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1 The thermal properties of hydrothermally altered andesites from La
2 Soufrière de Guadeloupe (Eastern Caribbean)

3

4 **Michael J. Heap^{1,2*}, David E. Jessop^{3,4,5}, Fabian B. Wadsworth⁶, Marina Rosas-Carbajal³,**
5 **Jean-Christophe Komorowski³, H. Albert Gilg⁷, Nadège Aron¹, Margaux Buscetti¹, Laura**
6 **Gential¹, Margaux Goupil¹, Mathilde Masson¹, Lucie Hervieu¹, Alexandra R.L. Kushnir¹,**
7 **Patrick Baud¹, Lucille Carbillet¹, Amy G. Ryan⁸, and Roberto Moretti^{3,4}**

8

9 ¹ *Université de Strasbourg, CNRS, Institut Terre et Environnement de Strasbourg, UMR 7063,*
10 *5 rue Descartes, Strasbourg F-67084, France*

11 ² *Institut Universitaire de France (IUF), Paris, France*

12 ³ *Université de Paris, Institut de Physique du Globe de Paris, CNRS UMR 7154, F-75005 Paris,*
13 *France*

14 ⁴ *Observatoire Volcanologique et Sismologique de Guadeloupe, Institut de Physique du Globe*
15 *de Paris, F-97113 Gourbeyre, France*

16 ⁵ *CNRS, IRD, OPGC Laboratoire Magmas et Volcans, Université Clermont Auvergne, F-63000,*
17 *Clermont-Ferrand, France*

18 ⁶ *Earth Science, Durham University, Science Labs, Durham, DL1 3LE, United Kingdom*

19 ⁷ *Engineering Geology, TUM School of Engineering and Design, Technical University of*
20 *Munich, 80333 Munich, Germany*

21 ⁸ *Department of Earth and Environmental Sciences, University of Minnesota, MN 55455, USA*

22

23 *Corresponding author: Michael Heap (heap@unistra.fr)

24

25 **Abstract**

26 The heat flux of an active volcano provides crucial information on volcanic unrest. The
27 hydrothermal activity often responsible for volcanic unrest can be accompanied by an increase
28 in the extent and intensity of hydrothermal alteration, which could influence the thermal
29 properties of the volcanic edifice. Therefore, an understanding of the influence of alteration on
30 the thermal properties of rocks is required to better interpret volcano heat flux data. We provide
31 laboratory measurements of thermal conductivity, thermal diffusivity, and specific heat
32 capacity for variably altered (intermediate to advanced argillic alteration) andesites from La
33 Soufrière de Guadeloupe (Eastern Caribbean). We complement these data with previously
34 published data for altered basaltic-andesites from Merapi (Indonesia) and new data for altered
35 rhyodacites from Chaos Crags (USA). Our data show that thermal conductivity and thermal
36 diffusivity decrease as a function of increasing porosity, whereas the specific heat capacity does
37 not change systematically. Thermal conductivity decreases as a function of alteration (the
38 percentage of secondary minerals) for the rocks from La Soufrière and Merapi (from ~ 1.6 to
39 $\sim 0.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ as alteration increases from ~ 1.5 to >75 wt.%), but increases for the rocks from
40 Chaos Crags (from ~ 1.1 to $\sim 1.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ as alteration increases from ~ 6 to ~ 15 wt.%).
41 Although the thermal diffusivity of the rocks from Chaos Crags increases from ~ 0.65 to
42 $\sim 0.75\text{--}0.95 \text{ mm}^2\cdot\text{s}^{-1}$ as alteration increases from ~ 6 to ~ 15 wt.%, the thermal diffusivity of the
43 rocks from La Soufrière and Merapi does not appear to be greatly influenced by alteration. The
44 specific heat capacity is not significantly affected by alteration, although there is a slight trend
45 of increasing specific heat capacity with alteration for the rocks from La Soufrière. We conclude
46 that the decrease in thermal conductivity as a function of alteration in the rocks from La
47 Soufrière and Merapi is the result of the low conductivity of the secondary mineral assemblage,
48 and that a combination of the high thermal conductivity of cristobalite and the reduction in
49 porosity as a result of the void-filling mineral precipitation can explain the increase in thermal
50 conductivity in the rocks from Chaos Crags. Calculations show that an increase in alteration of

51 a dome or edifice can result in decreases and increases in heat flow density, depending on the
52 type of alteration. Therefore, alteration-induced changes in the thermal properties of dome or
53 edifice rocks should be considered when interpreting volcano heat flux data. We conclude that
54 it is important not only to monitor the extent and evolution of alteration at active volcanoes, but
55 also the spatial distribution of alteration type.

56

57 **Keywords:** heat flux; alteration; porosity; thermal conductivity; thermal diffusivity; specific
58 heat capacity; Merapi; Chaos Crags

59

60 **Highlights:**

- 61 • Hydrothermal alteration changes the thermal properties of volcanic rocks.
- 62 • Alteration can increase or decrease thermal properties, depending on alteration type.
- 63 • Alteration can change the conductive heat flow density at a volcano.
- 64 • Alteration should be considered when interpreting volcano heat flux data.

65

66 **1 Introduction**

67 Active volcanoes, and particularly those with very active hydrothermal systems, emit a
68 prodigious amount of heat (Wright and Flynn, 2004). The heat from magma at depth is
69 transported to the surface by a combination of mechanisms, including the movement of magma,
70 the advection (e.g., by convection) of hot fluids, and the conduction of heat through rock.
71 Because changes in the heat emitted by a volcano can indicate the movement of magma and/or
72 modifications to the hydrothermal system, monitoring heat flux at active volcanoes provides
73 crucial information on volcanic activity and unrest (Jessop et al., 2021). Indeed, heat flux has
74 been measured, or is continuously monitored, at volcanoes worldwide (Dehn et al., 2001;
75 Wright et al., 2004), including Whakaari (New Zealand; Bloomberg et al., 2014), Vulcano

76 (Italy; Mannini et al., 2019), Campi Flegrei (Italy; Chiodini et al., 2005), Kīlauea (USA; Harris
77 et al., 2001), Mt Etna (Italy; Harris et al., 1997), Volcán de Colima (Mexico; Stevenson and
78 Varley, 2008), Lascar (Chile, Wooster and Rothery, 1997), and La Soufrière de Guadeloupe
79 (Eastern Caribbean; Jessop et al., 2021). The link between the thermal output of a volcano and
80 volcanic activity was recently emphasised by Girona et al. (2021), who highlighted that large-
81 scale thermal unrest was a reliable indicator of impending magmatic and phreatic eruptions.

82 The thermal properties of volcanic rocks comprising the edifice or dome, and processes
83 that increase or decrease these properties, are therefore informative for those charged with
84 interpreting surficial heat flux data at volcanoes. Furthermore, the thermal properties of
85 volcanic rocks are also an important input parameter in a wide range of models, such as those
86 designed to model heat loss from magma chambers, conduits, sills, dykes, lavas, and
87 ignimbrites (Irvine, 1970; Norton and Knight, 1977; Carrigan, 1984; Bruce and Huppert, 1989;
88 Carrigan et al., 1992; Fialko and Rubin, 1999; Wooster et al., 1997; Annen et al., 2008; Nabelek
89 et al., 2012; Heap et al., 2017a; Annen, 2017; Mattsson et al., 2018; Tsang et al., 2019). In
90 general, for simplicity, the thermal properties of rocks in and around volcanoes are often taken
91 to be constant, which facilitates calculations of heat flow and how it changes with, for example,
92 hydrothermal circulation or magma movement. And yet, laboratory data show that thermal
93 properties are variable with porosity (e.g., Robertson and Peck, 1974) and temperature (e.g.,
94 Whittington et al., 2009).

95 For the reasons given above, there is a clear outstanding need to better understand the
96 evolution of thermal properties as other rock characteristics change. This endeavour is well
97 suited to laboratory investigation in which the thermal properties of volcanic rocks are
98 constrained (Horai et al., 1970; Fujii and Osako, 1973; Robertson and Peck, 1974; Bagdassarov
99 and Dingwell, 1994; Whittington et al., 2009; Romine et al., 2012; Mielke et al., 2015, 2016,
100 2017; Vélez et al., 2018; Heap et al., 2020a; Weydt et al., 2021). For example, the thermal

101 conductivity of variably porous basalt from Hawai'i (USA) was found to decrease from ~ 1.7
102 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a porosity ~ 0.05 down to $\sim 0.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a porosity of ~ 0.85 (Robertson and
103 Peck, 1974). The average thermal conductivities of andesite (average porosity of 0.095) and
104 rhyolite lava (average porosity of 0.275) from the Tauhara geothermal field (New Zealand)
105 were measured to be 1.32 and 1.11 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively (Mielke et al., 2015), and the
106 thermal conductivities of andesite (porosity of 0.023–0.130), dacite (porosity of 0.108), and
107 rhyolite (porosity of 0.231) from the Taupō Volcanic Zone (New Zealand) were measured to
108 be 1.19–1.70, 1.18, and 1.04 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively (Mielke et al., 2016). More recently,
109 Heap et al. (2020a) measured the thermal properties (thermal conductivity, thermal diffusivity,
110 and specific heat capacity) of variably porous andesites from Mt Ruapehu (New Zealand) and
111 variably altered basaltic-andesites from Merapi volcano (Indonesia). The thermal conductivity
112 of low-porosity (~ 0.05) samples from Mt Ruapehu was measured to be between ~ 1.4 and ~ 1.6
113 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, but decreased to $\sim 0.4 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a porosity of ~ 0.6 (Heap et al., 2020a). These
114 authors also found that saturation with water increased the bulk specific heat capacity and
115 thermal conductivity, and decreased the thermal diffusivity, relative to the dry state. Thermal
116 property measurements on samples from Merapi volcano suggested that hydrothermal
117 alteration decreases thermal conductivity and thermal diffusivity, and increases specific heat
118 capacity (Heap et al., 2020a).

119 Despite these laboratory studies, and others, our understanding of the thermal properties
120 of volcanic rocks stands to benefit from new laboratory data. In particular, little is known about
121 the influence of hydrothermal alteration on the thermal properties of volcanic rocks. However,
122 an understanding as to how hydrothermal alteration can influence thermal properties is
123 important. This is because increasing volcanic unrest resulting in an elevated heat flux, which
124 is linked to impending magmatic and phreatic eruptions (Girona et al., 2021), is often associated
125 with a visible increase in alteration (Jessop et al., 2021). Here, we report findings from a

126 laboratory study designed to better understand the influence of hydrothermal alteration on the
127 thermal properties of volcanic rocks, using La Soufrière de Guadeloupe as a case study volcano.
128

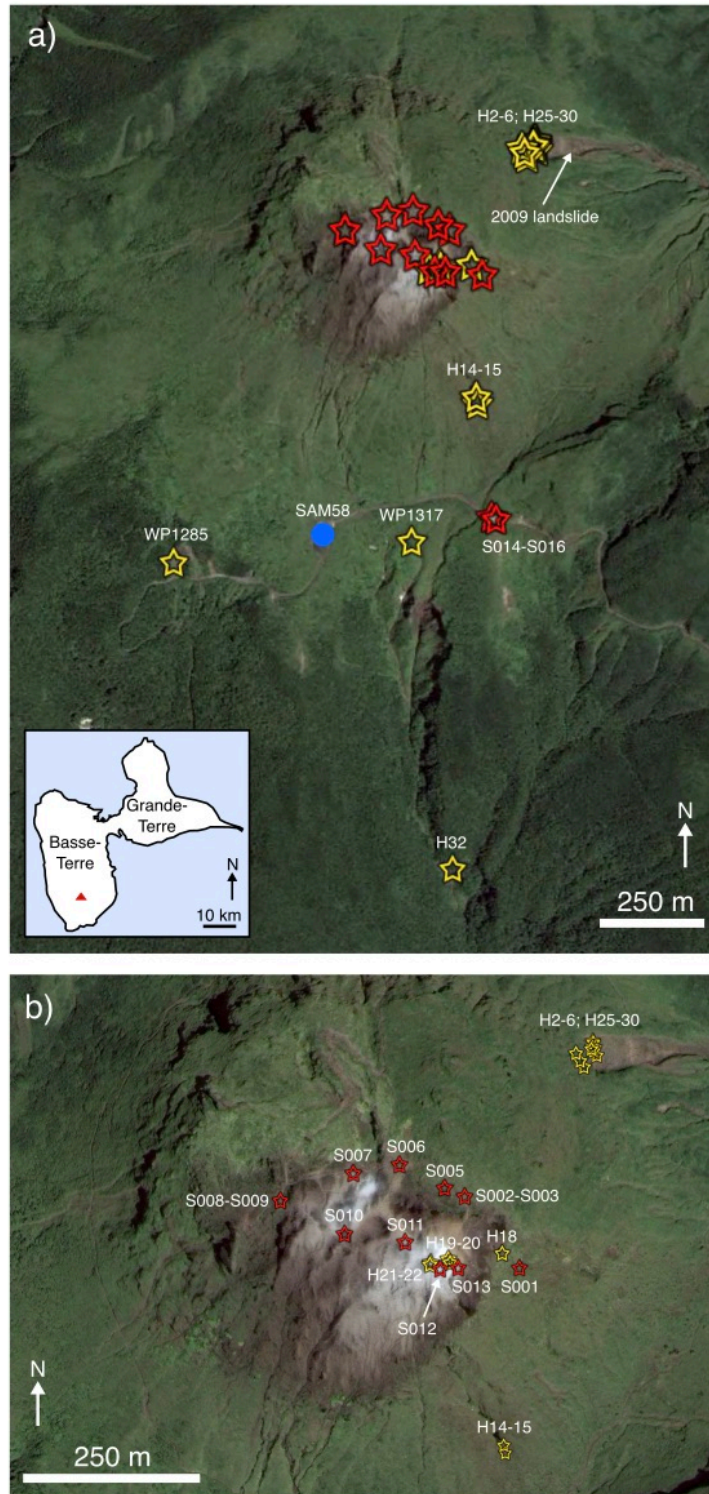
129 **2 La Soufrière de Guadeloupe (Eastern Caribbean)**

130 La Soufrière de Guadeloupe (hereafter called La Soufrière) is an active andesitic
131 stratovolcano located on the island of Guadeloupe in the Eastern Caribbean (Figure 1a). There
132 have been six non-magmatic phreatic or hydrothermal eruptions at La Soufrière since 1635 CE,
133 the largest and most-recent eruption of which occurred in 1976–1977 (Komorowski et al.,
134 2005). The 1976–1977 volcanic crisis resulted in the evacuation of ~70,000 people and severe
135 socio-economic consequences for the island of Guadeloupe (Feuillard et al., 1983; Hincks et
136 al., 2014; Komorowski et al., 2005, 2015). Volcanic unrest has been increasing at La Soufrière
137 since 1992, manifest as (1) the formation of new fumaroles and an expansion of the outgassing
138 area on top of the lava dome (Brombach et al., 2000; Komorowski et al., 2005; Villemant et al.,
139 2014; Allard et al., 2014; Moretti et al., 2020), (2) the appearance of high-flow-rate thermal
140 acid sulfate-chloride springs on the slopes and at the base of the dome (Villemant et al., 2005;
141 2014), (3) an increase in the heat output from the dome (Gaudin et al., 2013; Jessop et al., 2021),
142 (4) an increase in seismic activity (hundreds of shallow, low-magnitude earthquakes each
143 month) and the largest felt volcano-tectonic earthquake since 1976–1977 (Moretti et al., 2020),
144 and (5) large flank displacements (Moretti et al., 2020; Heap et al., 2021a).

145 Activity within the shallow hydrothermal system, which has been imaged by a variety
146 of geophysical methods (Nicollin et al., 2006; Brothelande et al., 2014; Bouligand et al., 2016;
147 Rosas-Carbajal et al., 2016), is considered responsible for much of the recent volcanic unrest
148 (Moretti et al., 2020; Heap et al., 2021a). Indeed, La Soufrière has a history of unrest associated
149 with hydrothermal activity. Not only were hydrothermally altered rocks ejected from the vent
150 during the 1976–1977 crisis (Feuillard et al., 1983), but geological evidence have also suggests

151 that historic partial edifice collapses at La Soufrière were associated with hydrothermal activity,
152 owing to the abundance of hydrothermally altered materials in debris avalanche deposits
153 (Boudon et al., 1987; Komorowski et al., 2005; Le Friant et al., 2006; Salaün et al., 2011; Rosas-
154 Carbajal et al., 2016; Peruzzetto et al., 2019). Because the tropical climate of the island of
155 Guadeloupe (i.e. high yearly precipitation) contributes to dome washout and the shallow
156 hydrothermal regime, La Soufrière represents an ideal natural laboratory to study the influence
157 of hydrothermal alteration on the thermal properties of a nearly fluid-saturated volcanic dome.
158 Further, and importantly, such data are timely due to the link between magmatic and phreatic
159 eruptions and the increase in heat output associated with increased hydrothermal activity at the
160 volcano (Moretti et al., 2020; Jessop et al., 2021; Girona et al., 2021).

161



162

163 **Figure 1.** (a) Map of La Soufrière de Guadeloupe (Eastern Caribbean; taken from Google

164 Earth®) showing the sampling locations for the 19 rock blocks of the main sample suite

165 (yellow stars; the “H” series), the 3 rock blocks and 13 bags of unlithified material from the

166 second field campaign (red stars; the “S” series), and the borehole rock sample (blue circle;

167 SAM58). Inset shows a map of Guadeloupe in which the location of La Soufrière de
168 Guadeloupe is indicated by a red triangle. (b) Zoom of the dome summit area showing the
169 sampling sites on the dome (also taken from Google Earth®).

170

171 **3 Materials and Methods**

172 Materials (rock blocks and unlithified deposits) were collected over two field campaigns
173 conducted at La Soufrière in 2019. The main sample suite of 19 rock blocks were collected
174 during the first field campaign (“H” series), which were supplemented by 3 rock blocks and 13
175 bags of unlithified material collected during the second campaign (“S” series). The sampling
176 locations are provided in Figure 1. Our aim was to sample a range of materials from different
177 locations on the volcano that best represent the observed variability in porosity and
178 hydrothermal alteration.

179 Of the 19 rock blocks from the main sample suite, the “H” series, nine blocks were taken
180 from the collapse scar of the 2009 landslide (H2A, H2B, H3, H4A, H5A, H6, H25, H29, and
181 H30). Five blocks were collected from the dome summit: four blocks were taken from the lava
182 spines of the 1530 CE dome (two blocks from *Cratère Sud Central*, H19 and H20, and two
183 blocks from an adjacent site, H21 and H22), and one block was taken from the *Lacroix*
184 *Supérieure* outgassing fracture (H18). We also collected blocks from the West wall of the fault
185 *Faille 30 août* (H14 and H15), the collapse scar of the landslide triggered by the 21 November
186 2004 *Les Saintes* magnitude Mw 6.3 regional earthquake (Feuillet et al., 2011) (WP1285), and
187 adjacent to the *Galion* waterfall (H32). The final block, a volcanic bomb from the 1976–1977
188 eruption, was taken from the roof of a small disused thermal bathhouse to the South of the dome
189 (WP1317).

190 Samples in the “S” series were mostly tephra (millimetric granular matter in an argillic
191 matrix). Out of the 16 samples collected, only S005, S008, and S016 were rock blocks. S001

192 was taken from just below the summit plateau to the East of the dome. Notably, this location is
193 upwind of the predominant wind direction and thus the sample was relatively uncontaminated
194 by fumarolic gases. Samples S002–S006 were taken from within the thermally-active zone
195 (called “ZFNN” in Jessop et al., 2021), but from locations mostly unaffected by strong
196 outgassing. S007 was taken directly downwind and from the rim of the *Tarrisan* crater. Samples
197 S008–S011 were taken from various sites downwind of the main outgassing sites, and samples
198 S012 and S013 were collected in proximity to the main outgassing vents. Finally, samples
199 S014–S016 were taken from the base of the volcano, close to a site with low-level, passive
200 degassing and highly-altered soils due to historical high-activity. An additional rock sample
201 was taken from a borehole (SAM58) located at the *Savane à Mulets* carpark on the southwest
202 base of the dome (Figure 1a). This sample was taken from a depth of ~58 m.

203 Multiple cylindrical samples were cored from each of the rock blocks (from the “H” and
204 “S” series) to a diameter of 20 mm, and then cut and precision-ground to a nominal length of
205 40 mm. The samples were washed and then dried in a vacuum-oven at 40 °C for at least 48 h.
206 The connected porosity of each sample was calculated using the bulk sample volume and the
207 skeletal (solid) sample volume measured by a helium pycnometer. The cylindrical samples
208 prepared from each block were then grouped into pairs of similar porosity. For most blocks,
209 three pairs of samples were measured. For certain blocks (H15, H30, SAM58, S005, S008, and
210 S016), which were smaller, only two pairs of samples were measured.

211 The unlithified materials were first sieved to a grain diameter < 2 mm and then dried in
212 a vacuum-oven at 40 °C for at least 48 h. The solid density of each unlithified sample was then
213 measured using the mass and volume, measured by a helium pycnometer, of an aliquot of the
214 oven-dry powder.

215 The thermal conductivity, λ (in units of $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$), and thermal diffusivity, D (in units
216 of $\text{mm}^2\cdot\text{s}^{-1}$), were measured using a Hot Disk® TPS 500 Thermal Constants Analyser using the

217 transient plane source method (Gustafsson, 1991; Gustavsson et al., 1994; Harlé et al., 2019;
218 Heap et al., 2020a). The transient plane source method is a periodic method to measure the
219 thermal properties of materials (Hofmeister, 2019). The standard uncertainties for thermal
220 conductivity and thermal diffusivity values using the transient hot-strip method, arising from
221 contact losses and ballistic radiative transfer gains (Hofmeister, 2019), were determined to be
222 2.6 and 11%, of the measured values respectively (Hammerschmidt and Sabuga, 2000).

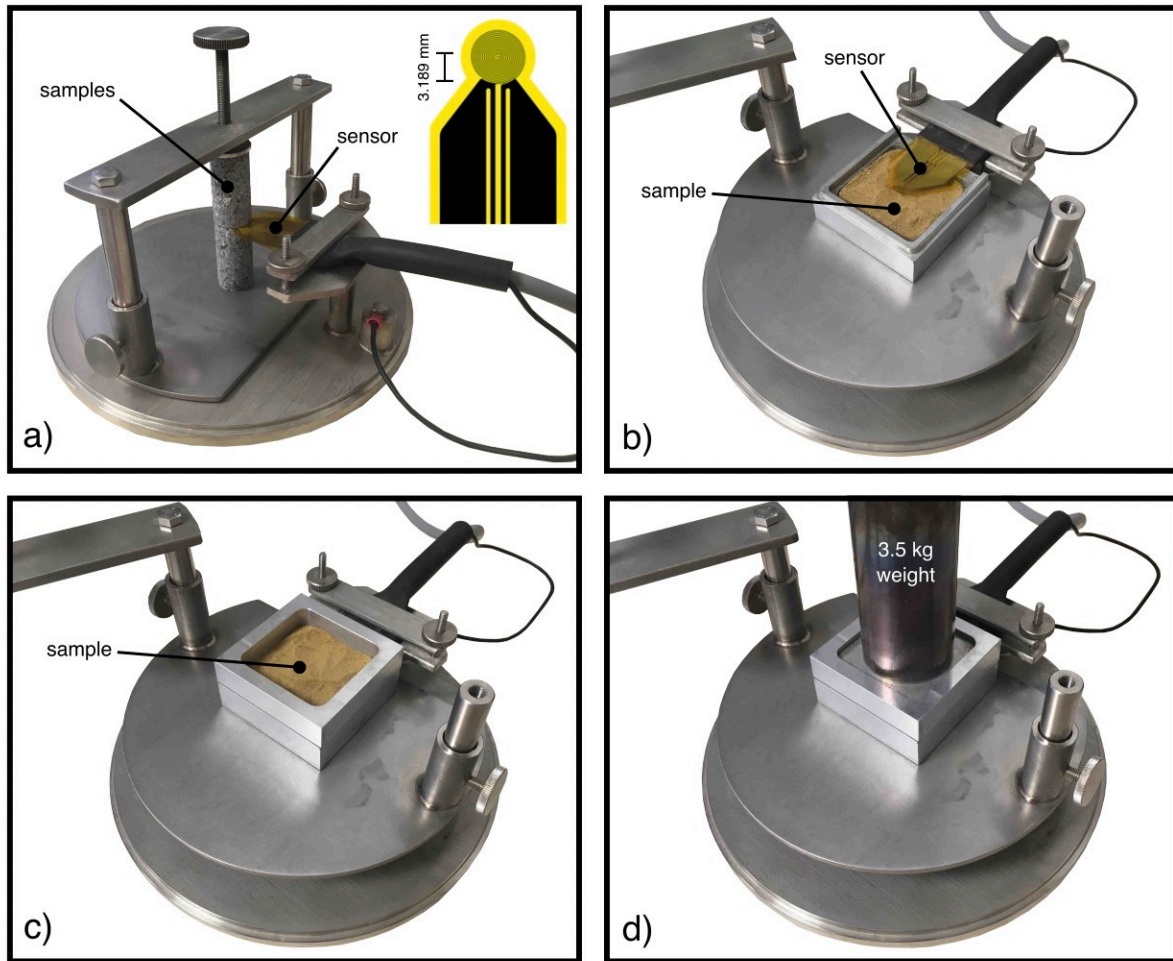
223 Thermal conductivity and thermal diffusivity were measured using a sensor consisting
224 of two 10 μm -thick nickel foil spirals (radius of 3.189 mm) that are encased and insulated by
225 30 μm -thick kapton (see inset on Figure 2a). For the rock samples, the sensor was sandwiched
226 between two cylindrical samples, of similar porosity, cored from the same block (Figure 2a).
227 The rock samples were held in place using a screw positioned at the top of the sample jig (Figure
228 2a), which ensured good contact between the sensor and the surface of the samples.

229 The unlithified samples were measured using a sample holder of known volume
230 supplied by Hot Disk®. Powder was first spooned into the lower part of the sample holder, and
231 the sensor was placed on top of the powder (Figure 2b). The top part of the sample holder was
232 then placed on top of the lower part, and more powder was spooned on top of the sensor (Figure
233 2c). A plate was then placed on top of the sample assembly, and the entire sample assembly
234 was compacted using a 3.5 kg weight (Figure 2d). The 3.5 kg weight ensured (1) a similar
235 compaction from sample to sample and (2) a good contact between the powder and the sensor.

236 The temperature adjacent to the sample, measured using a thermocouple, was inputted
237 into the Thermal Constants Analyser prior to starting each measurement. An electrical current
238 of known power and duration was passed through the sensor during the measurement, which
239 also recorded the increase in sample temperature as a function of time. The output power and
240 duration required for a reliable measurement varied from sample to sample and were found
241 using trial-and-error. The output power and test duration were typically 100–200 mW and 5–

242 10 s, respectively. Four consecutive measurements were performed on each pair of samples (on
243 the four different combinations of sample end-faces) and each powder, and we report herein an
244 average of these four measurements. Each measurement was performed at least five minutes
245 apart to ensure that the sample had cooled back to the ambient temperature. The sensor
246 measured the temperature drift of the sample for 40 seconds prior to each measurement to check
247 whether the sample was in thermal equilibrium. If the sample temperature was not constant
248 during this 40 second period, the data were not considered and the measurement was repeated.
249 Following each set of measurements on the powder samples, the mass of the powder was
250 measured to provide the bulk sample density and, using the solid density of each powder, their
251 total porosities were calculated. The volumetric heat capacity, $\rho_b c_p$ (in units of $\text{J}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$),
252 calculated by the Hot Disk® device, was divided by the independently determined bulk sample
253 density, ρ_b , to provide the bulk sample specific heat capacity, c_p (in units of $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$). All
254 measurements were conducted in a far-field environment that was at ambient laboratory
255 temperature and pressure.

256



257

258 **Figure 2.** (a) Photograph of the experimental setup for the rock-rock measurements.

259 Insets show the sensor used, consisting of two 10 μm -thick nickel foil spirals (radius of 3.189

260 mm) that are encased and insulated by 30 μm -thick kapton (used for all measurements). The

261 screw on the top of the setup ensures a good contact between the sensor and the samples.

262 Panels (b), (c), and (d) are photographs that show the procedure for measuring a powder

263 sample. (b) The powder was first spooned into the lower part of the holder, underneath the

264 sensor. (c) The upper part of the holder was placed onto the lower part and powder was

265 spooned over the sensor. (d) The top of the holder (a flat piece) was placed on top of the

266 powder and a 3.5 kg weight was placed on top of the setup to ensure reproducible

267 compaction and a good contact between the sensor and the powder.

268

269 Polished thin sections were prepared from offcuts of the main sample suite (the “H”
270 series) for microstructural analysis using a Tescan Vega 2 XMU scanning electron microscope
271 (SEM). The mineral contents of 17 of the 19 blocks from the “H” series, collected during the
272 first field campaign, were quantified using X-ray powder diffraction (XRPD) in Heap et al.
273 (2021a). We provide here the mineral contents of the two remaining samples from this sample
274 suite (samples H30 and H32), using the same technique described in Heap et al. (2021a). To do
275 so, powdered offcuts of the core material were ground for 8 min with 10 ml of isopropyl alcohol
276 in a McCrone Micronising Mill using ZrO₂ cylinder elements. The XRPD analyses were
277 performed on side-loaded powder mounts using a Bruker D8 Advance Eco X-ray diffractometer
278 (CuK α , 40 kV, 25 mA, 2–75° 2 θ , 0.01° step size, 15 mm irradiated length, 2.5° primary and
279 secondary sollers, and a LynxEye XE-T detector). The phases in the whole rock powders were
280 then quantified using the Rietveld program BGMN (Bergmann et al., 1998) and the Profex
281 graphical user interface (Döbelin and Kleeberg, 2015). To identify clay minerals, < 2 μ m
282 fractions were separated by gravitational settling, and oriented mounts were X-rayed in an air-
283 dried state, an ethylene-glycolated state, and following exposure to 550 °C. Selected mineral
284 phases were additionally identified by micro-Raman spectroscopy using a Horiba Jobin Yvon
285 XploRA PLUS confocal Raman microscope. The spectrometer was equipped with a frequency-
286 double Nd:YAG laser (532 nm, with a maximum power of 22.5 mW) and an Olympus
287 LMPLFLN 100 \times long-working-distance objective with a numerical aperture of 0.9.

288

289 **4 Results**

290 4.1 Microstructure, mineralogy, and alteration

291 The mineral contents for each of the 19 rock blocks from the main sample suite (“H”
292 series) are available in Table 1 (data for all but samples H30 and H32 were presented in Heap
293 et al., 2021a). All of the andesite blocks are characterised by a porphyritic texture comprising

294 phenocrysts (often a few hundred microns long, but occasionally as large as 1–2 mm) of
 295 dominantly plagioclase and pyroxene (orthopyroxene and clinopyroxene) within a crystallised
 296 groundmass (Figures 3 and 4; Heap et al., 2021a). Microcracks and pores (with a diameter
 297 ranging from a couple of tens of microns to almost 1 mm) are present in all of the samples
 298 (Figures 3 and 4; Heap et al., 2021a).

299 All of the samples also contain variable quantities of secondary minerals, including
 300 kaolinite, alunite or natro-alunite, silica polymorphs (quartz, cristobalite, tridymite, and opal-
 301 A), hematite, pyrite, gypsum, and talc (secondary minerals are indicated by asterisks in Table
 302 1). The most abundant secondary minerals are kaolinite, natro-alunite, and cristobalite (Table
 303 1). Kaolinite is present as a replacement mineral in altered plagioclases and as a microcrack-
 304 and pore-filling precipitate, and natro-alunite and cristobalite are present as microcrack- and
 305 pore-filling precipitates (Figures 3 and 4). The observed secondary mineral assemblage
 306 corresponds to intermediate to advanced argillic alteration (Heap et al., 2021a).

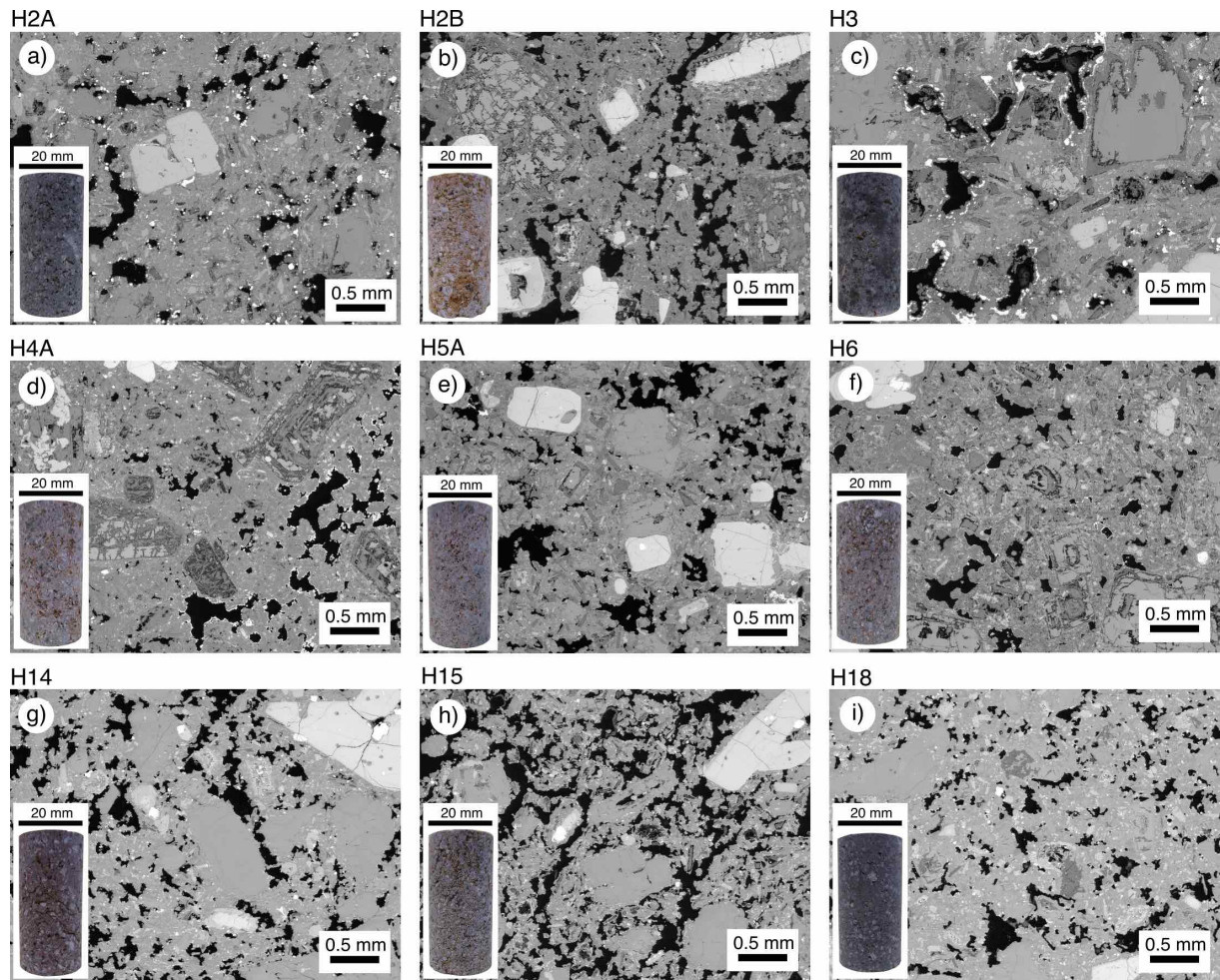
307 Based on the results of the XRPD, each of the 19 rock blocks from the “H” series are
 308 assigned a value of alteration—the weight percentage of secondary minerals in the block—
 309 which ranges from 6 wt.% (sample H32) up to 85.4 wt.% (sample H30) (Table 2).

310

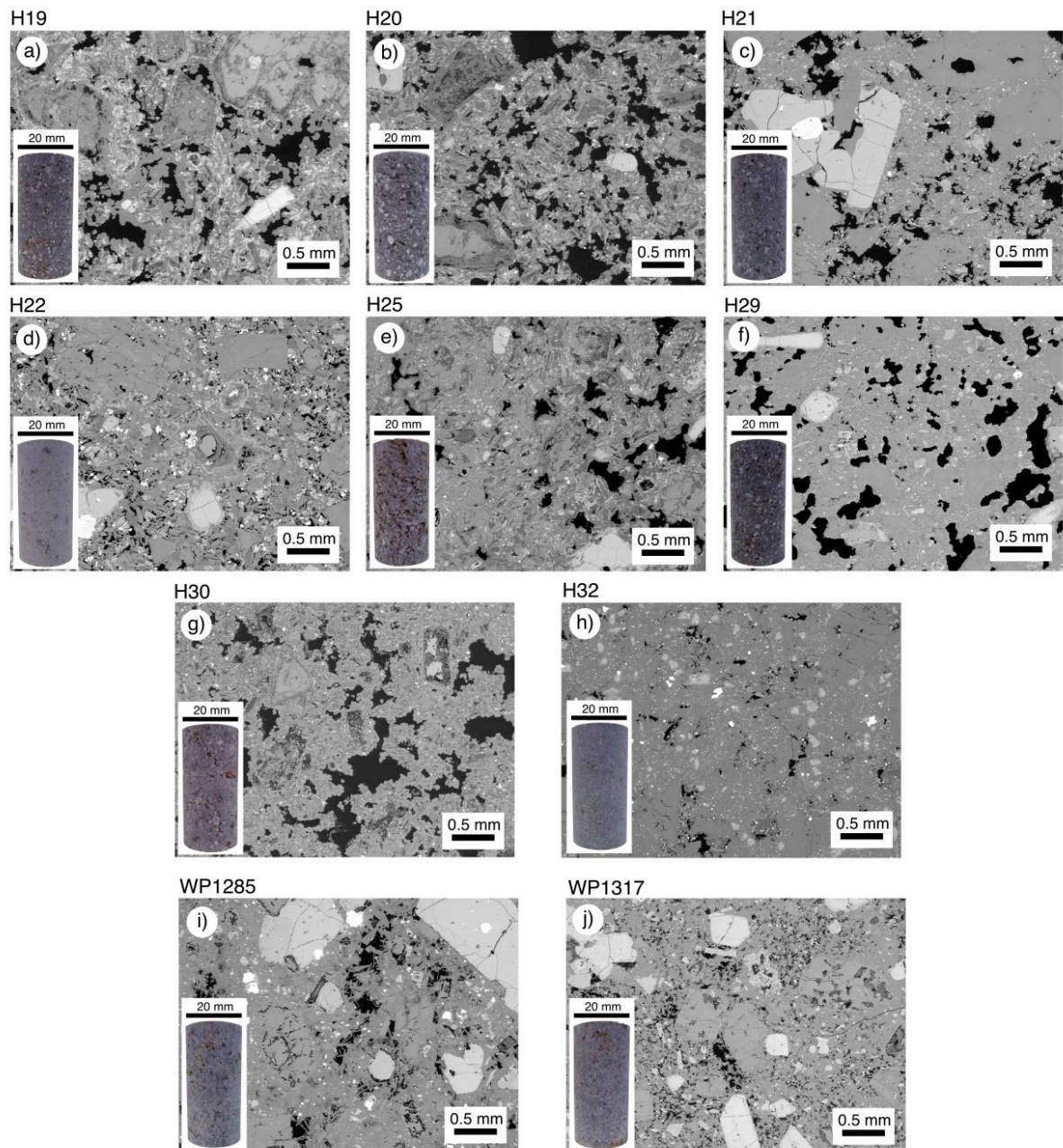
| Mineral | H2 A | H2 B | H3 | H4 A | H5 A | H6 | H1 4 | H1 5 | H1 8 | H1 9 | H2 0 | H2 1 | H2 2 | H2 5 | H2 9 | H3 0 | H3 2 | WP128 5 | WP131 7 |
|-----------------|---------|---------|------|---------|---------|------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|------------|------------|
| 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 | 8.9 | 64.4 | 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 | 2.5 | 9.5 | 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 | 3.3 | 15.1 | 13.2 | 15.6 |
| (Ti-) Magnetite | 0.7 | - | 0.8 | - | - | - | 0.8 | - | 2.9 | - | 2.4 | 3.1 | 0.8 | - | 2.7 | - | 4.9 | 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 | 0.9 | 0.3 | 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 | 9 | 5.7 | - | - |
| Tridymite* | - | - | - | - | - | - | - | 0.7 | - | - | - | - | - | - | - | - | - | 13.2 | 13.2 |
| Hematite* | - | - | - | - | - | - | 3.4 | - | 2.8 | 2.4 | - | - | - | - | 3.1 | 4.3 | - | - | - |
| 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 | - | 25.6 | - | - | - |
| 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 | - | 35.6 | - | - | |
| Talc* | - | - | - | - | - | - | - | - | - | - | - | - | 2.9 | - | - | - | - | - | - |
| Opal-A* | - | - | - | - | - | - | - | - | 33.0 | 30.0 | 25.0 | - | - | - | 10.0 | 10 | - | - | - |

311

312 **Table 1.** Mineral contents, measured by X-ray powder diffraction, of the 19 rock blocks from
313 the main sample suite (“H” series). Values in wt.%. Asterisk denotes a secondary mineral (i.e.
314 alteration mineral). All data, except those for H30 and H32, were taken from Heap et al.
315 (2021a).
316



317
318 **Figure 3.** Backscattered scanning electron microscope images of andesites from La Soufrière
319 de Guadeloupe (Eastern Caribbean). Insets show a photograph of 20 mm-diameter sample
320 prepared from the block. (a) H2A. (b) H2B. (c) H3. (d) H4A. (e) H5A. (f) H6. (g) H14. (h)
321 H15. (i) H18.
322



323

324 **Figure 4.** Backscattered scanning electron microscope images of andesites from La Soufrière

325 de Guadeloupe (Eastern Caribbean). Insets show a photograph of 20 mm-diameter sample

326 prepared from the block. (a) H19. (b) H20. (c) H21. (d) H22. (e) H25. (f) H29. (g) H30. (h)

327

H32. (i) WP1285. (j) WP1317.

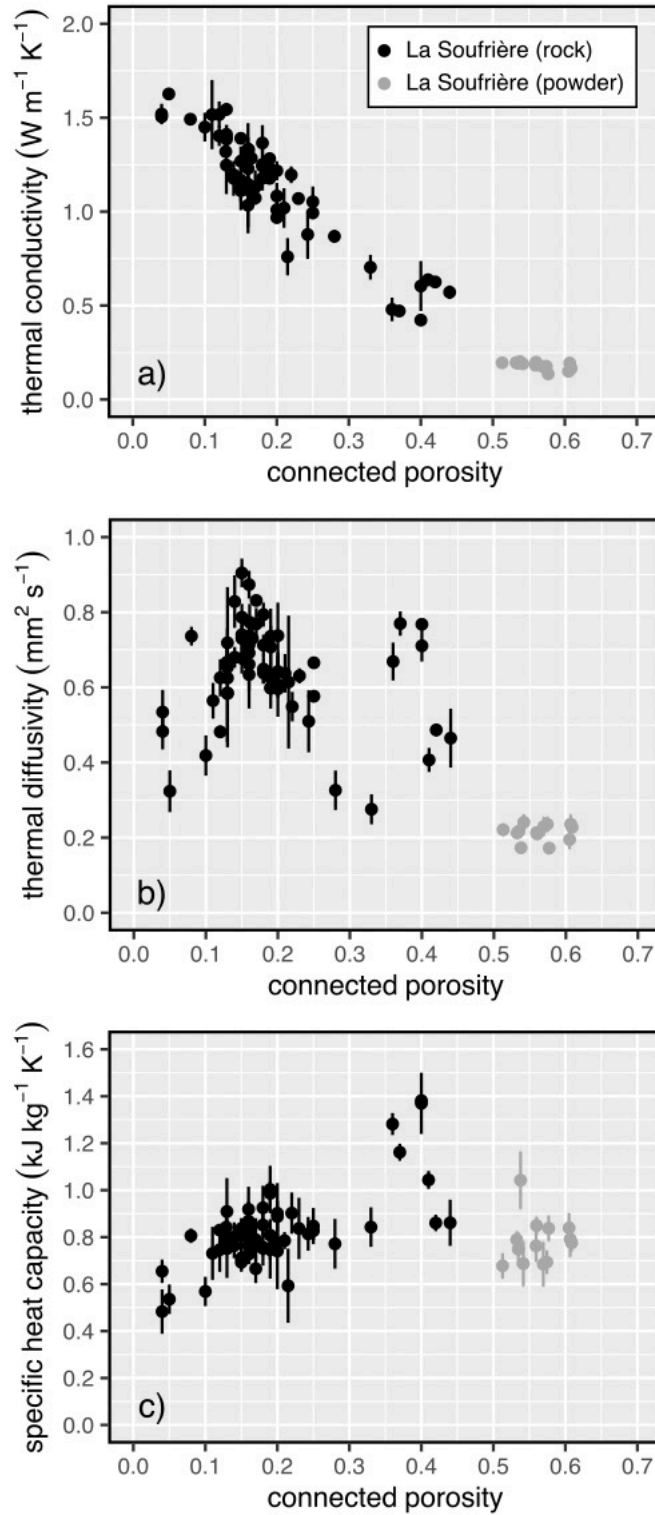
328

329 4.2 Thermal properties

330 Thermal conductivity, thermal diffusivity, and specific heat capacity for all the samples
331 (rocks and powders) are plotted as a function of porosity in Figure 5 (data available in Table
332 2). If we consider both types of sample (rocks and powders), we find that thermal conductivity
333 and thermal diffusivity decrease as a function of porosity, and that specific heat capacity does
334 not change systematically as a function of porosity (Figure 5). The trend with increasing
335 porosity is much more evident in the thermal conductivity data (Figure 5a) than in the thermal
336 diffusivity data (Figure 5b).

337 The porosity of the rock samples varies from 0.04 to 0.44, and the porosity of the
338 powders during the measurements, compacted using a 1 kg weight, varies between 0.51 and
339 0.61. For the rock samples, thermal conductivity decreases from $\sim 1.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a porosity
340 of ~ 0.04 to $\sim 0.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at a porosity of ~ 0.4 (Figure 5a). The thermal conductivity of the
341 powder samples is $\sim 0.2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for all samples (Figure 5a). The thermal diffusivity of the
342 rock samples generally decreases from ~ 0.9 to $\sim 0.3 \text{ mm}^2\cdot\text{s}^{-1}$ as porosity increases, and the
343 thermal diffusivity of the powder samples is $\sim 0.22 \text{ mm}^2\cdot\text{s}^{-1}$ for all samples (Figure 5b).
344 Excluding a few outliers, the specific heat capacity is $\sim 0.8 \text{ kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ for all samples (rocks
345 and powders), regardless of their porosity (Figure 5c).

346



347

348 **Figure 5.** Thermal conductivity, thermal diffusivity, and specific heat capacity as a function

349 of connected porosity for the rocks (black symbols) and unlithified materials (powders; grey

350 symbols) from La Soufrière de Guadeloupe (Eastern Caribbean).

351

| sample number | sample type | connected porosity | weight percentage of secondary minerals | thermal conductivity ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) | thermal diffusivity ($\text{mm}^2\cdot\text{s}^{-1}$) | specific heat capacity ($\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$) |
|---------------|-------------|--------------------|---|---|---|---|
| H2A | rock | 0.18 | 23.2 | 1.37 ± 0.10 | 0.79 ± 0.03 | 0.75 ± 0.03 |
| H2A | rock | 0.19 | 23.2 | 1.23 ± 0.08 | 0.74 ± 0.07 | 0.75 ± 0.13 |
| H2A | rock | 0.20 | 23.2 | 1.01 ± 0.04 | 0.60 ± 0.02 | 0.77 ± 0.04 |
| H2B | rock | 0.42 | 74.6 | 0.63 ± 0.02 | 0.49 ± 0.02 | 0.86 ± 0.04 |
| H2B | rock | 0.44 | 74.6 | 0.57 ± 0.04 | 0.46 ± 0.08 | 0.86 ± 0.10 |
| H2B | rock | 0.41 | 74.6 | 0.64 ± 0.03 | 0.41 ± 0.03 | 1.04 ± 0.04 |
| H3 | rock | 0.19 | 35.2 | 1.28 ± 0.03 | 0.62 ± 0.04 | 0.99 ± 0.05 |
| H3 | rock | 0.16 | 35.2 | 1.33 ± 0.06 | 0.69 ± 0.10 | 0.87 ± 0.15 |
| H3 | rock | 0.15 | 35.2 | 1.39 ± 0.03 | 0.73 ± 0.01 | 0.84 ± 0.02 |
| H4A | rock | 0.23 | 60.0 | 1.07 ± 0.03 | 0.63 ± 0.02 | 0.84 ± 0.13 |
| H4A | rock | 0.25 | 60.0 | 0.99 ± 0.03 | 0.67 ± 0.01 | 0.85 ± 0.08 |
| H4A | rock | 0.20 | 60.0 | 1.08 ± 0.07 | 0.64 ± 0.04 | 0.89 ± 0.07 |
| H5A | rock | 0.16 | 42.4 | 1.28 ± 0.06 | 0.66 ± 0.03 | 0.80 ± 0.03 |
| H5A | rock | 0.18 | 42.4 | 1.25 ± 0.02 | 0.64 ± 0.03 | 0.85 ± 0.05 |
| H5A | rock | 0.15 | 42.4 | 1.27 ± 0.08 | 0.68 ± 0.04 | 0.82 ± 0.05 |
| H6 | rock | 0.18 | 52.7 | 1.18 ± 0.02 | 0.65 ± 0.04 | 0.93 ± 0.09 |
| H6 | rock | 0.19 | 52.7 | 1.18 ± 0.02 | 0.60 ± 0.06 | 1.00 ± 0.10 |
| H6 | rock | 0.16 | 52.7 | 1.23 ± 0.02 | 0.71 ± 0.05 | 0.82 ± 0.08 |
| H14 | rock | 0.19 | 23.7 | 1.20 ± 0.03 | 0.71 ± 0.06 | 0.75 ± 0.04 |
| H14 | rock | 0.20 | 23.7 | 1.22 ± 0.05 | 0.61 ± 0.09 | 0.90 ± 0.13 |
| H14 | rock | 0.18 | 23.7 | 1.24 ± 0.13 | 0.71 ± 0.03 | 0.76 ± 0.08 |
| H15 | rock | 0.28 | 60.9 | 0.87 ± 0.03 | 0.33 ± 0.05 | 0.77 ± 0.11 |
| H15 | rock | 0.33 | 60.9 | 0.70 ± 0.07 | 0.28 ± 0.04 | 0.84 ± 0.08 |
| H18 | rock | 0.13 | 15.2 | 1.39 ± 0.08 | 0.59 ± 0.02 | 0.80 ± 0.05 |
| H18 | rock | 0.13 | 15.2 | 1.41 ± 0.05 | 0.65 ± 0.21 | 0.79 ± 0.05 |
| H18 | rock | 0.12 | 15.2 | 1.40 ± 0.05 | 0.48 ± 0.02 | 0.83 ± 0.02 |
| H19 | rock | 0.15 | 62.8 | 1.13 ± 0.04 | 0.74 ± 0.04 | 0.69 ± 0.03 |
| H19 | rock | 0.17 | 62.8 | 1.07 ± 0.04 | 0.77 ± 0.04 | 0.67 ± 0.06 |
| H19 | rock | 0.20 | 62.8 | 0.97 ± 0.03 | 0.74 ± 0.09 | 0.74 ± 0.16 |
| H20 | rock | 0.37 | 45.0 | 0.47 ± 0.03 | 0.77 ± 0.03 | 1.16 ± 0.04 |
| H20 | rock | 0.36 | 45.0 | 0.48 ± 0.06 | 0.67 ± 0.05 | 1.28 ± 0.05 |
| H20 | rock | 0.40 | 45.0 | 0.42 ± 0.02 | 0.71 ± 0.04 | 1.37 ± 0.13 |
| H21 | rock | 0.15 | 41.0 | 1.17 ± 0.12 | 0.90 ± 0.04 | 0.71 ± 0.06 |
| H21 | rock | 0.17 | 41.0 | 1.13 ± 0.09 | 0.83 ± 0.01 | 0.79 ± 0.09 |
| H21 | rock | 0.16 | 41.0 | 1.12 ± 0.20 | 0.87 ± 0.04 | 0.77 ± 0.04 |
| H22 | rock | 0.12 | 17.2 | 1.52 ± 0.07 | 0.63 ± 0.05 | 0.75 ± 0.09 |
| H22 | rock | 0.11 | 17.2 | 1.52 ± 0.18 | 0.56 ± 0.05 | 0.73 ± 0.11 |
| H22 | rock | 0.13 | 17.2 | 1.54 ± 0.03 | 0.62 ± 0.02 | 0.77 ± 0.03 |
| H25 | rock | 0.16 | 45.8 | 1.15 ± 0.07 | 0.63 ± 0.09 | 0.85 ± 0.10 |
| H25 | rock | 0.21 | 45.8 | 1.02 ± 0.11 | 0.64 ± 0.05 | 0.79 ± 0.04 |
| H25 | rock | 0.14 | 45.8 | 1.20 ± 0.03 | 0.68 ± 0.03 | 0.76 ± 0.05 |
| H29 | rock | 0.22 | 25.9 | 1.20 ± 0.04 | 0.55 ± 0.04 | 0.90 ± 0.09 |
| H29 | rock | 0.25 | 25.9 | 1.05 ± 0.08 | 0.58 ± 0.02 | 0.83 ± 0.04 |
| H29 | rock | 0.19 | 25.9 | 1.20 ± 0.04 | 0.60 ± 0.02 | 0.81 ± 0.06 |

| | | | | | | |
|--------|--------|------|------|-------------|-------------|-------------|
| H30 | rock | 0.16 | 85.4 | 1.30 ± 0.17 | 0.74 ± 0.01 | 0.92 ± 0.01 |
| H30 | rock | 0.40 | 85.4 | 0.60 ± 0.13 | 0.77 ± 0.01 | 1.38 ± 0.02 |
| H32 | rock | 0.04 | 6.0 | 1.52 ± 0.05 | 0.48 ± 0.05 | 0.65 ± 0.05 |
| H32 | rock | 0.04 | 6.0 | 1.51 ± 0.01 | 0.53 ± 0.06 | 0.48 ± 0.09 |
| H32 | rock | 0.05 | 6.0 | 1.63 ± 0.01 | 0.32 ± 0.06 | 0.54 ± 0.06 |
| WP1317 | rock | 0.16 | 16.3 | 1.03 ± 0.15 | 0.73 ± 0.03 | 0.85 ± 0.05 |
| WP1317 | rock | 0.15 | 16.3 | 1.11 ± 0.10 | 0.79 ± 0.04 | 0.81 ± 0.07 |
| WP1317 | rock | 0.14 | 16.3 | 1.18 ± 0.09 | 0.83 ± 0.07 | 0.80 ± 0.06 |
| WP1285 | rock | 0.13 | 13.4 | 1.40 ± 0.02 | 0.72 ± 0.04 | 0.75 ± 0.12 |
| WP1285 | rock | 0.10 | 13.4 | 1.45 ± 0.08 | 0.42 ± 0.05 | 0.57 ± 0.06 |
| WP1285 | rock | 0.08 | 13.4 | 1.49 ± 0.01 | 0.74 ± 0.02 | 0.81 ± 0.03 |
| SAM58 | rock | 0.14 | - | 1.18 ± 0.02 | 0.66 ± 0.03 | 0.75 ± 0.05 |
| SAM58 | rock | 0.14 | - | 1.21 ± 0.05 | 0.65 ± 0.03 | 0.78 ± 0.04 |
| S005 | rock | 0.22 | - | 0.76 ± 0.10 | 0.61 ± 0.18 | 0.59 ± 0.16 |
| S005 | rock | 0.24 | - | 0.88 ± 0.13 | 0.51 ± 0.08 | 0.81 ± 0.07 |
| S008 | rock | 0.17 | - | 1.28 ± 0.04 | 0.73 ± 0.03 | 0.75 ± 0.02 |
| S008 | rock | 0.16 | - | 1.33 ± 0.02 | 0.77 ± 0.05 | 0.73 ± 0.04 |
| S016 | rock | 0.13 | - | 1.32 ± 0.03 | 0.67 ± 0.05 | 0.84 ± 0.06 |
| S016 | rock | 0.13 | - | 1.25 ± 0.15 | 0.58 ± 0.13 | 0.91 ± 0.14 |
| S001 | powder | 0.61 | - | 0.19 ± 0.00 | 0.24 ± 0.02 | 0.79 ± 0.08 |
| S002 | powder | 0.56 | - | 0.20 ± 0.01 | 0.21 ± 0.01 | 0.85 ± 0.04 |
| S003 | powder | 0.53 | - | 0.20 ± 0.01 | 0.21 ± 0.00 | 0.79 ± 0.03 |
| S004 | powder | 0.61 | - | 0.15 ± 0.01 | 0.19 ± 0.03 | 0.84 ± 0.06 |
| S006 | powder | 0.57 | - | 0.18 ± 0.01 | 0.23 ± 0.03 | 0.68 ± 0.09 |
| S007 | powder | 0.56 | - | 0.18 ± 0.01 | 0.21 ± 0.02 | 0.76 ± 0.07 |
| S009 | powder | 0.51 | - | 0.20 ± 0.01 | 0.22 ± 0.02 | 0.68 ± 0.05 |
| S010 | powder | 0.54 | - | 0.19 ± 0.01 | 0.22 ± 0.01 | 0.75 ± 0.07 |
| S011 | powder | 0.57 | - | 0.18 ± 0.00 | 0.24 ± 0.02 | 0.69 ± 0.05 |
| S012 | powder | 0.61 | - | 0.17 ± 0.01 | 0.23 ± 0.01 | 0.78 ± 0.04 |
| S013 | powder | 0.54 | - | 0.19 ± 0.01 | 0.24 ± 0.02 | 0.69 ± 0.10 |
| S014 | powder | 0.58 | - | 0.14 ± 0.00 | 0.17 ± 0.01 | 0.84 ± 0.05 |
| S015 | powder | 0.54 | - | 0.20 ± 0.02 | 0.17 ± 0.01 | 1.04 ± 0.12 |

352

353 **Table 2.** Connected porosity, weight percentage of secondary minerals, thermal conductivity,
354 thermal diffusivity, and specific heat capacity for the samples from La Soufrière de
355 Guadeloupe (Eastern Caribbean) measured for this study. The standard deviations provided
356 relate to measurement precision (calculated using the four measurements). The standard
357 uncertainty for values of thermal conductivity and thermal diffusivity using the transient hot-
358 strip method has been determined to be 2.6 and 11%, respectively (Hammerschmidt and
359 Sabuga, 2000).

| volcano | rock type | connected porosity | weight percentage of secondary minerals | thermal conductivity ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) | thermal diffusivity ($\text{mm}^2\cdot\text{s}^{-1}$) | specific heat capacity ($\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$) |
|-------------|-------------------|--------------------|---|---|---|---|
| Chaos Crags | rhyodacite | 0.153 | 6.4 | 1.14 ± 0.03 | 0.65 ± 0.02 | 0.78 ± 0.02 |
| Chaos Crags | rhyodacite | 0.139 | 14.6 | 1.29 ± 0.03 | 0.73 ± 0.07 | 0.78 ± 0.08 |
| Chaos Crags | rhyodacite | 0.125 | 14.6 | 1.52 ± 0.26 | 0.84 ± 0.09 | 0.77 ± 0.07 |
| Chaos Crags | rhyodacite | 0.138 | 14.6 | 1.40 ± 0.24 | 0.94 ± 0.06 | 0.66 ± 0.15 |
| Chaos Crags | rhyodacite | 0.076 | 16.5 | 1.66 ± 0.09 | 0.89 ± 0.07 | 0.77 ± 0.02 |
| Chaos Crags | rhyodacite | 0.142 | 16.5 | 1.51 ± 0.28 | 0.90 ± 0.13 | 0.72 ± 0.05 |
| Chaos Crags | rhyodacite | 0.115 | 16.5 | 1.44 ± 0.17 | 0.78 ± 0.11 | 0.78 ± 0.03 |
| Merapi | basaltic-andesite | 0.080 | 7.5 | 1.43 ± 0.02 | 0.70 ± 0.04 | 0.79 ± 0.03 |
| Merapi | basaltic-andesite | 0.084 | 7.5 | 1.37 ± 0.03 | 0.73 ± 0.03 | 0.74 ± 0.02 |
| Merapi | basaltic-andesite | 0.077 | 7.5 | 1.48 ± 0.02 | 0.73 ± 0.04 | 0.79 ± 0.04 |
| Merapi | basaltic-andesite | 0.079 | 29.0 | 1.20 ± 0.02 | 0.57 ± 0.02 | 0.86 ± 0.04 |
| Merapi | basaltic-andesite | 0.080 | 29.0 | 1.23 ± 0.05 | 0.57 ± 0.03 | 0.87 ± 0.02 |
| Merapi | basaltic-andesite | 0.083 | 29.0 | 1.28 ± 0.01 | 0.53 ± 0.02 | 0.98 ± 0.03 |
| Merapi | basaltic-andesite | 0.154 | 45.0 | 1.06 ± 0.07 | 0.51 ± 0.03 | 0.90 ± 0.04 |
| Merapi | basaltic-andesite | 0.182 | 45.0 | 0.90 ± 0.06 | 0.51 ± 0.03 | 0.81 ± 0.01 |
| Merapi | basaltic-andesite | 0.144 | 45.0 | 1.07 ± 0.05 | 0.53 ± 0.01 | 0.91 ± 0.06 |
| Merapi | basaltic-andesite | 0.155 | 45.0 | 1.04 ± 0.01 | 0.52 ± 0.02 | 0.88 ± 0.04 |
| Merapi | basaltic-andesite | 0.160 | 45.0 | 0.97 ± 0.08 | 0.54 ± 0.03 | 0.81 ± 0.06 |
| Merapi | basaltic-andesite | 0.162 | 45.0 | 0.97 ± 0.01 | 0.66 ± 0.21 | 0.73 ± 0.23 |
| Merapi | basaltic-andesite | 0.182 | 45.0 | 0.94 ± 0.00 | 0.43 ± 0.00 | 0.99 ± 0.00 |
| Merapi | basaltic-andesite | 0.215 | 62.0 | 0.78 ± 0.08 | 0.60 ± 0.09 | 0.66 ± 0.18 |
| Merapi | basaltic-andesite | 0.233 | 62.0 | 0.80 ± 0.04 | 0.51 ± 0.09 | 0.79 ± 0.10 |
| Merapi | basaltic-andesite | 0.220 | 62.0 | 0.86 ± 0.07 | 0.51 ± 0.04 | 0.82 ± 0.02 |
| Merapi | basaltic-andesite | 0.188 | 62.0 | 0.86 ± 0.06 | 0.50 ± 0.03 | 0.82 ± 0.02 |
| Merapi | basaltic-andesite | 0.163 | 62.0 | 0.88 ± 0.01 | 0.55 ± 0.04 | 0.75 ± 0.06 |
| Merapi | basaltic-andesite | 0.242 | 62.0 | 0.79 ± 0.05 | 0.46 ± 0.00 | 0.86 ± 0.05 |
| Merapi | basaltic-andesite | 0.263 | 62.0 | 0.79 ± 0.01 | 0.47 ± 0.04 | 0.88 ± 0.07 |
| Merapi | basaltic-andesite | 0.168 | 62.0 | 0.85 ± 0.03 | 0.44 ± 0.09 | 0.92 ± 0.19 |
| Merapi | basaltic-andesite | 0.231 | 32.5 | 0.75 ± 0.06 | 0.45 ± 0.06 | 0.80 ± 0.11 |
| Merapi | basaltic-andesite | 0.236 | 32.5 | 0.76 ± 0.05 | 0.51 ± 0.02 | 0.70 ± 0.07 |
| Merapi | basaltic-andesite | 0.262 | 32.5 | 0.76 ± 0.04 | 0.55 ± 0.11 | 0.70 ± 0.11 |
| Merapi | basaltic-andesite | 0.256 | 32.5 | 0.75 ± 0.05 | 0.47 ± 0.05 | 0.78 ± 0.06 |
| Ruapehu | andesite | 0.021 | - | 1.54 ± 0.02 | 0.70 ± 0.02 | 0.80 ± 0.03 |
| Ruapehu | andesite | 0.040 | - | 1.62 ± 0.02 | 0.77 ± 0.02 | 0.78 ± 0.01 |
| Ruapehu | andesite | 0.024 | - | 1.47 ± 0.06 | 0.77 ± 0.07 | 0.72 ± 0.10 |
| Ruapehu | andesite | 0.036 | - | 1.46 ± 0.05 | 0.75 ± 0.01 | 0.73 ± 0.02 |
| Ruapehu | andesite | 0.042 | - | 1.53 ± 0.01 | 0.76 ± 0.03 | 0.74 ± 0.04 |
| Ruapehu | andesite | 0.047 | - | 1.51 ± 0.05 | 0.72 ± 0.05 | 0.79 ± 0.09 |
| Ruapehu | andesite | 0.038 | - | 1.45 ± 0.03 | 0.70 ± 0.03 | 0.77 ± 0.05 |
| Ruapehu | andesite | 0.036 | - | 1.50 ± 0.00 | 0.72 ± 0.01 | 0.78 ± 0.01 |
| Ruapehu | andesite | 0.024 | - | 1.48 ± 0.02 | 0.71 ± 0.02 | 0.77 ± 0.03 |
| Ruapehu | andesite | 0.027 | - | 1.46 ± 0.03 | 0.68 ± 0.01 | 0.79 ± 0.00 |
| Ruapehu | andesite | 0.048 | - | 1.39 ± 0.01 | 0.83 ± 0.06 | 0.64 ± 0.04 |
| Ruapehu | andesite | 0.042 | - | 1.41 ± 0.00 | 0.66 ± 0.00 | 0.80 ± 0.00 |
| Ruapehu | andesite | 0.184 | - | 1.06 ± 0.01 | 0.65 ± 0.04 | 0.73 ± 0.05 |
| Ruapehu | andesite | 0.205 | - | 1.00 ± 0.05 | 0.58 ± 0.05 | 0.79 ± 0.11 |
| Ruapehu | andesite | 0.098 | - | 1.26 ± 0.01 | 0.70 ± 0.04 | 0.72 ± 0.04 |
| Ruapehu | andesite | 0.118 | - | 1.22 ± 0.06 | 0.65 ± 0.05 | 0.77 ± 0.10 |

| | | | | | | |
|---------|----------|-------|---|-----------------|-----------------|-----------------|
| Ruapehu | andesite | 0.153 | - | 1.16 ± 0.05 | 0.66 ± 0.08 | 0.75 ± 0.06 |
| Ruapehu | andesite | 0.140 | - | 1.23 ± 0.05 | 0.71 ± 0.06 | 0.73 ± 0.08 |
| Ruapehu | andesite | 0.149 | - | 1.14 ± 0.04 | 0.65 ± 0.02 | 0.73 ± 0.05 |
| Ruapehu | andesite | 0.167 | - | 1.08 ± 0.09 | 0.72 ± 0.09 | 0.65 ± 0.03 |
| Ruapehu | andesite | 0.129 | - | 1.21 ± 0.04 | 0.59 ± 0.01 | 0.86 ± 0.04 |
| Ruapehu | andesite | 0.151 | - | 1.13 ± 0.05 | 0.60 ± 0.06 | 0.80 ± 0.05 |
| Ruapehu | andesite | 0.204 | - | 1.01 ± 0.05 | 0.61 ± 0.03 | 0.75 ± 0.00 |
| Ruapehu | andesite | 0.182 | - | 1.09 ± 0.02 | 0.62 ± 0.05 | 0.78 ± 0.05 |
| Ruapehu | andesite | 0.308 | - | 0.81 ± 0.00 | 0.64 ± 0.03 | 0.66 ± 0.03 |
| Ruapehu | andesite | 0.320 | - | 0.84 ± 0.00 | 0.75 ± 0.11 | 0.61 ± 0.10 |
| Ruapehu | andesite | 0.345 | - | 0.81 ± 0.04 | 0.52 ± 0.05 | 0.85 ± 0.05 |
| Ruapehu | andesite | 0.348 | - | 0.81 ± 0.06 | 0.59 ± 0.02 | 0.76 ± 0.05 |
| Ruapehu | andesite | 0.333 | - | 0.79 ± 0.05 | 0.53 ± 0.06 | 0.81 ± 0.07 |
| Ruapehu | andesite | 0.382 | - | 0.72 ± 0.04 | 0.63 ± 0.09 | 0.68 ± 0.14 |
| Ruapehu | andesite | 0.602 | - | 0.43 ± 0.03 | 0.51 ± 0.04 | 0.79 ± 0.02 |
| Ruapehu | andesite | 0.628 | - | 0.38 ± 0.03 | 0.55 ± 0.08 | 0.71 ± 0.16 |

361

362 **Table 3.** Connected porosity, weight percentage of secondary minerals, thermal conductivity,
363 thermal diffusivity, and specific heat capacity for the samples from Chaos Crags (USA; data
364 unique to this study), Merapi volcano (Indonesia; data from Heap et al., 2020a), and Ruapehu
365 (New Zealand; data from Heap et al, 2020a). All of the samples listed are rock samples.

366 Alteration (percentage of secondary minerals) of the samples from Merapi volcano was not
367 reported in Heap et al. (2020a). XRPD data are not available for the samples from Ruapehu
368 and so alteration values are not reported. The standard deviations provided relate to
369 measurement precision (calculated using the four measurements). The standard uncertainty
370 for values of thermal conductivity and thermal diffusivity using the transient hot-strip method
371 has been determined to be 2.6 and 11%, respectively (Hammerschmidt and Sabuga, 2000).

372

373 **5 Discussion**

374 5.1 The influence of porosity on thermal properties

375 To better understand the influence of porosity on the thermal properties of volcanic
376 rocks, we compare our new data for rocks and powders La Soufrière (Table 2) with published
377 data from Heap et al. (2020a) for rocks from Ruapehu and Merapi volcano (Table 3) and new
378 data for variably-altered rhyodacites from Chaos Crags (Lassen Volcanic National Park,

379 California, USA) (Table 3). The blocks from Ruapehu are variably porous (from <0.05 to ~0.6)
380 and relatively unaltered porphyritic andesites that contain large phenocrysts of plagioclase and
381 pyroxene within a glassy microlite-rich groundmass (Heap and Kennedy, 2016; Heap et al.,
382 2020a). The blocks from Merapi volcano are variably altered basaltic-andesites with porphyritic
383 texture comprising phenocrysts of dominantly plagioclase and pyroxene within a crystallised
384 groundmass (Heap et al., 2019, 2020a). The alteration phases present are similar to those found
385 in the blocks from La Soufrière, and include natro-alunite, alunite, quartz, hematite, cristobalite,
386 gypsum, and unclassified amorphous phases (Heap et al., 2019, 2020a). The alteration of the
387 blocks from Merapi volcano—the percentage of secondary minerals in the block—ranges from
388 7.5 wt.% up to 62 wt.% (Table 3). The blocks from Chaos Crags, described in Ryan et al. (2020)
389 and Heap et al. (2021b), are porphyritic rhyodacites containing phenocrysts of dominantly
390 plagioclase, K-feldspar, and quartz within a crystallised groundmass. Secondary minerals
391 include cristobalite, hematite, smectite, and kaolinite (Heap et al., 2021b). The alteration of the
392 rhyodacite blocks from Chaos Crags ranges from 6.4 wt.% up to 16.5 wt.% (Table 3). The
393 thermal properties of cylindrical samples prepared from these blocks (Ruapehu, Merapi
394 volcano, and Chaos Crags) were measured using the same Hot Disk® Thermal Constants
395 Analyser used to measure the samples from La Soufrière, as described above, adding
396 confidence to direct data comparisons.

397 The effective thermal conductivity, $\lambda(\phi)$, can be determined using the Maxwell result
398 for conduction in a porous medium,

399

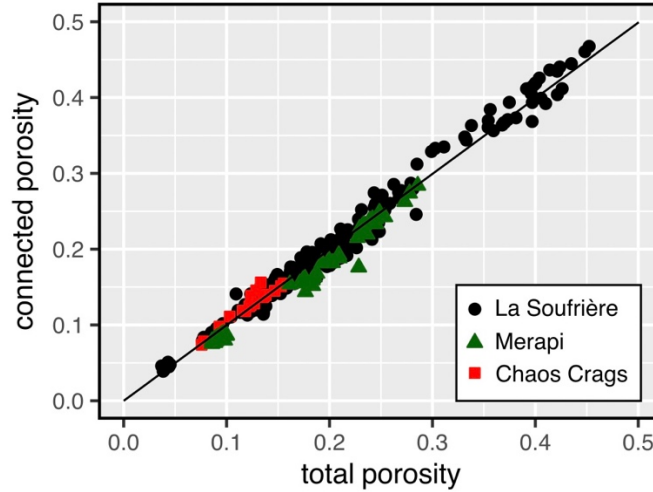
$$400 \quad \frac{\lambda(\phi)}{\lambda_0} = \frac{(1 - \phi)(1 - r) + r\beta\phi}{(1 - \phi)(1 - r) + \beta\phi}, (1)$$

401

402 where ϕ is the total porosity, $r = \lambda_f/\lambda_0$ (where λ_f and λ_0 are the thermal conductivities of
403 the pore-filling fluid and the rock groundmass, respectively; Zimmerman, 1989). We assume

404 spherical pores, and so $\beta = 3(1 - r)/(2 + r)$ (Zimmerman, 1989). The Maxwell model is a
405 dilute approximation and so assumes no interaction between the spherical pores. Because our
406 thermal conductivity data are for dry materials, we assume a pore fluid thermal conductivity,
407 λ_f , of $0 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (i.e. we assume that the conduction of the porosity-filling air is negligible).
408 The Maxwell model for the effect of pores (or any inclusion) on the thermal properties of a
409 medium (Equation (1)) is a dilute approximation for spherical pores or inclusions. By contrast,
410 powders are the effective geometric inverse of spherical pores of gas in a continuum matrix,
411 and are instead discrete (sub-angular or sub-spherical) particles in a continuum gas. Therefore,
412 not only does the Maxwell model (Equation (1)) not apply, but it also is reasonable to expect
413 that the functional form of the appropriate model for powders is different. This is explicitly
414 documented in Torquato and Haslach (2002), who gives a range of effective medium models
415 for the physical properties of inclusions of one phase in another phase, as well as the inverse
416 case. As an applied example, Vasseur et al. (2016) found that the elastic properties of coherent
417 versus powdered materials were best scaled by two different effective medium models, and that
418 the transition from one to the other (from coherent to powdered) could be captured by a cross
419 over between models. As a result, in the following we will only discuss the measurements
420 performed on rock samples. Finally, we highlight that the Maxwell equation (Equation (1)), and
421 the effective medium models below (Equations (2) and (3)), consider the total porosity, whereas
422 we have measured the connected porosity of our samples. To assess the suitability of using
423 these models to discuss our experimental data (Figure 5; Tables 2 and 3), we provide a plot of
424 connected porosity as a function of total porosity for numerous 20 mm-diameter cylindrical
425 samples from La Soufrière, Chaos Crags, and Merapi volcano (Figure 6), which shows that
426 there is little to no isolated porosity in the studied materials.

427



428

429 **Figure 6.** Connected porosity as a function of total porosity for 20 mm-diameter cylindrical
 430 samples from La Soufrière, Chaos Crags, and Merapi volcano. The black line shows the 1:1
 431 line, whereat the connected porosity equals the total porosity.

432

433 We find that our data for La Soufrière, as well as data for rhyodacites from Chaos Crags
 434 and basaltic-andesites from Merapi volcano, are bracketed by curves for which λ_0 is 1.9 and
 435 $0.85 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (the black lines in Figure 7a). A third curve, which best captures the overall
 436 trend of the dataset, is also provided in Figure 7a (for which λ_0 is $1.45 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$). The
 437 effective thermal diffusivity $D(\phi)$ was then determined using,

438

$$439 \quad D(\phi) = \frac{\lambda(\phi)}{\rho_s C_p (1 - \phi) + \rho_f C_{p,f} \phi}, \quad (2)$$

440

441 where ρ_s and ρ_f are the groundmass and pore fluid densities, respectively, and C_p and $C_{p,f}$ are
 442 the groundmass and pore fluid specific heat capacity, respectively (Connor et al., 1997). We
 443 take values of ρ_f and $C_{p,f}$ of $1.275 \text{ kg}\cdot\text{m}^{-3}$ and $1.007 \text{ kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$, respectively. Values for ρ_s
 444 and C_p , guided by our data, were taken as $2650 \text{ kg}\cdot\text{m}^{-3}$ and $0.8 \text{ kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$, respectively.

445 Theoretical curves are provided using the $\lambda(\phi)$ values taken from the three models shown in

446 Figure 7a (i.e. $\lambda_0 = 1.9, 1.45, \text{ and } 0.85 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$; Equation (1); the black lines in Figure 7b).
447 Based on Equation (2), the effective specific heat capacity $C_p(\phi)$ is given by the porosity-
448 weighted average,

449

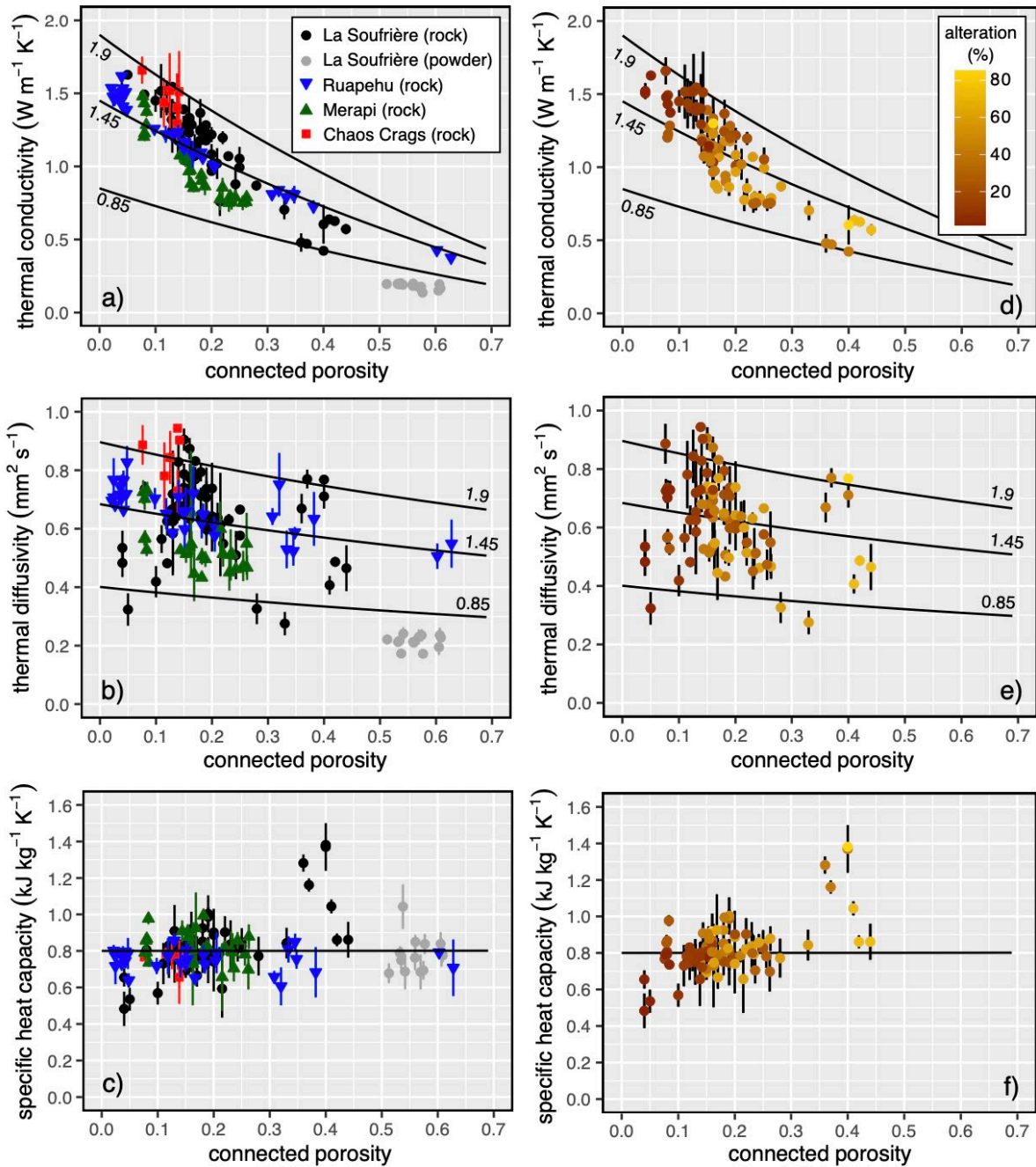
$$450 \quad C_p(\phi) = \frac{\rho_s C_p (1 - \phi) + \rho_f C_{p,f} \phi}{\rho_b}, (3)$$

451

452 where ρ_b is the bulk sample density, where $\rho_b = \rho_s(1 - \phi) + \rho_f \phi$. To plot $C_p(\phi)$, we take the
453 same values outlined above for the different variables (the black line in Figure 7c).

454 The effective medium models (Equations (1), (2), and (3)) are in reasonable agreement
455 with the data (Figure 7), suggesting that porosity plays an important role in governing their
456 thermal properties (as discussed in Heap et al., 2020a). The scatter in these data, i.e. the reason
457 why even data from the same volcano cannot be described by a single theoretical curve (Figure
458 7), is the result of factors not considered in Equations (1), (2), and (3), such as differences in
459 the nature of the void space (e.g., pore size, shape, and number density, microcrack density;
460 factors that can vary significantly in volcanic rocks, see the review of Heap and Violay, 2021)
461 and differences in their mineral componentry (e.g., variable alteration intensities and secondary
462 mineral assemblages). The large differences in the alteration of these rocks (Tables 2 and 3),
463 and that the effective medium approaches well describe the relatively unaltered suite of rocks
464 from Ruapehu (Figure 7; Heap et al., 2020a) and Hawai'i (Robertson and Peck, 1974) with
465 variable pore sizes and shapes, suggest that the scatter in the data can be explained by their
466 variable alteration, discussed in the next section.

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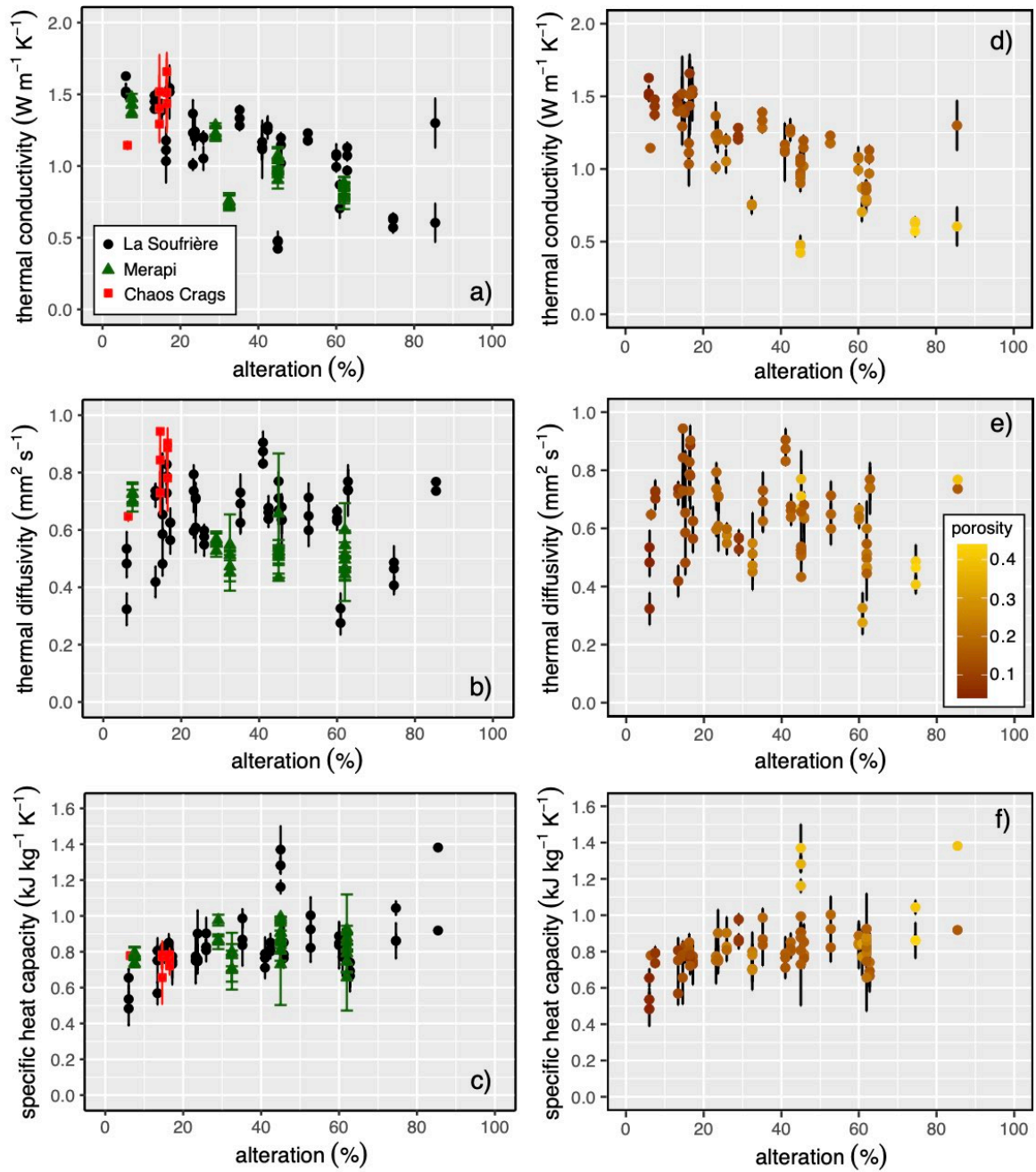
474

Figure 7. Thermal conductivity (a and d), thermal diffusivity (b and e), and specific heat capacity (c and f) as a function of connected porosity for the samples from La Soufrière de Guadeloupe (Eastern Caribbean), Merapi volcano (Indonesia; data from Heap et al., 2020a), Ruapehu (New Zealand; data from Heap et al., 2020a), and Chaos Crags (USA). In panels (a–c), the symbols and colours differentiate the data from the different volcanoes (La Soufrière de Guadeloupe: black (rocks) and grey circles (powders); Merapi volcano: green triangles;

475 Ruapehu: blue triangles; Chaos Crags: red squares). In panels (d–f), the colour of the symbol
476 (where red and yellow indicate low and high values, respectively) indicates the alteration
477 (percentage of secondary minerals). Numbers next to the modelled curves in panels (a), (b),
478 (d), and (e) indicate the assumed rock groundmass conductivity, λ_0 (in $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$). The
479 modelled curves for λ_0 values of 0.85 and 1.9 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ are designed to bracket the dataset.
480

481 5.2 The influence of alteration on thermal properties

482 To explore the influence of hydrothermal alteration on the thermal properties of volcanic
483 rocks, we plot thermal conductivity, thermal diffusivity, and specific heat capacity as a function
484 of alteration (taken to be the weight percentage of secondary minerals) in Figure 8. We
485 supplement these data with published data for variably-altered basaltic-andesites from Merapi
486 volcano (from Heap et al., 2020a; Table 3) and new data for variably-altered rhyodacites from
487 Chaos Crags (Table 3). The methods used, XRPD to quantify their mineral contents and the
488 Hot Disk® Thermal Constants Analyser to measure their thermal properties, were the same as
489 for the measured materials from La Soufrière (presented in Table 2), adding confidence to direct
490 data comparisons. Similar to the samples from La Soufrière, the secondary mineral assemblages
491 of the rocks from Merapi volcano and Chaos Crags correspond to intermediate to advanced
492 argillic alteration.



493

494 **Figure 8.** Thermal conductivity (a and d), thermal diffusivity (b and e), and specific heat
 495 capacity (c and f) as a function of alteration (percentage of secondary minerals) for the
 496 samples from La Soufrière de Guadeloupe (Eastern Caribbean), Merapi volcano (Indonesia;
 497 data from Heap et al., 2020a), and Chaos Crags (USA). In panels (a–c), the symbols and
 498 colours differentiate the data from the different volcanoes (La Soufrière de Guadeloupe: black
 499 (rocks) and grey circles (powders); Merapi volcano: green triangles; Ruapehu: blue triangles;

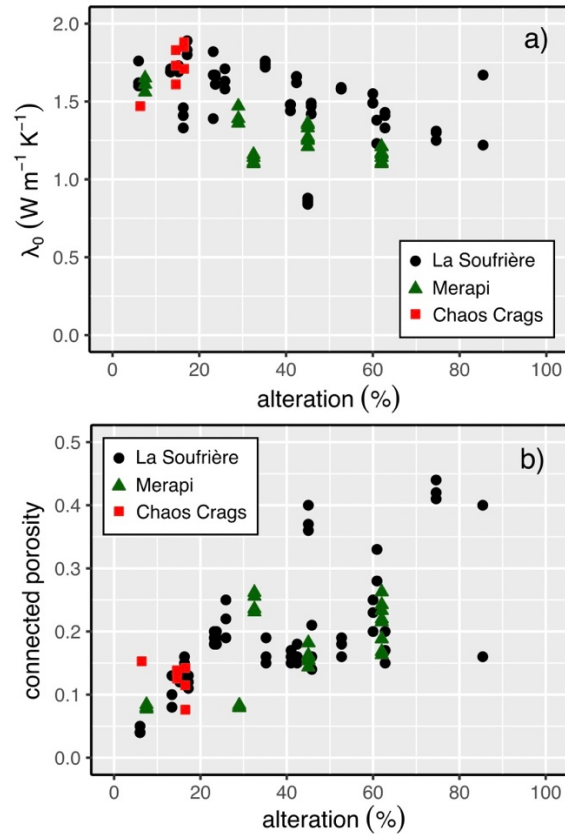
500 Chaos Crags: red squares). In panels (d–f), the colour of the symbol (where red and yellow
501 indicate low and high values, respectively) indicates the connected porosity.

502

503 The thermal conductivity of the rocks from La Soufrière and Merapi volcano decreases
504 as a function of increasing alteration, from $\sim 1.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at an alteration of $\sim 1.5 \text{ wt.}\%$ to
505 $\sim 0.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at an alteration $> 75 \text{ wt.}\%$ (Figure 8a). The thermal conductivity of the rocks
506 from Chaos Crags, on the other hand, increases as alteration increases, from $\sim 1.1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
507 at an alteration of $\sim 6 \text{ wt.}\%$ to $\sim 1.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at an alteration of $\sim 15 \text{ wt.}\%$ (Figure 8a).
508 Although the thermal diffusivity of the rocks from La Soufrière and Merapi volcano does not
509 appear to be influenced by alteration, the thermal diffusivity of the rocks from Chaos Crags
510 increases from ~ 0.65 to $\sim 0.75\text{--}0.95 \text{ mm}^2\cdot\text{s}^{-1}$ as alteration increases from ~ 6 to $\sim 15 \text{ wt.}\%$ (Figure
511 8b). The specific heat capacity of all rocks does not appear to be significantly affected by
512 alteration, although there is a slight trend of increasing specific heat capacity with increasing
513 alteration for the rocks from La Soufrière (Figure 8c).

514 The reasons for the observed changes in thermal properties as a function of
515 hydrothermal alteration (Figure 8) are twofold. First, the thermal properties of the primary and
516 secondary minerals differ (Horai, 1971; Brigaud and Vasseur, 1989; Clauser and Huenges,
517 1995). Therefore, increasing the proportion of secondary minerals at the expense of the primary
518 minerals will either increase or decrease the thermal properties of a sample, depending on
519 whether the secondary minerals have higher values of thermal conductivity, diffusivity, and
520 specific heat capacity than the primary minerals. Second, hydrothermal alteration can promote
521 either increases in porosity (dissolution) or decreases in porosity (pore- and crack-filling
522 mineral precipitation), a physical property known to influence thermal conductivity (as
523 discussed in the previous section; Figure 5; Robertson and Peck, 1974; Heap et al., 2020a).

524



525

526 **Figure 9.** The thermal conductivity of the rock groundmass, λ_0 , calculated using Equation (1)
 527 (a) and the connected porosity (b) as a function of alteration (percentage of secondary
 528 minerals) for the samples from La Soufrière de Guadeloupe (Eastern Caribbean; black
 529 circles), Merapi volcano (Indonesia; green triangles; data from Heap et al., 2020a), and Chaos
 530 Crags (USA; red squares).

531

532 To explore whether the different thermal properties of secondary minerals, compared to
 533 primary minerals, is affecting the thermal properties of the studied rocks, we exploit the fact
 534 that Equation (1) shows that λ is only a function of λ_0 (the thermal conductivity of the
 535 groundmass) and the porosity, such that we can solve for λ_0 for each sample. The value of λ_0
 536 for each sample represents the thermal conductivity of the sample at zero porosity, allowing us
 537 to assess solely the influence of alteration on thermal conductivity. λ_0 is plotted as a function
 538 of alteration in Figure 9a. Figure 9a shows that λ_0 broadly decreases as a function of alteration

539 for the samples from La Soufrière and Merapi volcano, but appears to increase for the samples
540 from Chaos Crags. The decrease in thermal conductivity as a function of alteration for the
541 samples from La Soufrière and Merapi volcano can be, at least partly, explained by the lower
542 thermal conductivity of the secondary minerals compared to the primary minerals (Horai, 1971;
543 Brigaud and Vasseur, 1989; Clauser and Huenges, 1995). For example, the thermal conductivity
544 of kaolinite, a common replacement mineral in the rocks from La Soufrière (Table 1; Figures 3
545 and 4), is lower than that of plagioclase (Horai, 1971; Brigaud and Vasseur, 1989). For the rocks
546 from Chaos Crags, we note that, although smectite and kaolinite have low thermal
547 conductivities (Brigaud and Vasseur, 1989), the high thermal conductivity of cristobalite (~ 6
548 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$; Kunugi et al., 1972) could explain why their thermal conductivities appear to
549 increase as a function of increasing alteration (Figure 8a).

550 The above analysis suggests that hydrothermal alteration can modify the thermal
551 conductivity of volcanic rock by changing the mineral assemblage. However, alteration can
552 also result in either increases in porosity (dissolution) or decreases in porosity (pore- and crack-
553 filling mineral precipitation), a parameter that we have already established as important in
554 dictating the thermal properties of volcanic rock (Figure 7). To explore whether the observed
555 alteration as resulted in changes to porosity, and therefore thermal properties, we plot alteration
556 as a function of porosity in Figure 9b. We also provide plots in Figure 7 that show thermal
557 conductivity, thermal diffusivity, and specific heat capacity as a function of porosity in which
558 the symbol colour indicates the alteration (Figures 7d, 7e, and 7f). Similarly, in Figures 8d, 8e,
559 and 8f, which show plots of thermal conductivity, thermal diffusivity, and specific heat capacity
560 as a function of alteration, the symbol colour indicates the porosity. Figure 9b shows that
561 porosity increases as a function of alteration for the rocks from La Soufrière and Merapi
562 volcano, but appears to decrease as a function of alteration for the rocks from Chaos Crags.
563 Figures 7d–f and 8d–f also show that the more altered samples are typically the most porous.

564 Therefore, we could conclude from these data that the changes in thermal properties as a
565 function of alteration (Figure 8a) are also the result of alteration-induced increases (in the case
566 of the rocks from La Soufrière and Merapi volcano) and decreases (in the case of the rocks from
567 Chaos Crags) to the porosity, alongside the changes resulting from modifications to the mineral
568 assemblage (Figure 9a). However, because alteration efficiency is increased by higher fluid-
569 rock ratios (Giggenbach, 1984), another possibility is that the high-porosity rocks collected
570 from La Soufrière and Merapi volcano are simply more altered because they were initially more
571 porous and permeable. We also note that detailed microstructural analysis on the rocks from La
572 Soufrière (Figures 3 and 4; Heap et al., 2021a) and Merapi volcano (Heap et al., 2019, 2021c)
573 shows that not all alteration was associated with mineral dissolution, and that samples from
574 both sample suites show evidence of microcrack- and pore-filling alteration (Figures 3 and 4;
575 Heap et al., 2019, 2021a). Therefore, although we observe changes in the thermal properties of
576 the rocks collected from La Soufrière and Merapi volcano as a function of alteration (Figure 8),
577 it is difficult at present to conclude whether changes in porosity resulting directly from
578 alteration influenced their thermal properties. It appears more likely, due to the abundance of
579 void-filling mineral precipitation (Figures 3 and 4; Heap et al., 2021a), that the trend of
580 increasing porosity with alteration (Figure 9b) is simply the result of increasing alteration
581 efficiency at higher porosity due to an increase in fluid-rock ratio (i.e. higher interfacial surface
582 area). Therefore, in the case of the andesites and basaltic-andesites from La Soufrière and
583 Merapi volcano, it is likely that the decrease in thermal conductivity as a function of alteration
584 (Figure 8a) is the result of the low conductivity of the secondary mineral assemblage (Horai,
585 1971; Brigaud and Vasseur, 1989; Clauser and Huenges, 1995).

586 The explanation that higher porosities, and therefore higher fluid-rock ratios, led to more
587 efficient alteration, however, cannot explain the decrease in porosity as alteration is increased
588 from ~6 wt.% to ~15 wt.% in the rocks from Chaos Crags (Figure 9b). Detailed microstructural

589 work on the rocks from Chaos Crags showed that the alteration was manifest as microcrack-
590 and pore-filling mineral precipitation of mainly cristobalite and kaolinite (Heap et al., 2021b).
591 Therefore, in the case of the rhyodacites from Chaos Crags, it is likely that the increase in
592 thermal conductivity (Figure 8a) and thermal diffusivity (Figure 8b) as alteration is increased
593 from ~6 to ~15 wt.% is the result of a combination of the high thermal conductivity of
594 cristobalite and the reduction in porosity as a result of the void-filling mineral precipitation.

595

596 5.3 Volcanological implications

597 The heat flux from an active volcano provides crucial information on volcanic unrest
598 (Jessop et al., 2021) and provides an indication as to the likelihood of magmatic and phreatic
599 eruptions (Girona et al., 2021). As a result, processes that can alter the thermal properties of
600 dome- or edifice-forming rocks are of interest to those tasked with interpreting volcano heat
601 flux data. We have shown here that porosity (Figure 7) and alteration (Figure 8) can influence
602 the thermal properties of suites of volcanic rocks (basaltic-andesites, andesites, and rhyodacites)
603 from La Soufrière, Merapi volcano, and Chaos Crags. The main focus of this contribution is
604 the influence of alteration on thermal properties.

605 To explore the influence of alteration on the heat output of a dome, we can provide
606 estimates for the contribution of the heat flux from the dome, q , from conduction alone using
607 Fourier's law,

608

$$609 \quad q = -\lambda \frac{dT}{dz} \approx \lambda \frac{\Delta T}{H}, (4)$$

610

611 where λ is the thermal conductivity, T is the temperature, z is the vertical position in the dome
612 ($z = 0$ is the dome top and $z = H$ is the base of the dome), and ΔT is the difference in
613 temperature between the top and base of the dome. Although in a real volcanic setting, heat

614 may be transported by a number of additional mechanisms that would represent additional terms
615 in Equation (4), here we assume that heat transport is by conduction alone, which allows us to
616 investigate the relative influence of alteration on heat flux. We additionally neglect a
617 temperature-dependence of λ , which is valid at the relatively low temperatures of edifice rocks
618 (Whittington et al., 2009; Romaine et al., 2012) and assume that the dome is homogeneous. We
619 assume at dome height $H = 200$ m, and a temperature difference $\Delta T = 300$ °C. Based on our
620 experimental results (Figure 8a), we consider two scenarios: (1) a scenario in which alteration
621 decreases the thermal conductivity and (2) a scenario in which alteration increases the thermal
622 conductivity. For each scenario, we consider three dome states: (1) unaltered, (2) slightly
623 altered, and (3) altered. For the first scenario we assume thermal conductivities of 1.5, 1, and
624 $0.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the unaltered, slightly altered, and altered dome, respectively (Figure 7a).
625 And for the second scenario we assume thermal conductivities of 1, 1.25, and $1.5 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
626 for the unaltered, slightly altered, and altered dome, respectively (Figure 8a). The calculated
627 conductive heat flux for the first scenario are 2.25, 1.5, and $0.75 \text{ W}\cdot\text{m}^{-2}$, respectively, and 1.5,
628 1.875, and $2.25 \text{ W}\cdot\text{m}^{-2}$ for the second scenario, respectively. These simple calculations should
629 be considered for illustrative purposes only as they assume (1) that the dome is homogenous in
630 terms of its thermal conductivity, and (2) there is no temperature dependence of thermal
631 conductivity, a factor that can influence the thermal properties of volcanic rocks (Whittington
632 et al., 2009; Romaine et al., 2012). Nevertheless, for conductive heat transport alone, it is clear
633 from Equation (4) that the alteration-dependence of λ will have a linearly proportional impact
634 on q . Our results show that λ can change by up to a factor of three as alteration increases, which
635 will reduce q by a third. More importantly, our results suggest that dynamic changes in q
636 observed directly at volcanoes could represent changes in sub-surface alteration.

637 The mean ground heat flux measured at La Soufrière, for example, was measured to be
638 $406 \pm 24 \text{ W}\cdot\text{m}^{-2}$ in 2020 (Jessop et al., 2021). Therefore, although alteration-induced changes

639 to the thermal conductivity of the dome can influence the measured heat flux, as discussed
640 above, the contribution from conduction is likely much less than that from other heat transport
641 mechanisms, such as the convection of hydrothermal fluids within the core of the edifice;
642 although we note that conduction becomes the dominant mode of heat transfer between the
643 condensation isotherm and the surface (Harris, 2013), and that ongoing work at La Soufrière
644 supports this view (Lebas, 2021). However, changes in porosity resulting from alteration can
645 also modify permeability (Sruoga et al., 2004; Mayer et al., 2016; Heap et al., 2017b, 2019,
646 2020b; Revil et al., 2020; Kennedy et al., 2020; Heap et al., 2021b; Kanakiya et al., 2021),
647 which can influence the efficiency of the migration of hot fluids up through the dome structure
648 and therefore affect the measured volcano heat flux by increasing or decreasing the influence
649 of convection. Therefore, alteration can influence volcanic heat flux by changing the thermal
650 conductivity of the rocks, as explored in this contribution, and also by changing the ease of
651 convection within the dome by modifying permeability. A full description of heat transfer, in
652 which conduction and convection are solved together, that also incorporates the influence of
653 hydrothermal alteration on thermal properties, porosity, and permeability represents the logical
654 next step in unravelling the influence of alteration on volcano heat flux. Although this complete
655 description currently eludes us, we conclude with the available data that hydrothermal alteration
656 can influence volcano heat flux and should therefore be considered when interpreting heat flux
657 data collected at active volcanoes worldwide.

658

659 **6 Conclusions**

660 We have provided laboratory measurements of thermal conductivity, thermal
661 diffusivity, and specific heat capacity for variably altered volcanic rocks. These data show that
662 thermal conductivity and thermal diffusivity decrease as a function of porosity, and that specific
663 heat capacity does not change systematically as a function of porosity (as discussed in Heap et

664 al., 2020a). When plotted as a function of alteration (the percentage of secondary minerals), we
665 find that thermal conductivity decreases for the rocks from La Soufrière and Merapi volcano,
666 but appears to increase for the rocks from Chaos Crags. Although the thermal diffusivity of the
667 rocks from Chaos Crags increases as a function of alteration, the thermal diffusivity of the rocks
668 from La Soufrière and Merapi volcano does not appear to be influenced by alteration. The
669 specific heat capacity is not significantly affected by alteration, although there is a slight trend
670 of increasing specific heat capacity with alteration for the rocks from La Soufrière.

671 We conclude that the decrease in thermal conductivity as a function of alteration in the
672 rocks from La Soufrière and Merapi volcano is the result of the low conductivity of the
673 secondary mineral assemblage, and that a combination of the high thermal conductivity of
674 cristobalite and the reduction in porosity as a result of the void-filling mineral precipitation can
675 explain the increase in thermal conductivity in the rocks from Chaos Crags.

676 Although Fourier's law shows that an increase in alteration of a dome or edifice can
677 modify thermal conductivity and therefore heat flux, the contribution from conduction is likely
678 much less than that from other heat transport mechanisms, such as convection (see, for example,
679 Jessop et al., 2021). However, we conclude that not only is it important to be aware that
680 alteration can influence the thermal conductivity of the dome or edifice, but that alteration can
681 also influence permeability and therefore the convection of heat, a factor that can greatly modify
682 the heat flux measured at the volcano.

683 Because the heat flux of an active volcano provides crucial information about volcanic
684 unrest (Jessop et al., 2021; Girona et al., 2021), we conclude that the extent and, importantly, the
685 type of alteration should be routinely monitored. Monitoring can be performed by remote and/or
686 ground-based optical and spectroscopic methods (Crowley and Zimbelman, 1997; John et al.,
687 2008; Darmawan et al., 2018; Kereszturi et al., 2020; Mueller et al., 2021), gas and thermal
688 monitoring (Edmonds et al., 2003; Tamburello et al., 2019; de Moor et al., 2019; Jessop et al.,

689 2021; Moretti et al., 2020), geological mapping (van Wyk de Vries et al., 2000), magnetic
690 methods (Finn et al., 2007), deformation monitoring (Moretti et al., 2020), near-surface seismic
691 imaging (Amoroso et al., 2018), electrical tomography (Ahmed et al., 2018; Byrdina et al.,
692 2017; Ghorbani et al., 2018; Rosas-Carbajal et al., 2016), and muon tomography (Lesparre et
693 al., 2012; Rosas-Carbajal et al., 2017). A better understanding of hydrothermal alteration, and
694 how alteration can influence the thermal properties, porosity, and permeability of dome- and
695 edifice-forming rock, will allow for a deeper understanding of volcano heat fluxes and therefore
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697

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714

715 **CRedit author statement**

716 **Michael J. Heap:** Conceptualization; Methodology; Formal analysis; Investigation; Resources;

717 Writing - Original draft; Visualization; Supervision; Project administration; Funding

718 acquisition

719 **David Jessop:** Conceptualization; Investigation; Resources

720 **Marina Rosas-Carbajal:** Investigation; Resources; Project administration; Funding

721 acquisition

722 **Jean-Christophe Komorowski:** Investigation; Resources; Project administration; Funding

723 acquisition

724 **H. Albert Gilg:** Investigation

725 **Nadège Aron:** Investigation

726 **Margaux Buscetti:** Investigation

727 **Laura Gential:** Investigation

728 **Margaux Goupil:** Investigation

729 **Mathilde Masson:** Investigation

730 **Lucie Hervieu:** Investigation

731 **Alexandra R.L. Kushnir:** Methodology; Supervision

732 **Patrick Baud:** Investigation; Resources

733 **Lucille Carbillet:** Investigation; Resources

734 **Amy G. Ryan:** Investigation; Resources

735 **Roberto Moretti:** Resources; Project administration

736

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