37	6.1	1.8	45.0
H20 10	0.37	3.4	1.1	45.0
H21 3	0.17	90.2	26.6	41.0
H21_8	0.16	65.2	17.4	41.0
H21 11	0.16	86.9	25.8	41.0
H21 12	0.16	73.8	19.2	41.0
H21 13	0.16	87.2	20.1	41.0
H22 2	0.12	145.7	29.7	17.2
H22_3	0.12	144.3	30.1	17.2
 H22_4	0.12	139.1	30.5	17.2
 H22_5	0.12	149.7	28.9	17.2
H22_6	0.12	133.6	31.1	17.2
H25 2	0.14	99.9	29.3	45.8
H25 4	0.14	97.7	29.3	45.8
H25 5	0.14	94.2	26.2	45.8
H25 9	0.14	87.1	29.9	45.8
H25 12	0.15	83.1	24.0	45.8
H29_2	0.22	53.9	25.4	25.9
H29 8	0.23	59.1	24.5	25.9
H29 9	0.22	75.2	27.9	25.9
H29 12	0.25	53.4	23.5	25.9
H29 16	0.24	50.7	21.3	25.9
WP1285 2	0.13	78.5	22.8	13.4
WP1285_8	0.13	63.0	25.7	13.4
WP1285 10	0.13	78.3	21.4	13.4
WP1285 11	0.12	87.2	21.1	13.4
WP1285 15	0.12	84.6	25.6	13.4
WP1317 2	0.16	67.3	20.4	16.3
WP1317_6	0.16	84.5	24.7	16.3
WP1317 7	0.15	91.5	22.6	16.3
····/_/	0.15	71.5	22.0	10.5

WP1317_8	0.15	95.8	25.9	16.3
WP1317_12	0.15	104.0	24.7	16.3

329 Table 2. Connected porosity, uniaxial compressive strength, Young's modulus, and the
330 percentage of secondary minerals (i.e. alteration) for the samples prepared for this study.
331 Sample locations are provided in Figure 1b (site descriptions and GPS coordinates are
332 available in the Supplementary Information).

333

334 Samples from three blocks were selected for triaxial compression experiments: a 335 relatively unaltered block (H18), a moderately altered block (H3), and a very altered block 336 (H2B). The microstructure of these samples is provided in Figure 2 and their mineral 337 componentry in Table 1. The percentages of secondary minerals (determined using the XRPD data; Table 1) for these rocks are 15.2, 35.2, and 74.6%, respectively. Triaxial experiments were 338 339 performed on H18 samples at effective pressures of 0, 2.5, 5, 10, and 15 MPa (Figure 4a), on 340 H3 samples at effective pressures of 0, 2.5, 5, and 7.5 MPa (Figure 4b), and on H2B samples at 341 effective pressures of 0, 0.5, 1, and 2 MPa (Figure 4c) (data available in Table 3). Different 342 effective pressures were chosen for the different samples in an attempt to ensure the brittle 343 behavior required to calculate the cohesion and angle of internal friction. Blocks H18 (Figure 344 4a) and H3 (Figure 4b) were brittle over the chosen range of effective pressures. The 345 compressive strength of blocks H18 and H3 increased as a function of increasing effective 346 pressure: H18 increased from 65.4 to 143.8 MPa and H3 increased from 31.5 to 54.8 MPa as 347 effective pressure was increased from 0 to 15 and 7.5 MPa, respectively (Figure 4). The stress-348 strain curves for block H2B are very different to those for blocks H18 and H3 (Figure 4). Subtle 349 strain softening associated with brittle behavior was observed for the experiments performed at 350 effective pressures of 0 and 0.5 MPa, but no strain softening was seen at 1 and 2 MPa, 351 suggestive of ductile behavior (Figure 4c).

The cohesion and angle of internal friction calculated using these triaxial data (excluding the two ductile H2B experiments), necessary for the large-scale numerical modeling, were 14.8 MPa and 46.7° for H18, 7.1 MPa and 37.7° for H3, and 0.4 MPa and 42.5° for H2B, respectively (Figure 4). The method for determining these values is explained in the Supplementary Information.

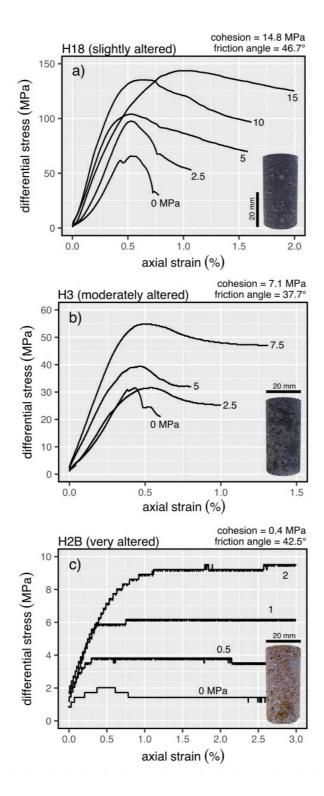


Figure 4. Stress-strain curves for andesites from La Soufrière de Guadeloupe (Eastern
Caribbean, France) deformed under different effective pressures (given by the number
adjacent to each curve). (a) Relatively unaltered sample H18. (b) Moderately altered sample
H3. (c) Very altered sample H2B. Differential stress is the axial stress minus the confining

363 pressure. Insets show photographs of the 20 mm-diameter cylindrical samples. The values of

364 cohesion and internal friction angle, calculated using these data, are also provided for each of

365

the three rocks.

366

Sample	Connected porosity	Percentage of secondary	Pore fluid pressure (MPa)	Confining pressure (MPa)	Effective pressure (MPa)	Peak differential stress (MPa)
		minerals	(IVIF a)	(IVIF a)	(MIFA)	stress (wira)
H18_11	0.13	15.2	0 (wet)	0	0	65.7
H18_10	0.14	15.2	10	12.5	2.5	97.7
H18_2	0.12	15.2	10	15	5	104.0
H18_8	0.12	15.2	10	20	10	135.2
H3_14	0.16	35.2	0 (wet)	0	0	31.5
H3_12	0.15	35.2	10	12.5	2.5	31.7
H3_4	0.15	35.2	10	15	5	39.4
H3_5	0.16	35.2	10	17.5	7.5	54.9
H2B_7	0.44	74.6	0 (wet)	0	0	2.0
H2B_8	0.44	74.6	10	10.5	0.5	3.8
H2B_6	0.39	74.6	10	11	1	-
H2B_5	0.38	74.6	10	12	2	-

367

368 Table 3. Experimental summary for the triaxial deformation experiments (and wet uniaxial 369 experiments) performed on relatively unaltered (H18), moderately altered (H3), and very 370 altered (H2B) andesites from La Soufrière de Guadeloupe (Eastern Caribbean, France). 371 Sample locations are provided in Figure 1b (site descriptions and GPS coordinates are available in the Supplementary Information) and microstructural images are provided in 372 373 Figure 2. 374

375 **5** Discussion

376 5.1 Hydrothermal alteration and mineral precipitation

377 Volcanic andesitic systems discharging acid-chloride-sulfate hydrothermal fluids 378 (Browne, 1978; Ellis and Mahon, 1977; Taran and Kalacheva, 2020) are typically characterized 379 by either propylitic alteration, resulting from the rapid ascension of acidic fluids and the lack

380 of time available for neutralization (Taran and Kalacheva, 2020), or intermediate to advanced 381 argillic alteration in zones where fluids can circulate. The samples measured for this study were 382 sourced from (1) zones proximal to summit fumarolic vents that tap the central zone 383 characterized by the rapid ascent of steam separated from a boiling hydrothermal aquifer 384 (Brombach et al., 2000; Moretti et al., 2020) (e.g., H18-22; Table 1) and (2) from zones far 385 from currently active fumarolic zones, where the shallow groundwaters are heated via 386 conduction and mixed with Na-Cl liquids from the hydrothermal aquifer (Brombach et al., 387 2000; Villemant et al., 2014) (e.g., those sampled from the collapse scar of the 2009 landslide: 388 H2A, H2B, H3, H4A, H5A, H6, H25, and H29; Table 1) (Figure 1b).

389 The secondary mineral assemblage in our samples (Table 1) is essentially the same as 390 that found in the debris avalanche deposits to the southwest of La Soufrière (Salaün et al., 2011). 391 However, the debris-avalanche deposits investigated by Salaün et al. (2011) contain abundant 392 smectite, whereas the samples collected for this study contain kaolinite and no smectite (Table 393 1). The presence of kaolinite, a key mineral in the argillic alteration facies, typically indicates 394 fluid-rock interaction at relatively low fluid temperatures (< 150–200 °C) and a pH of ~4.5–6 395 (Fulignati, 2020; Inoue, 1995), but can also occur in the advanced argillic alteration facies (pH 396 < 3; temperatures up to about 300 °C). The altered samples analyzed in this study therefore 397 represent a more advanced stage of hydrolysis reactions than the smectite-bearing products 398 from the Holocene debris-avalanche deposits described in Salaün et al. (2011), indicating that 399 the rocks analyzed here were altered by hydrothermal fluids with a higher H⁺ activity that led 400 to complete alkali leaching.

401 The most abundant secondary minerals are cristobalite, amorphous silica, natroalunite, 402 and kaolinite (Table 1). Cristobalite is a silica polymorph stable at low-pressure and high-403 temperature in its cubic β -form (Schipper et al., 2020). Because it is kinetically favored, 404 metastable cristobalite is commonly found in preference to quartz in lava domes, where it 405 precipitates from a vapor-phase enriched in magmatic components or results from groundmass 406 devitrification (Horwell et al., 2013; Martel et al., 2021; Schipper et al., 2020). Our samples 407 contain tetragonal α-cristobalite, suggesting a final temperature lower than 240 °C, the nominal 408 temperature for the transition from β - to α -cristobalite (Damby et al., 2014). Schipper et al. 409 (2020) identified a lower and broader range for the β - α transition in volcanic cristobalites 410 (~150–200 °C), which overlaps with the temperature interval inferred by the presence of 411 kaolinite. The cristobalite in the studied samples precipitated onto pore walls (see Figure 2 and 412 the Supplementary Information) and so we consider that it predominately formed by vapor 413 deposition. Amorphous silica (opal-A) partially to completely replaced the plagioclase 414 phenocrysts (see Figure 2 and the Supplementary Information), as often observed in acid-sulfate 415 alteration zones (McCollom et al., 2013).

416 Alunite is a characteristic family of sulfate minerals produced by hypogene magmatic-417 hydrothermal and steam-heated alteration or supergene processes (Rye et al., 1992). As such it 418 is commonly found in the alteration products of rhyolitic to andesitic rock compositions, 419 including surficial solfataric or acid hot spring environments. Alunite and natroalunite are the 420 two most common minerals of the alunite group (Stoffregen et al., 2000). Only the volcanic 421 bomb from the 1976–1977 eruption (sample WP1317) contains alunite, whereas natroalunite is 422 commonly observed (Table 1). Natroalunite is the high temperature end-member of the alunite group and typically forms at 200–350 °C, but is also stable at 380–450 °C (Henley and Berger, 423 424 2011; Meyer and Hemley 1967). Natroalunite precipitates from an acidic (pH < 4) fluid with a 425 high sulfate content and high [Na]/[K] ratio (Hemley et al., 1969; Stoffregen and Cygan, 1990). 426 Natroalunite precipitation requires fluids with [Na]/[K] about 100 (at pH = 3; Devell and 427 Dipple, 2005), much higher than measured in circulating fluids and in whole rock and 428 groundmass compositions at La Soufrière (Boudon et al., 2008; Metcalfe et al., 2021; Villemant 429 et al., 2005), suggesting that equilibrium was not obtained or that the fluids responsible are

430 different from those discharged at the surface (Deyell and Dipple, 2005). If the fluids are 431 different, acidic Na-Cl-SO₄ fluids would be required, which form H₂S and H₂SO₄ following the 432 interaction between Na-Cl waters and elemental sulfur at depth (Taran and Kalacheva, 2020). 433 This hypothesis is compatible with the observation that highly acidic chlorine-sulfate rich fluids 434 (pH < 1) have been rising and circulating within the dome as of early 1998, forming acid ponds 435 within summit craters (Komorowski et al., 2005, Rosas-Carbajal et al., 2016; Villemant et al., 436 2014). Alternative hypotheses include the interaction of Na-Cl neutral water (water/rock 437 volume ratio >> 1) with rocks already showing advanced argillic alteration, and that the rocks 438 collected represent old and buried solfataric fields (Marini et al., 2003) resulting from previous 439 volcanic cycles.

440 We conclude that the observed secondary mineral assemblages (Table 1) are the result 441 of intense fluid-rock interactions. Advanced argillic alteration of the shallow portion of the 442 edifice was promoted by the efficient circulation of cooled (below 350 °C, and down to 150°C-443 200 °C), acidic (pH < 4) hydrothermal fluids, possibly mixed with meteoric waters. The 444 alteration assemblages observed at La Soufrière are similar to those commonly identified at 445 domes and craters of active volcanoes worldwide (Heap et al., 2019; Zimbelman et al., 2005; Yilmaz et al., 2021), although the rocks from Mt Unzen volcano (Japan) were overprinted by 446 447 propylitic alteration (Yilmaz et al., 2021), not observed here. Multiple reaction paths and 448 variable alteration intensity reflect a variable degree of reaction progress. For a given alteration 449 rate, reaction progress will increase as exposure to altering fluids and pathway length increase, 450 or as water flow and water/rock volume ratio decrease. Variations in these environmental 451 parameters simply reflect the heterogeneity and structural complexity of the dome and thus the complex architecture of the hydrothermal system, as revealed by geological and geophysical 452 453 data (Brothelande et al., 2014; Jessop et al., 2021; Moretti et al., 2020; Nicollin et al., 2006; Rosas-Carbajal et al., 2016; Tamburello et al., 2019). In particular, the complex 3D geometry 454

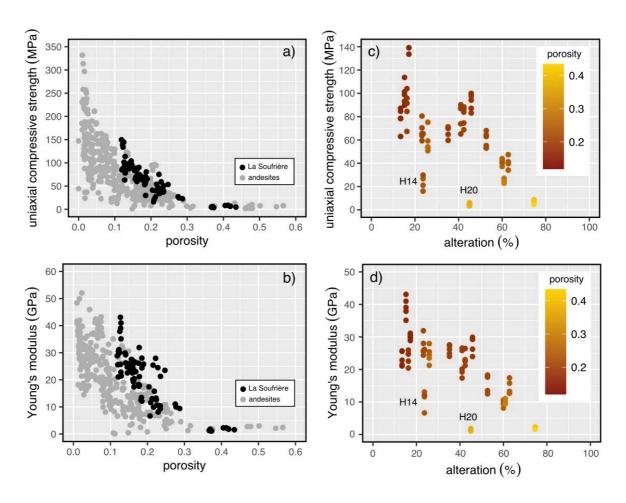
455 of large faults and fractures in the dome allow for the efficient drainage of fluids and the 456 compartmentalization of supergene and hypogene alteration. In such an environment: (1) 457 intermediate to advanced argillic alteration will develop preferentially along discontinuities 458 favoring fluid circulation, leading to the formation of low-strength layers dominated by clay 459 and sulfate minerals; (2) partial edifice collapse or erosion favored in these weakened zones 460 will expose pyrite-rich zones of the hydrothermal system below the water table and promote 461 the deepening of supergene oxidation and the alteration aureole (as seen in the rain-triggered 462 landslide in 2009; Rosas-Carbajal et al., 2016); and (3) H₂S will be easily released by deeper, 463 boiling fluids or by the disproportionation of magmatic SO₂ to H₂S and aqueous sulfate during 464 the condensation of magmatic vapor plume at intermediate depths.

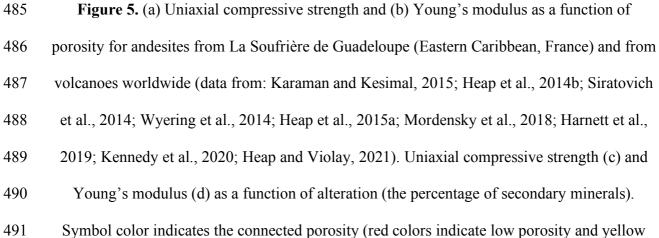
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466 5.2 The influence of porosity and alteration on uniaxial compressive strength

467 New mechanical data show that the uniaxial compressive strength and Young's 468 modulus of variably-altered andesites from La Soufrière both decrease as a function of 469 increasing porosity (Figure 3), in accordance with previous studies on volcanic rocks (Heap et 470 al., 2014a, 2020a; Heap and Violay, 2021; Mordensky et al., 2018; Schaefer et al., 2015; Zhu 471 et al., 2016). We compare our new data with those previously published for andesites in Figure 472 5a and 5b, which show that the data for the rocks from La Soufrière are in broad agreement 473 with those previously published. This comparison shows that the andesites forming La 474 Soufrière are generally more porous, and therefore weaker (Figure 5a) and softer (Figure 5b), 475 than andesites from other stratovolcanoes (the block with the lowest porosity, H22, has a 476 porosity of 0.12). Indeed, muon tomography data have shown that the material forming the 477 dome at La Soufrière is typically low-density (typically ≤ 1500 kg.m⁻³; Lesparre et al., 2012; 478 Rosas-Carbajal et al., 2017) and, therefore, likely comprises high-porosity rocks. However, we 479 note that the relative abundance of high-porosity rocks with a low strength and a low Young's

480 modulus from La Soufrière may be a consequence of sampling only 17 blocks. In other words,
481 low-porosity rocks may exist at La Soufrière and, if many more blocks were sampled, maybe
482 the data would encompass a similar range to the compiled dataset for other volcanoes.





colors indicate high porosity). Outliers-blocks H14 and H20-are labelled on panels (c) and

(d).

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- 494

495 Of interest to the goal of this study is whether hydrothermal alteration has influenced 496 the compressive strength and Young's modulus of the materials collected. Our triaxial data 497 highlight that alteration (percent of secondary minerals) reduces rock cohesion, but does not 498 systematically influence the angle of internal friction (Figure 4). Experimental studies have 499 shown that alteration can influence the strength of volcanic rock (del Potro and Hürlimann, 500 2009; Farquharson et al., 2019; Frolova et al., 2014; Heap et al., 2020b, 2021b, Mordensky et 501 al., 2018, 2019; Wyering et al., 2014). The type of alteration, porosity increasing dissolution 502 and alteration to clay (del Potro and Hürlimann, 2009; Farquharson et al., 2019; Opfergelt et 503 al., 2006; Watters and Delahaut, 1995) or porosity decreasing mineral precipitation (Heap et 504 al., 2020b, 2021), dictates whether the alteration decreases or increases the strength and 505 Young's modulus of the rock. Figure 5c and 5d show plots of uniaxial compressive strength 506 and Young's modulus, respectively, as a function of the percentage of secondary minerals for 507 the rocks from La Soufrière. The symbols in Figure 5c and 5d are color-coded to show their 508 porosity: red colors indicate low porosity and yellow colors indicate high porosity. Although 509 there are two notable outliers, Figure 5c and 5d, respectively, show that uniaxial compressive 510 strength and Young's modulus decrease as a function of increasing hydrothermal alteration. 511 Uniaxial compressive strength and Young's modulus are reduced from $\sim 60-140$ to < 10 MPa 512 and from 20–45 to < 5 GPa as alteration increases from $\sim 16-18$ to $\sim 75\%$ (Figure 5c and 5d). 513 The low uniaxial compressive strengths and Young's moduli of the two relatively unaltered blocks that form the most obvious outliers (H14 and H20) can be explained by meso-scale 514 515 fractures found within sample H14 (sample photographs are provided in the Supplementary 516 Information) and the high porosity of sample H20, respectively. Although the data in Figure 5c

517 and 5d suggest that an increase in alteration results in a decrease in strength and Young's 518 modulus, it is difficult to untangle the influence of porosity. In other words, are the weak 519 samples weak because of alteration or are the more porous samples simply more altered because 520 of their higher surface area available for rock-fluid interactions? Indeed, our data do show that, 521 in general, alteration increases as a function of increasing porosity. Although laboratory-522 controlled hydrothermal alteration experiments (Farquharson et al., 2019) are required to 523 definitively answer this question, we contend here that the observed alteration served to reduce 524 the strength and Young's modulus of these andesites from La Soufrière, regardless of their porosity, due to the presence of abundant clay minerals, phases considered to reduce the 525 526 strength of volcanic rocks (del Potro and Hürlimann, 2009; Nicolas et al., 2020; Opfergelt et 527 al., 2006; Watters and Delahaut, 1995).

528

529 5.3 Influence of alteration on the stability of La Soufrière de Guadeloupe: large-scale numerical
530 modeling

531 We used the open-source hydro-thermo-mechanical modeling code LaMEM 532 (Lithosphere and Mantle Evolution Model; <u>https://bitbucket.org/bkaus/lamem/src/master/;</u> 533 Kaus et al., 2016), which models the non-linear, visco-elastoplastic deformation of rocks, to 534 investigate the influence of upscaling and alteration on the stability of La Soufrière. This model 535 has recently been used to model large-scale volcano deformation (Heap et al., 2021a). 536 Mathematically, the model solves a coupled system of conservation equations, the conservation 537 equations of momentum and mass. The various deformation mechanisms are connected in an 538 additive constitutive relationship, where the total strain rate is the sum of the individual strain 539 rates of elastic, viscous, and plastic deformation. LaMEM uses the Drucker-Prager failure 540 criterion to determine the magnitude of the plastic strain rate. Numerically, LaMEM uses a 541 staggered-grid finite difference method for discretization and a marker-and-cell method to 542 assign and track rock properties within a Eulerian advection framework (Harlow and Welch, 543 1965). This approach enables the model to attain the large deformations required in, for 544 example, salt tectonics, continental collision, and magmatic systems (Baumann et al., 2014; 545 Pusok and Kaus, 2015; Reuber et al., 2018). The ability to model large deformations is seen 546 here as an advantage of LaMEM over commonly-used commercial packages such as FLAC (by 547 Itasca Consulting Group), RS2 (by Rocscience), and PLAXIS 2D (by Virtuosity). We refer the 548 reader to Kaus et al. (2016) and the Supplementary Information for a more detailed description 549 of LaMEM.

550 Models were run on simplified 2D North-South cross-sections of the volcano (643 km 551 Easting), built using the geomIO tool (Bauville and Baumann, 2019). The model domain was first separated into deformable and non-deformable domains, guided by geological 552 553 observations and the recent electrical survey of Rosas-Carbajal et al. (2016). The models have 554 a dimension of 2000×800 m, a model resolution of 4×3 m, free-slip boundary conditions at 555 the sides, and a free surface at the top. All our models assumed an entirely liquid saturated 556 volcano. The average topographic height was used as the reference surface for determining the hydrostatic pressure, p_h , using $p_f = p_h + \lambda_i (p_{lith} - p_h)$, where p_f and p_{lith} are the pore 557 fluid and lithostatic pressure, respectively, and λ_i is the pore fluid pressure ratio. Following 558 559 Heap et al. (2021a), we assumed a value of 0.2 for λ_i (Table 4). To account for the uncertainty 560 in λ_i , additional models using $\lambda_i = 0.4$ are provided in the Supplementary Information. We 561 highlight that, although the assumption of complete volcano saturation is an oversimplification 562 (Ball et al., 2015, 2018; Hurwitz et al., 2003), the main goal of our modeling was to understand, 563 all else being equal, whether hydrothermal alteration is a contributing factor to the deformation 564 observed at La Soufrière. Further, the distribution of liquid-saturated and dry zones within the 565 volcano is currently poorly constrained. However, using the average topographic height as the reference surface resulted in a "dry" volcano summit with a pore fluid pressure of zero (seeSupplementary Information for further details).

568 By modifying the segmentation of the deformable domain, we prepared three cross-569 sections: (1) a homogeneous volcano, (2) a volcano containing alteration zones identified by 570 the electrical conductivity data of Rosas-Carbajal et al. (2016), and (3) a volcano in which we 571 artificially enlarged the highly altered zone identified by Rosas-Carbajal et al. (2016). Zones 572 within the volcano cross-sections were designated as either slightly altered (V3), moderately 573 altered (V2), or highly altered (V1) (Figures 6a-d). Zones V3, V2, and V1 were assigned the 574 mechanical properties of samples H18, H6, and H2B, respectively, the three blocks chosen to 575 represent relatively unaltered, moderately altered, and highly altered rock (Figure 2; Table 1). 576 The laboratory data used in the modeling were: water-saturated bulk density, Young' modulus, 577 cohesion, and the angle of internal friction. Poisson's ratio, not measured here, was assumed to 578 be 0.2 (as recommended by Heap et al., 2020a). The model parameters for V3, V2, and V1 are 579 provided in Table 4. As the models are dynamic, we must decide when the models reach a 580 quasi-steady state and can be compared (see Baumann and Kaus, 2015). Here, we stop the 581 models after an initial deformation phase that is dominated by an elastic response. For each of 582 the three modeled scenarios, we ran one model in which we used the laboratory data and one 583 model in which we used upscaled values of the laboratory data (Table 4).

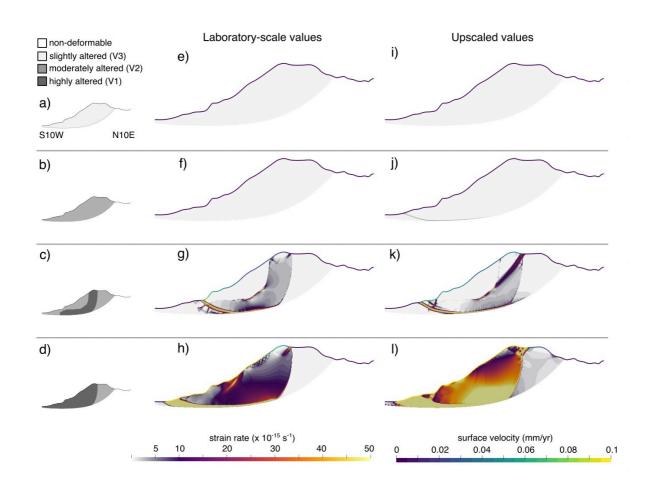
	V1 (highly altered)	V2 (moderately altered)	V3 (relatively					
			unaltered)					
Laboratory-scale input parameters								
Bulk density(kg/m ³)	1900	2400	2500					
Young's modulus (GPa)	0.4	9.6	20.3					
Poisson's ratio	0.2	0.2	0.2					
Cohesion (MPa)	0.4	7.1	14.8					
Internal friction angle (°)	42.5	37.7	46.7					
Pore fluid pressure ratio	0.2	0.2	0.2					
	Upscaled inp	ut parameters						
Bulk density(kg/m ³)	1900	2400	2500					
Young's modulus (GPa)	0.2	3.9	8.3					
Poisson's ratio	0.3	0.3	0.3					

Cohesion (MPa)	0.67	1.09	2.02
Internal friction angle (°)	30.1	33.5	46.4
Pore fluid pressure ratio	0.2	0.2	0.2

Table 4. Laboratory-scale and upscaled input parameters for the numerical modeling
performed using hydro-thermo-mechanical modeling code LaMEM (Lithosphere and Mantle
Evolution Model).

589

590 The laboratory-scale Young's modulus and Poisson's ratio were upscaled using 591 empirical relationships for Young's modulus (Hoek and Diederichs, 2006) and Poisson's ratio 592 (Vásárhelyi, 2009) that account for rock-mass structure using the Geological Strength Index 593 (GSI), as described in Heap et al. (2020a). Following Heap et al. (2020a), GSI was assumed to 594 be 55 and the damage parameter in the Hoek-Diederichs equation was considered zero. The 595 cohesion and angle of internal friction were upscaled by transforming the generalized form of 596 the Hoek-Brown failure criterion to the Mohr-Coulomb failure criterion (as in Hoek et al., 597 2002). We used the mean uniaxial compressive strength of samples H18, H6, and H2B for layers V3, V2, and V1, respectively. The empirical fitting parameter m_i required for the 598 599 generalized Hoek-Brown failure criterion was determined from the triaxial data (Figure 4) using 600 RocData (Rocscience; https://www.rocscience.com). To do so, we again assumed a GSI of 55 601 and a damage parameter of zero. Since a depth is required to upscale the cohesion and angle of 602 internal friction, we selected a depth half of that between the summit of the volcano and the top 603 of the undeformable zone in the model (275 m). The upscaled model parameters for V3, V2, 604 and V1 are provided in Table 4. A more detailed description of the upscaling methods can be 605 found in the Supplementary Information. 606



607

608 Figure 6. Results of the large-scale 2D numerical modeling designed to better understand the 609 influence of alteration on the stability of La Soufrière de Guadeloupe Eastern (Caribbean, 610 France). Panels (a-d) show the model setup, in which white, light grey, grey, and dark grey 611 indicate non-deformable, slightly altered (V3), moderately altered (V2), and highly altered 612 (V1) zones. Panels (e-h) and (i-l) show the model results for the models run using laboratory-613 scale and upscaled mechanical properties, respectively. The color of the line representing the 614 surface of the volcano indicates the surface velocity (mm/yr) and the color inside the volcano indicates the strain rate (s⁻¹). 615 616

The model results, the velocity magnitude at the surface and the 2nd invariant of the deviatoric strain rate tensor, which we refer to as the strain rate, within the deformable domain, are presented in Figure 6. The strain rate allows us to understand whether the deformation is widespread or localized, and the velocity at the surface can be compared to geophysical data.
Figures 6e-h and 6i-l show model runs using the laboratory-scale and upscaled values,
respectively. We first highlight that our results emphasize the importance of using upscaled
values for large-scale modeling. Not only are the strain rates and velocities much lower when
using the laboratory-scale values, but the deformation also manifests slightly differently (Figure
625
6).

626 To discuss the influence of alteration, we will restrict our discussion to the upscaled 627 models (Figures 6i-1). We first note that strain rates and velocities are low in the homogenously 628 altered volcanoes, that deformation is largely restricted to the surface and the base of southern 629 slope, and that there is little difference in strain rates and velocities when the alteration is 630 increased from slightly to moderately altered (Figures 6i and 6j). When the highly-altered zone 631 guided by the recent electrical survey of Rosas-Carbajal et al. (2016) is included, the strain rates 632 and velocities increase significantly (Figure 6k). Surface velocities increase from < 0.01 to 633 ~ 0.05 mm/yr and a large listric, concave-upward sliding surface emerges, which stretches from 634 the top of the dome to near the base of the southern flank (Figure 6k). A localized slip surface 635 at the base of the deformable domain and two smaller slip surfaces also emerge near the base 636 of the southern flank (Figure 6k). Therefore, the inclusion of the highly altered zone, the 637 consequence of pervasive alteration resulting from the circulation of acid hydrothermal fluids 638 (Rosas-Carbajal et al., 2016), has significantly increased volcano deformation rates and 639 therefore decreased the stability of the volcano. As outlined above, recent manifestations of 640 increased hydrothermal activity, such as fumaroles and hot springs (Moretti et al., 2020; Jessop 641 et al., 2021), also provide evidence that the measured ground deformation is, in part, due to the 642 alteration of the rock forming the dome. Continuous and campaign Global Navigation Satellite 643 System (GNSS) measurements over ~ 20 years show that the dome is sliding at a rate of 0.3– 644 0.7 cm/yr (Moretti et al., 2020). Therefore, although our modeling captures the dynamics of the

deformation of the southern flank, our simplified 2D model setup (Figure 6) underestimates the deformation measured at the volcano. This underestimation is likely the result of the presence of large-scale faults, discontinuities, extremely weak clay-rich layers, and zones of high pore pressure, not included in our modeling. Indeed, the strain rate in the southern flank increases when we increase the pore fluid pressure ratio, λ_i , from 0.2 to 0.4 (see additional models presented in the Supplementary Information). Future models will aim to incorporate these features.

652 In a final step, and to further explore how alteration can influence volcano stability, we 653 modeled a scenario in which the highly altered zone identified by Rosas-Carbajal et al. (2016) 654 was artificially enlarged to incorporate the entire southern flank (Figure 6d). Increasing the size 655 of the alteration zone increases the surface velocities from ~0.05 to ~0.1 mm/yr and 656 significantly increases the strain rate within the flank (Figure 6l). The slip surface that at the 657 base of the deformable domain is now larger and has a higher strain rate, and the entire southern 658 flank is now characterized by higher strain rates (Figure 61). In addition, we also observe that 659 expanding the alteration zone likely increases the potential collapse volume (Figure 61). Based 660 on these models, increasing the size of the highly altered zone (i.e. to a size larger than the 661 present-day altered zone) will significantly increase the risk posed by the volcano. Deformation 662 snapshots from additional models, presented in the Supplementary Information, show the 663 progression of deformation for the present-day and increased-alteration scenarios from 5 to 100 664 kyrs (using upscaled physical and mechanical properties). These additional models also show 665 that increasing the extent of the alteration increases the rate of volcano spreading.

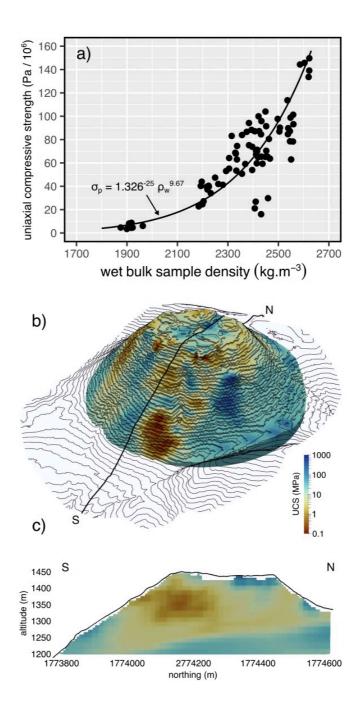
666

667 5.4 Three-dimensional strength model for La Soufrière de Guadeloupe

668 The results of the previous section highlight the importance of combining geophysical669 surveys, laboratory data, and large-scale modeling to evaluate the influence of alteration on

volcanic flank instability. Our 2D model geometry is based on qualitative interpretations from an electrical conductivity model to delimit different regions based on their degree of alteration. A quantitative approach would require a petrophysical relation to convert the conductivity values to rock strength and cohesion, but such relations are highly non-unique as electrical conductivity depends on many factors including clay content, fluid temperature, and composition. Instead, rock strength is controlled to a first order by porosity, as shown in Figure 3b, and thus to the bulk rock density.

677 Here, we combine a 3D model of the bulk density of the present-day lava dome (Rosas-Carbajal et al., 2017) with an empirical power-law function relating wet bulk density and 678 uniaxial compressive strength, σ_p , where $\sigma_p = 1.326^{-25} \rho_w^{9.67}$ (σ_p in Pa and ρ_w in kg/m³) to 679 680 create a 3D strength map of the volcano (Figure 7). The empirical function was specifically 681 fitted to our new data for La Soufrière (Table 2; Figure 7a). Because the distribution of liquid-682 saturated and dry zones within the volcano is currently poorly constrained, we chose to use the 683 bulk density of our samples saturated with liquid water, a scenario we consider better represents 684 the natural state of the volcano than a completely dry volcano. The 3D bulk density model of 685 Rosas-Carbajal et al. (2017) was obtained by jointly inverting gravity data and muon data 686 acquired using three simultaneous muon detectors that scanned the lava dome from different 687 points of view. Muon tomography is a novel technique, which uses the attenuation experienced 688 by cosmic muons as they pass through the volcano to estimate its density (Tanaka et al., 2010). 689 Whereas a single detector can provide 2D profiles of the average density of the volcano in the 690 different directions sampled, multiple simultaneous detectors from different points of view can 691 be used to create a 3D model of the volcano (Bonechi et al., 2020).



692

Figure 7. (a) Uniaxial compressive strength as a function of wet bulk sample density for
andesites from La Soufrière de Guadeloupe (Eastern Caribbean, France). Curve is the best-fit
empirical power-law function. (b) 3D uniaxial compressive strength map of La Soufrière de
Guadeloupe (Eastern Caribbean, France). The black line, which runs North-South, indicates
the position of the cross-section in panel (c). (c) North-South cross-section through La
Soufrière showing the distribution of uniaxial compressive strength. Color scale is the Roma

699

(seismic tomography) color scale of Fabio Crameri (Crameri et al., 2020). UCS – uniaxial compressive strength.

700 701

702 Figure 7 highlights that large portions of the dome are characterized by very low values 703 of strength (< 5 MPa). In particular, the southern flank of the volcano hosts a significant weak 704 zone (Figures 7b and 7c). This low-strength zone is coincident with the location of the main 705 hydrothermal reservoir at La Soufrière, as exposed by electrical conductivity measurements 706 (Lesparre et al., 2012; Rosas-Carbajal et al., 2016). Based on (1) the influence of 707 hydrothermally altered rock on edifice stability and spreading rates at La Soufrière revealed by 708 the numerical modeling in the previous section, and (2) the large volume of low-strength rock 709 in the La Soufrière dome highlighted by our 3D strength model, we conclude that the stability 710 of La Soufrière is compromised by its high degree of rock alteration, and that the rate of volcano 711 spreading and the probability of an eventual partial edifice collapse may increase solely due to 712 the ongoing alteration caused by circulating hydrothermal fluids, and may not necessarily 713 require intensified hydrothermal and/or magmatic activity.

714 Based on our 3D strength model, we can provide estimates for the volumes of rock 715 within the lava dome below a certain strength threshold. For example, we estimate that rock with a strength lower than 1 and 10 MPa comprises minimum dome volumes of 2.7×10^6 and 716 36×10^6 m³, respectively, corresponding to at least 4% and 55% of the dome above 1200 m. In 717 718 other words, more than half of the La Soufrière dome could be considered to have a low 719 strength. We highlight that these volume and percentage estimates should be considered 720 conservative due to the positioning of the three telescopes at La Soufrière, which provides better 721 coverage of the southern flank of the volcano (Rosas-Carbajal et al., 2017). It is likely, 722 therefore, that the volumes and percentages of weak rock within the dome at La Soufrière are 723 higher than estimated here.

Although the strength map shown in Figure 7 is currently simplistic (for example, it is well known that strength increases with depth in the brittle field; Heap et al., 2015b; Figure 4), this 3D model permits the quick identification of zones within the volcano that could promote instability and more experimental data, when available, will allow us to account for depth and other factors that can influence rock mechanical behavior, such as temperature and saturation state (Heap and Violay, 2021).

730

731 6 Conclusions

732 Hydrothermal alteration is often invoked as a mechanism that greatly contributes to 733 volcano instability (Reid et al., 2001; van Wyk de Vries et al., 2000). However, owing to the 734 paucity of experimental data and the difficulty in accurately imaging subsurface alteration 735 zones, the rock properties and the shape and size of the alteration zones required for large-scale 736 modeling are often poorly constrained or even assumed. Here we use a combination of 737 laboratory and geophysical data to numerically model the influence of hydrothermal alteration 738 on volcano stability, using La Soufrière de Guadeloupe as a case study. Our numerical modeling 739 suggests that hydrothermal alteration can significantly increase the deformation rate within the 740 flanks of the volcano, and also increase the potential collapse volume.

741 The volcano stability modeling and 3D strength map presented herein, firsts for La 742 Soufriere, help to validate the hypothesis forwarded by the numerous geophysical studies 743 performed at the volcano: that the identified hydrothermal reservoir characterized by altered 744 materials promotes volcano instability that could result in collapse (Bouligand et al., 2016; 745 Brothelande et al., 2014; Lesparre et al., 2012; Nicollin et al., 2006; Rosas-Carbajal et al., 2016, 746 2017). As discussed above, eventual partial edifice collapse at La Soufrière may not necessarily 747 require intensified hydrothermal and/or magmatic activity. Not only would the debris avalanche 748 resulting from partial or major dome collapse threaten the inhabited areas surrounding the 749 volcano (Le Friant, 2006; Peruzzetto et al., 2019), but collapse could also depressurize the 750 hydrothermal system leading to steam-driven eruptions, dangerous laterally-directed 751 explosions, and associated high energy pyroclastic density currents, similar to the activity 752 during the 1976–1977 volcanic crisis (Feuillard et al., 1983; Komorowski et al., 2005). We also 753 highlight that the fragmentation threshold (the overpressure required for complete 754 fragmentation upon depressurization), the proportion of fines produced as a result of 755 fragmentation, and the ejection speed are also influenced by hydrothermal alteration (Mayer et 756 al., 2015, 2016; 2017; Montanaro et al., 2016), reinforcing the need for a multidisciplinary 757 approach to unravel how hydrothermal alteration affects volcanic hazards (see also de Moor et 758 al., 2019; Mick et al., 2021).

759 Our modeling and 3D strength map therefore highlight the importance of monitoring the extent and evolution of hydrothermal alteration, and the formation of low-strength altered 760 761 layers, at La Soufrière and at other active volcanoes worldwide. Such monitoring can be 762 achieved, for example, using geophysical methods such as electrical tomography (Ahmed et 763 al., 2018; Byrdina et al., 2017; Ghorbani et al., 2018; Rosas-Carbajal et al., 2016) and muon 764 tomography (Lesparre et al., 2012; Rosas-Carbajal et al., 2017), near-surface seismic imaging 765 (Amoroso et al., 2018), thermal and gas monitoring (de Moor et al., 2019; Edmonds et al., 2003; 766 Jessop et al., 2021; Moretti et al., 2020; Tamburello et al., 2019), geological mapping (van Wyk 767 de Vries et al., 2000), deformation monitoring (Moretti et al., 2020), magnetic methods (Finn 768 et al., 2007), and remote and/or ground-based optical and spectroscopic methods (Crowley and 769 Zimbelman, 1997; Darmawan et al., 2018; John et al., 2008; Kereszturi et al., 2020; Mueller et 770 al., 2021). Further, although we document a reduction in strength as a function of alteration 771 (Figure 5c), alteration can also increase strength and potentially promote volcanic instability by 772 creating zones of high pore fluid pressure (Heap et al., 2021a; Reid, 2004). As noted in Heap 773 et al. (2021a), it is important not only to map the extent and evolution of hydrothermal

alteration, but also the locations in the volcano prone to porosity increasing and porositydecreasing alteration.

776 We have also shown that microtextural and mineralogical analysis of host-rock material 777 erupted during phreatic explosive eruptions, as well as material from landslides and debris-778 avalanche deposits, can provide highly valuable insights into the nature, extent, and dynamics 779 of alteration processes and fluid circulation in the pre-collapse edifice (see also Boudon et al., 780 1998; Komorowski et al., 2010; Salaün et al., 2011). Hence, we highlight that it is also important 781 to quantify the mineral content of new material exposed by landslides and collapses, as well as 782 their deposits, and to continuously monitor fluid compositions and water table fluctuations 783 (Hurwitz et al., 2003) with respect to the geometry of weakened low-strength structures that are characteristic of the internal framework volcanic edifices hosting hydrothermal systems. A 784 785 better understanding of the spatio-temporal evolution of hydrothermal alteration, and how it 786 can influence volcano stability using large-scale modeling, will help improve the mitigation of 787 volcanic hazards at active volcanoes worldwide.

788

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