1	A multidisciplinary approach to quantify the permeability of the
2	Whakaari/White Island volcanic hydrothermal system (Taupo
3	Volcanic Zone, New Zealand)
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5	Michael J. Heap ¹ , Ben M. Kennedy ² , Jamie I. Farquharson ¹ , James Ashworth ³ , Klaus
6	Mayer ⁴ , Mark Letham-Brake ⁵ , Thierry Reuschlé ¹ , H. Albert Gilg ⁶ , Bettina Scheu ⁴ , Yan
7	Lavallée ³ , Paul Siratovich ² , Jim Cole ² , Arthur D. Jolly ⁷ , Patrick Baud ¹ , Donald B. Dingwell ⁴
8	
9	¹ Géophysique Expérimentale, Institut de Physique de Globe de Strasbourg (UMR 7516
10	CNRS, Université de Strasbourg/EOST), 5 rue René Descartes, 67084 Strasbourg cedex,
11	France.
12	² Geological Sciences, University of Canterbury, Private Bag 4800, 8140 Christchurch, New
13	Zealand
14	³ Earth, Ocean and Ecological Sciences, University of Liverpool, Liverpool L69 3GP, United
15	Kingdom.
16	⁴ Earth & Environmental Sciences, Ludwig Maximilians Universität, Theresienstr. 41/III,
17	80333 Munich, Germany.
18	⁵ Kakapo Disaster Resilience, 2/48 Brockworth Place, Riccarton 8011, Christchurch, New
19	Zealand.
20	⁶ Lehrstuhl für Ingenieurgeologie, Technische Universität München, Arcisstr. 21, 80333
21	Munich, Germany.
22	⁷ GNS Science, 1 Fairway Avenue Avalon, 5040, New Zealand.
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26	Corresponding author: M.J Heap (heap@unistra.fr)

27 Highlights

28	•	Whakaari chiefly comprises lavas, lava breccias, ash tuffs, and tephra.
29	•	Porosity varies from ~0.01 to ~0.7 and permeability from ~10^{-19} to ~10^{-11} m^2.
30	•	The spatial distribution of rock types in crater walls and crater-fill deposits
31		identifies vertical and horizontal barriers to and pathways for fluid transport.
32	•	Pore- and fracture-filling precipitation and cementation can locally reduce
33		porosity and permeability and therefore augment pore pressure.
34	•	Our data will assist the modelling of deformation, seismicity, and explosive
35		behaviour at active hydrothermal systems worldwide.
36		

37 Abstract

38 Our multidisciplinary study aims to better understand the permeability of active 39 volcanic hydrothermal systems, a vital prerequisite for modelling and understanding 40 their behaviour and evolution. Whakaari/White Island volcano (an active stratovolcano 41 at the north-eastern end of the Taupo Volcanic Zone of New Zealand) hosts a highly 42 reactive hydrothermal system and represents an ideal natural laboratory to undertake 43 such a study. We first gained an appreciation of the different lithologies at Whakaari and 44 (where possible) their lateral and vertical extent through reconnaissance by land, sea, 45 and air. The main crater, filled with tephra deposits, is shielded by a volcanic 46 amphitheatre comprising interbedded lavas, lava breccias, and tuffs. We deployed field 47 techniques to measure the permeability and density/porosity of (1) more than 100 48 hand-sized sample blocks and (2) layered unlithified deposits in eight purpose-dug 49 trenches. Our field measurements were then groundtruthed using traditional laboratory 50 techniques on almost 150 samples. Our measurements highlight that the porosity of the 51 materials at Whakaari varies from ~ 0.01 to ~ 0.7 and permeability varies by eight 52 orders of magnitude (from $\sim 10^{-19}$ to $\sim 10^{-11}$ m²). The wide range in physical and 53 hydraulic properties is the result of the numerous lithologies and their varied 54 microstructures and alteration intensities, as exposed by a combination of macroscopic 55 microscopic (scanning electron microscopy) observations, quantitative and 56 mineralogical studies (X-ray powder diffraction), and mercury porosimetry. An 57 understanding of the spatial distribution of lithology and alteration style/intensity is 58 therefore important to decipher fluid flow within the Whakaari volcanic hydrothermal 59 system. We align our field observations and porosity/permeability measurements to 60 construct a schematic cross section of Whakaari that highlights the salient findings of 61 our study. Taken together, the alteration typical of a volcanic hydrothermal system can 62 result in increases (due to alteration-induced dissolution and fracturing) and decreases 63 (due to hydrothermal precipitation) to permeability. Importantly, a decrease in

64 permeability—be it due to fracture sealing in the lava, pore-filling alunite precipitation 65 in the tuffs, near-vent cementation by sulphur, and/or well-sorted layers of fine ash-66 can result in pore pressure augmentation. An increase in pore pressure could result in 67 ground deformation, seismicity, jeopardise the stability of the volcanic slopes, and/or 68 drive the wide variety of eruptions observed at Whakaari. Our systematic study offers 69 the most complete porosity-permeability dataset for a volcanic hydrothermal system to 70 date. These new data will inform and support modelling, unrest monitoring, and 71 eruption characterisation at Whakaari and other hydrothermally modified volcanic 72 systems worldwide.

73 **1. Introduction**

74 The permeability of the materials within a volcanic hydrothermal system 75 controls the ease at which exsolved magmatic volatiles can escape the magma-filled 76 conduit (Eichelberger et al., 1987; Jaupart, 1998; Collinson and Neuberg, 2012), as well 77 as the ingress, circulation, and interaction of meteoric and/or marine (in the case of 78 volcanic islands, coastal, or submarine volcanoes) water and hydrothermal fluids (Bibby 79 et al., 1995; Edmonds et al., 2003; Rowland and Sibson, 2004; Hurwitz et al., 2007). The 80 permeability of a volcanic hydrothermal system therefore exerts a first-order control on 81 the magnitude and distribution of pore pressure (Hurwitz et al., 2007; Todesco et al., 82 2010; Fournier and Chardot, 2012). The build-up of pore pressure within a volcanic 83 hydrothermal system can lead to catastrophic flank collapse (Day, 1996; Voight and 84 Elsworth, 1997; Reid et al., 2001; Reid, 2004; Moon et al., 2009; Procter et al., 2014), 85 seismicity (Nishi et al., 1996; Sherburn et al., 1998; Bean et al., 2014; Chardot et al., 86 2015), and/or drive a wide variety of eruptions, from phreatic (Barberi et al., 1992; 87 Kaneshima et al., 1996; Christenson et al., 2010; Mayer et al., 2015; Montanaro et al., 88 2016; Mayer et al., 2016a; 2016b) and phreatomagmatic (Bertagnini et al., 1991; 89 Houghton and Nairn, 1991) explosions to larger, more sustained eruptions (Houghton 90 and Nairn, 1991; Christenson, 2000; Deino et al., 2004). Detailed knowledge of the 91 permeability of the materials found within hydrothermal systems will therefore provide 92 a deeper understanding of their behaviour and evolution (Hurwitz et al., 2007; 93 Collombet, 2009; Todesco et al., 2010; Christenson et al., 2010; Fournier and Chardot, 94 2012; Collinson and Neuberg, 2012).

While laboratory studies have exposed porosity-permeability relationships for a
variety of volcanic rocks (*Klug and Cashman*, 1996; *Saar and Manga*, 1999; *Rust and Cashman*, 2004; *Mueller et al.*, 2005; *Wright et al.*, 2009; *Farquharson et al.*, 2015; *Kushnir et al.*, 2016; *Wadsworth et al.*, 2016; *Heap and Kennedy*, 2016), studies that
provide values of permeability for the highly altered lithologies that typically comprise

100 hydrothermal systems are scarce (Siratovich et al., 2014; Mayer et al., 2016a; 2016b), 101 largely due to their microstructural complexity and variability. To emphasise, 102 hydrothermal alteration can be the result of dissolution, mineral precipitation, and 103 secondary mineralisation (Browne, 1978) and can therefore result in increases or 104 decreases in porosity, a physical property known to exert a first-order control on 105 permeability (e.g., Bourbié and Zinszner, 1985). For example, the hydrothermal 106 alteration of volcanic rocks can replace the existing minerals with weak minerals such as 107 clays (e.g., kaolinite, illite, and smectite; Steiner, 1968; Browne, 1978; Inoue, 1995) and 108 sulphates (e.g., alunite and jarosite; Johnston, 1977; Ece et al., 2008), leading to material 109 weakening (del Potro and Hürlimann, 2009; Pola et al., 2012; 2014; Frolova et al., 2014; 110 Wyering et al., 2014; 2015; Heap et al., 2015a), fracturing/disintegration, and therefore 111 an increase in permeability. On the other hand, hydrothermal alteration can infill pores 112 and fractures (both micro- and macrofractures) with precipitated minerals (Edmonds et 113 al., 2003; Wyering et al., 2014; Heap et al., 2015a; Ball et al., 2015) leading to reductions 114 in permeability (e.g., Griffiths et al., 2016). The type and intensity of hydrothermal 115 alteration will not only depend on the rock type, but also on the temperature (Wyering 116 et al., 2014; Frolova et al., 2014) and composition (including pH) of the circulating fluids, 117 and the effective fluid-rock ratio (Browne, 1978; Reed, 1997), factors that can vary 118 tremendously in both space and time. The consequence of this variability is that the 119 rocks within a volcanic hydrothermal system will suffer a broad array of chemical 120 alteration that will modify their physical properties, such as porosity and permeability, 121 in different ways and to different degrees.

And so it is that, especially for volcanic hydrothermal systems, an understanding of the permeability—and therefore behaviour—must rely on a detailed comprehension of the variability in porosity and permeability of the accumulated materials, and information as to their spatial distribution. Whakaari volcano (an active stratovolcano located at the north-eastern end of the Taupo Volcanic Zone of New Zealand) hosts an 127 open, highly reactive hydrothermal system (hot springs and mud pools, fumaroles, acid 128 streams and lakes) (Giggenbach et al., 2003 and references therein) and represents an 129 ideal natural laboratory to undertake such a study. We present herein a 130 multidisciplinary study designed to better understand the permeability of an active 131 volcanic hydrothermal system. The primary goal of this contribution is to produce a 132 very large dataset to inform future modelling efforts. The permeability of the materials 133 within active volcanic hydrothermal systems is, for example, required to understand 134 and accurately model the outgassing of magmatic volatiles from the magma-filled 135 conduit (Collombet, 2009; Collinson and Neuberg, 2012), subsurface hydrothermal 136 activity and therefore volcanic unrest (Hurwitz et al., 2007; Peltier et al., 2009; Todesco 137 et al., 2010; Christenson et al., 2010 Fournier and Chardot, 2012; Christenson et al., 2016), 138 gas monitoring (Bloomberg et al., 2014; Peiffer et al., 2014), and volcano seismicity (Leet, 139 1988; Nishi et al., 1996; Sherburn et al., 1998; Bean et al., 2014; Chardot et al., 2015).

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141 **2.** Whakaari (White Island volcano)

142 Whakaari is an active andesitic-dacitic stratovolcano located 48 km north of 143 New Zealand's North Island, at the north-eastern end of the Taupo Volcanic Zone (Figure 144 1). The island, the tip of a much larger submarine structure, summits at 321 m above sea 145 level and measures 2.4 km east-west by 2 km north-south (Clark and Cole, 1986; 146 Houghton and Nairn, 1991 and references therein). Whakaari's subaerial structure 147 consists of two cones: the extinct and partially eroded Ngatoro Cone and the active, 148 amphitheatre-shaped Central Cone (Figure 1). Both are constructed from lavas, tuffs, 149 agglomerates, tephra, dykes, and breccias (Cole et al., 2000 and references therein). 150 Geomorphic evidence suggests that 0.21 km³ of material was prehistorically removed 151 from the Central Cone due to major flank failures involving weak hydrothermally 152 altered rock (Moon et al., 2009).

153 Whakaari hosts an open, highly reactive hydrothermal system that has existed 154 for approximately 10, 000 years (*Giggenbach et al.*, 2003 and references therein). The 155 mélange of marine/meteoric water and hot magmatic fluids generate acid brines 156 (Giggenbach et al., 2003 and references therein; Christenson et al., 2016) that rise to the 157 surface forming hot springs, mud pools, fumaroles, and acid streams and lakes. The pH 158 of the acid streams can be as low as ~ 2 (*Hedenquist et al.*, 1993; *Donachie et al.*, 2002). 159 Crater lake, a large lake of boiling acidic waters in the western subcrater, which appears 160 to vary in volume due to meteorological conditions and fluctuating levels of 161 hydrothermal activity (Christenson et al., 2016), is the focus of present day volcanic 162 activity and outgassing (e.g., Werner et al., 2008; Bloomberg et al., 2014), although more 163 diffuse outgassing through the crater floor and outgassing through fumaroles that line 164 the crater rim is also observed (*Bloomberg et al.*, 2014).

165 Volcanic eruptions at Whakaari are generally phreatic or phreatomagmatic in 166 style (although Strombolian activity occurred from the late seventies to the mid-167 eighties; Houghton and Nairn, 1991), and form discrete craters within the main crater-168 fill deposits of the Central Cone (Houghton and Nairn, 1991; Cole et al., 2000). Indeed, a 169 minimum of 28 phreatic or phreatomagmatic eruptions has occurred since 1826 (Mayer 170 et al., 2015). Prehistoric eruptions at Whakaari, recorded in the stratigraphy of crater 171 walls of the Central Cone, were characterised by the extrusion of lava (*Cole et al.*, 2000). 172 Eruptions at Whakaari are frequent to this day, the most recent occurring on April 27 173 2016.

Geophysical surveys on main crater floor highlight ground deformation episodes within the recently formed eruption centres and fumarole fields, interpreted as an increase in pore pressure due to the migration of hydrothermal fluids and/or shallow magma injection (e.g., *Peltier et al.*, 2009; *Fournier and Chardot*, 2012) and/or lake level variations (*Christenson et al.*, 2016). The circulation of hydrothermal fluids within these zones has been blamed for swarms of low-amplitude, high-frequency earthquakes and volcanic tremors (*Nishi et al.*, 1996; *Sherburn et al.*, 1998; *Chardot et al.*, 2015). Although
the permeability of the main crater floor has been inferred from thermal infrared
mapping (*Mongillo and Wood*, 1995) and gas and heat flux mapping (*Bloomberg et al.*,
2014), a systematic study that provides direct measurements of permeability, to our
knowledge, is not currently available.

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186 **3. Reconnaissance of Whakaari by land, sea, and air**

In order to assess the lateral and (where possible) vertical extent of the main
lithologies at Whakaari, and therefore select the most appropriate rocks for our study,
we lean on decades of reconnaissance missions of the island by land, sea, and air
(summarised in Figure 2). We also consulted historical records.

The modern-day crater walls chiefly comprise interbedded coherent (albeit fractured) and brecciated lavas dipping away from the centre of the island. These sequences of interbedded lava and lava breccia deposits can reach >100 m in thickness (Figure 2, picture 1). Significant debris from the lava breccia deposit from the northeastern wall of the volcanic amphitheatre (Figure 2, picture 7) can be seen at the base of the slope (Figure 3c).

197 Interbedded tuffs with a dominantly ash-sized particle size (hereafter called ash 198 tuffs) and coarser lithified pyroclastic deposits (lapilli tuffs) are exposed in perched 199 subcraters within the main crater walls (Figure 2; pictures 9, 10, and 11) and sequences 200 can reach >10 m in thickness. Bedded sequences of ash and lapilli tuffs, interbedded 201 with the lavas and lava breccias, dip away from the volcano, as seen in the eastern and 202 western sea cliffs of the volcano (Figure 2; pictures 10, 12, 15, 16). Coarse lithified 203 pyroclastic deposits also fill pre-existing valleys and are exposed in the cliffs on the 204 outer flanks of the volcano.

All of the interbedded deposits forming the crater walls are blanketed by layers of recent tephra deposits comprised of mostly well sorted ash and lapilli airfall (Figure 207 2; Figures 3a-b).

The crater rim is lined with active fumaroles, boiling mud pools, and acid streams (Figure 2; pictures 2, 4, and 6; Figures 3d-f). The crater-fill deposits found in these areas have been locally cemented by hydrothermal activity (frequently by sulphur or sulphates), forming lithified crusts (Figure 2; pictures 2, 4, and 6; Figures 3d-f). Fumaroles are encrusted by sulphur and sulphur flows are seen—albeit rarely—to extend from fumaroles containing subsurface pools of liquid sulphur (Figure 3f).

214 The main crater is filled with unlithified ash and lapilli deposits that are at least 215 several metres in thickness and consist of numerous thin beds (from several mm to 216 several cm in thickness) that can be easily distinguished based on their differences in 217 colour (Figure 3g). The crater floor is strewn with conspicuous rocky hummocks, the 218 debris from the collapse of the crater wall in September 1914 (Houghton and Nairn, 219 1991) (Figure 2, picture 3). Some areas close to the current crater lake—such as the 220 area near Donald Duck and Noisy Nellie craters (Figures 3h-i)—are blanketed by a well-221 sorted layer of fine ash ~ 100 mm in thickness. Eruptions as recent as 2016 have 222 carpeted the crater floor deposits with blocks (Figure 2; Figures 3g-i) and poorly sorted 223 surge deposits that are rapidly reworked into fluvial deposits draining away from the 224 main crater towards the sea (Figure 2; pictures 2 and 3).

The mapped surface exposure of deposits is further informed by historic accounts, photos, published articles, and volcano monitoring (GeoNet) reports outlining geomorphological changes to the crater. Eruptions have generated ~15 overlapping explosion craters and subsidence pits (up to 200 m deep) in the northern end of the main crater (*Houghton and Nairn*, 1989) that have been rapidly filled with eruption and erosional debris. Lava is occasionally seen within these craters either as a small dome (*Chardot et al.*, 2014) or as a distinct glow (*Houghton and Nairn*, 1989). Surges and ballistic debris generate tuff cones (*Jolly et al.*, 2016) in and around the modern crater
lake. Debris avalanches, landslides, and rockfalls deposit material on the crater floor and
create curvilinear collapse scars in the crater walls (e.g., *Moon et al.*, 2009). These
catastrophic events are further modified by erosion and shifting drainages, as
hydrothermal waters make their way south-east towards the ocean via surface streams.

Our reconnaissance has highlighted the extreme complexity and variability of the lithologies that form or are found within the crater at Whakaari. To best capture this variability we chose to (1) collect in excess of a hundred rock blocks for field and laboratory porosity-permeability testing and, (2) perform field porosity-permeability measurements on numerous vertical transects through the unlithified crater floor deposits. We also collected two unlithified crater floor samples for porositypermeability measurement in the laboratory.

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245 **4. Materials and methods for the lithified materials**

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247 *4.1 Description of the lithified materials*

248 We collected 124 blocks representative of the deposits that form or are found 249 within the crater at Whakaari: 115 hand-sized sample blocks (approximately 100 × 100 250 × 100 mm, although their shape varied; see Figures A1-A5) and nine larger blocks 251 (approximately $300 \times 300 \times 300$ mm). The hand-sized sample blocks were collected 252 from three sites located within the crater: the accessible scree at (1) the foot of the 253 eastern wall of the volcanic amphitheatre near Shark Bay, (2) Wilson's Bay and, (3) the 254 foot of the northern wall of the volcanic amphitheatre near Noisy Nellie crater (sampling 255 sites are shown on Figure 1). These sites were selected due to their accessibility and 256 because they contained blocks representative of the range of materials observed during 257 our reconnaissance (Figures 2 and 3). The locations on the volcanic slopes that fed these 258 scree deposits could be readily identified by fresh rockfall scars at each collection site.

259 The blocks consisted of variably altered ash tuffs (i.e., tuffs with an ash-sized particle 260 size), lavas, and lava breccias (Figures A1-A5). The hand-sized sample blocks were 261 collected for measurements of field density/porosity and permeability and, after coring 262 20 mm-diameter cylindrical samples from the blocks, measurements of porosity and 263 permeability in the laboratory. The goal of these measurements was to provide a large 264 porosity-permeability dataset; more detailed analyses (scanning electron microscopy 265 (SEM), X-ray powder diffraction (XRPD), and mercury porosimetry) were performed on 266 the nine larger blocks collected. These nine blocks, five of which were used in the recent 267 studies of *Heap et al.* (2015a) and *Mayer et al.* (2015), were selected to best represent 268 the variability seen at Whakaari (Figures 2 and 3). We collected one block of lava, one 269 block of lava breccia, four blocks of ash tuffs, one block of sulphur flow, one block of 270 sulphur crust, and one block of jarosite crust. Thin sections (double-polished) of each of 271 these rocks were made for SEM analysis and XRPD analysis was performed at the 272 Technische Universität München (TUM, Germany) on powdered material of each of the 273 blocks. For the XRPD analysis, powdered samples were mixed with an internal standard 274 (10% ZnO) and ground for 8 min with 10 ml of isopropyl alcohol in a McCrone 275 Micronising Mill using agate cylinder elements. The XRPD analyses were performed on 276 powder mounts using a PW 1800 X-ray diffractometer (CuKα, graphite monochromator, 277 10 mm automatic divergence slit, step-scan 0.02° 20 increments per second, counting 278 time 1 s per increment, 40 mA, 40 kV). The crystalline and amorphous phases in the 279 whole rock powders were quantified using the Rietveld program BGMN (Bergmann et 280 al., 1998). To confirm the presence of any identified clay minerals, we also separated < 2281 µm fractions by gravitational settling and prepared oriented mounts that were X-rayed 282 in an air-dried and ethylene glycolated state.

The porphyritic andesite lava (WI20) contains a groundmass of hydrated amorphous glass embedded with phenocrysts and microlites of feldspar and pyroxene. The lava is locally fractured and oxidised (Figure 4a). The block was collected at the foot 286 of the eastern wall of the volcanic amphitheatre near Shark Bay (Figures 1, 2, and 3a-b) 287 and likely therefore originates from one of the competent lava units of the currently 288 active Central Cone (Cole et al., 2000). Scanning electron microscopy (SEM) has revealed 289 the presence of pervasive microcracking and minor precipitation of alteration minerals 290 (jarosite and gypsum) into some of the pores (Figure 4a). The pores are typically a 291 couple of hundred microns in diameter, although some phenocrysts have microporous 292 rims (Figure 4a). XRPD analysis highlights the main mineralogical components to be 293 plagioclase (37 wt.%), cristobalite (14 wt.%), amorphous silica (14 wt.%), K-feldspar 294 (14 wt.%), and pyroxene (10 wt.%) (Table 1).

295 The altered lava breccia collected (WI30) was taken from the deposit shown in 296 Figure 3c (located within the debris from the partial collapse of the north-eastern wall 297 of the volcanic amphitheatre (Figure 2, picture 7)). Although some of the clasts within 298 the deposit were on the order of 1 m (Figure 3c), we collected material that contained a 299 maximum clast size of ~ 10 mm in diameter due to the small size of our laboratory 300 samples (20 mm in diameter). The groundmass of the lava breccia is brown-red in 301 colour (due to oxidation) and hosts numerous angular lava fragments (Figure 41). SEM 302 analysis shows microcracks, zones of microlitic groundmass, and pores up to 500 303 microns in diameter (Figure 41). The sample consists of amorphous silica (74 wt.%), 304 kaolinite (10 wt.%), and plagioclase (8 wt.%) (Table 1).

305 The four ash tuff deposits are composed of partially cemented ash particles, 306 typically altered crystal fragments or completely opalised shards of glass (now 307 composed dominantly of hydrated amorphous silica). The ash tuff blocks collected were 308 selected based on observable differences in colour (alteration type), macroscopic 309 textures (presence/absence of bedding or laminations), particle size, and degree of 310 cementation. They were all collected in the accessible scree at the foot of the eastern 311 wall of the volcanic amphitheatre near Shark Bay (Figures 1, 2, and 3a-b). The ash tuffs 312 can be subdivided into two subgroups: fine-grained white deposits containing pore313 filling alunite (WI21 and WI24) and coarser-grained, bedded deposits that do not 314 contain alunite (WI22 and WI23). Of the deposits containing alunite (WI21 and WI24; 315 Table 1), WI21 contains a lower porosity grey zone (Figure 4b) and a higher porosity 316 white zone that contains gas elutriation pipes (Figure 4c). All gas elutriation pipes were 317 orientated perpendicular to bedding (Figure 4c). WI21 and WI24 contain average 318 particle diameters of ~ 0.1 and ~ 0.2 mm, respectively, and an average pore diameter of 319 \sim 100-200 microns (Figures 4b-c and 4f). Pore-filling alunite (Figure 5) likely 320 precipitated following complete devitrification and opal replacement (Heap et al., 321 2015a). Of the bedded deposits (WI22 and WI23), the grey-coloured WI22 contains an 322 average particle diameter of ~0.3 mm and 10 mm-thick alternating bands of high- and 323 low-porosity (Figure 4d). WI23 contains alternating 3-10 mm-thick alternating brown-324 red and grey coloured bands (Figure 4e). The average particle diameter varies between 325 the different bands in WI23, from \sim 0.3-0.4 mm in one band to \sim 0.1 mm in another 326 (Figure 4e). Both WI22 and WI23 contain and an average pore diameter of ~100-200 327 microns (Figures 4d and 4e). The mineral composition of all four ash tuffs is given in 328 Table 1. While these tuffs can be described as *completely* altered (they contain none of 329 their original mineral phases; classified following British Standard practice BS5930, 330 1999), the presence of alunite attests to the mobility of aluminium and therefore 331 precludes the use of chemical alteration indices such as the Chemical Index of Alteration 332 (CIA), the Chemical Index of Weathering (CIW), and the Plagioclase Index of Alteration 333 (PIA) (see *Pola et al.*, 2012).

The remaining three lithologies represent those that have been lithified as a result of their proximity to acid streams and fumaroles (Figures 2 and 3d-f). They are: sulphur flow, sulphur crust, and jarosite crust. All of these blocks were collected at the south-western margin of the volcanic amphitheatre (Figures 2 and 3d-f). The sulphur flow is a very dense rock that forms a layer ~100 mm in thickness located next to an active sulphur fumarole (Figure 3f). SEM (Figure 4k) and XRPD (Table 1) analyses 340 shows that the rock comprises few angular ash fragments that are efficiently cemented 341 together by sulphur. Pores are difficult to distinguish, although microcracks are present 342 (Figure 4k). The sulphur crust block was collected adjacent to the sulphur flow (Figure 343 3f). The sulphur crust is very heterogeneous and contains ash and lapilli fragments (of 344 devitrified glass, pyroxene, and plagioclase; Table 1) cemented by sulphur (Figure 4h). 345 The sulphur gives the rock its light-green/chartreuse colour. Pores in the sulphur crust 346 are typically \sim 400-500 microns in diameter (Figure 4h). The jarosite crust is similarly 347 heterogeneous, containing ash and lapilli fragments (of devitrified glass, pyroxene, 348 plagioclase, and cristobalite; Table 1) cemented by jarosite (Figure 4g). Pores in the 349 jarosite crust are typically a couple of hundreds microns in diameter (Figure 4h), but 350 can reach 1 mm in diameter. The mineral composition of all three deposits is given in 351 Table 1.

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- 353 *4.2 Methods for the lithified materials*
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355 4.2.1 Field methods

356 Following several hours of drying in the sun, the permeability of the 115 blocks 357 was estimated using a portable air permeameter (Vindum Engineering TinyPerm II). 358 The TinyPerm unit estimates the permeability by evacuating the air from the rock at the 359 nozzle-rock interface (inner diameter of the nozzle = 10 mm) and monitoring the 360 response function of the transient vacuum. The TinyPerm is a useful field tool that 361 quickly provides an estimate of the permeability of a porous rock, and has been recently 362 used in studies on volcanic materials (Farguharson et al., 2015; Schaefer et al., 2015; 363 Kendrick et al., 2016). Although the TinyPerm II unit cannot provide values of permeability below $6.92 \times 10^{-16} \text{ m}^2$, it has been shown to be reasonably reliable in the 364 range 10⁻¹² to 10⁻¹⁵ m² (*Farquharson et al.*, 2015). However, we note that some of our 365 366 bedded ash tuff blocks were rectangular prisms (~50-60 mm in thickness); as a result,

367 permeability estimates on these blocks were likely overestimated by the TinyPerm due 368 to the volume of rock required for near-surface measurements of permeability 369 (Selvadurai and Selvadurai, 2010). Estimates of permeability using the TinyPerm unit 370 are not necessarily reliable for small (<<100 mm) samples. Some of the blocks collected 371 appeared to have low-permeability skins; when present, we tried to remove the skin 372 with a file prior to measurement. The permeability of each block was estimated using 373 the mean of 3-4 TinyPerm values measured at different locations on the surface of the 374 block.

375 The bulk density of each of the 115 hand-sized sample blocks was estimated 376 using the Archimedean weighing technique similar to that employed by *Kueppers et al.* 377 (2005) and Farquharson et al. (2015). The dry weight Wd of each block was first 378 measured using a balance (precision 0.1 g). The apparent immersed weight Wi was then 379 measured by lowering each block into a water-filled bag suspended underneath a 380 balance mounted on a tripod. The immersed weight was taken quickly to avoid water 381 imbibition. The dry bulk density ρ_b of each block could then be determined using the 382 following relation (assuming that the density of water = 1000 kg/m^3):

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$$\rho_b = \frac{Wd}{Wd - Wi} \quad (1)$$

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Four of the samples (WI-F-16, 46, 59, and 77; Table A2) floated and we could nottherefore determine their dry bulk density.

These dry bulk density values were converted to values of total porosity by measuring the solid density of each block. Once back in the laboratory, the blocks were cored to provide 20 mm-diameter cylindrical samples for laboratory measurements of porosity and permeability (see next section). The remainder of each block was powdered using a pestle and mortar. The solid density ρ_s (the density of each powder) was then measured using a helium pycnometer (Micromeritics AccuPyc II 1340). Totalporosity was then determined for each block using the following relation:

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$$\phi_t = 1 - \left(\frac{\rho_b}{\rho_s}\right) \quad (2)$$

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The total porosity of the four samples that floated (WI-F-16, 46, 59, and 77; Table A2)
was estimated by assuming a bulk dry density of 1000 kg/m³.

398

399 4.2.1 Laboratory methods

400 While field methods provide quick estimates for values of permeability and dry 401 bulk density, such methods should always be groundtruthed by well-constrained 402 measurements in the laboratory. Cylindrical samples (20 mm in diameter) were cored 403 from the 115 hand-sized sample blocks and were precision ground so that their end 404 faces were flat and parallel (Figures A6-12). Samples were cored perpendicular to 405 bedding, where present. The length of each core, which varied from ~ 22 to ~ 40 mm, 406 was a function of the size and shape of the block. The length to width ratio of our 407 samples was therefore greater than one in each case (length to width ratios lower than 408 one are not recommended for laboratory permeability measurements). The samples 409 were washed using water and then dried at 40 °C in a vacuum oven for at least two days. 410 The connected porosity of each core sample was measured using a helium pycnometer 411 (Micromeritics AccuPyc II 1340). The samples were then jacketed (in a rubber jacket) 412 and their permeability measured using a benchtop steady-state permeameter (see 413 Farquharson et al., 2016; Heap and Kennedy, 2016) under a confining pressure of 1 MPa 414 and at ambient laboratory temperature. A confining pressure during routine 415 measurements of permeability is required to prevent gas travelling between the sample 416 and the jacket. Samples were first left for one hour at a confining pressure of 1 MPa to 417 ensure microstructural equilibrium. Flow rate measurements were taken (using either a 418 low- or high-flow gas flowmeter, depending on the permeability of the sample) under 419 several pressure gradients (typically from 0.05 to 0.2 MPa, equating to flow rates 420 between 0.2 and 400 ml/min) to determine the permeability using Darcy's law and to 421 assess the need for the Forchheimer and/or the Klinkenberg corrections, which were 422 applied on a case-by-case basis. The Klinkenberg correction corrects for gas slippage 423 (Klinkenberg, 1941), and was typically applied to low-porosity, low-permeability 424 samples with a high microcrack density. The Forchheimer correction corrects for flow 425 inertia and was typically employed for high-permeability samples that had to be 426 measured using high flow rates.

427 Cylindrical samples (20 mm in diameter) were also taken from the nine larger 428 blocks (WI20-26 and WI29-30) and precision ground to a nominal length of 40 mm 429 (Figures 4 and A6-12). Samples of the lava (WI20) were cored so as to contain none or 430 one macroscopic (i.e., sample size) fracture along their axis (Figure 4a). Samples of ash 431 tuff WI21 were prepared to contain (1) no gas elutriation pipes (Figure 4b), (2) gas 432 elutriation pipes parallel to the sample axis (Figure 4c) and, (3) gas elutriation pipes 433 perpendicular to the sample axis (Figure 4c). Samples WI22 and WI23 were cored 434 perpendicular to the bedding direction (Figures 4d and 4e). The porosity and 435 permeability of these samples were measured using the same techniques described 436 above (data are available in Tables A4-A6). Further, to understand the influence of 437 confining pressure (i.e., depth) on permeability, we performed experiments in which we 438 sequentially measured the permeability of a sample of lava (WI20) and ash tuff (WI21) 439 under confining pressures of 1, 2, 5, 10, 15, 20, 25, and 30 MPa (data available in Table 440 A7).

To explore their microstructure further, mercury injection tests were performed
on pieces (2-5 g) of lava (WI20) and ash tuff (WI21-24) using the Micromeritics
Autopore IV 9500 at the University of Aberdeen (Scotland). The evacuation pressure
and evacuation time were 50 μmHg and 5 minutes, respectively, and the mercury filling

pressure and equilibration time were 0.52 pounds per square inch absolute (psia) and
10 seconds, respectively. The pressure range was 0.1 to 60000 psia (i.e., up to a pressure
of about 400 MPa). Mercury injection data permit the calculation of the pore throat size
distribution within a particular sample.

449

450 **5. Materials and methods for the unlithified materials**

451

452 5.1 Field materials and methods for the unlithified materials

453 Eight localities within the recent, unlithified deposits of the crater floor at 454 Whakaari were chosen for porosity and permeability analysis: four close to the crater 455 lake (sites SP01, SP02, SP04, and SP07), two in a large gulley on the north-eastern wall 456 of the volcanic amphitheatre (sites SP06 and SP08), one near Troup Head (site SP05), 457 and one near Crater Bay (SP03) (localities shown in Figure 6). At each location, shovels 458 were used to expose up to several metres of fresh, vertical stratigraphy. Three of the 459 near-crater localities (SP01, SP02, and SP04) exposed a similar stratigraphy (Figure 6). 460 The base of these deposits, only exposed at trench SP02, consists of a yellow-coloured 461 ash/lapilli layer that contains a high percentage of large (~50 mm) lapilli fragments 462 overlain by a similarly coarse black-coloured horizon of ash/lapilli. Above this coarse 463 basal layer is a series of interbedded red- and grey/black-coloured deposits that likely 464 represent cyclic phreatic to phreatomagmatic eruptions. These layers contain a 465 noticeably smaller average particle size than the coarse basal layer. We interpret red-466 and yellow-coloured layers to be more hydrothermally altered than those black in 467 colour. The deposits of the 2013 phreatic eruption mark the top of these crater floor 468 deposits. The final near-crater trench (SP07) is located in a gully (several metres deep) 469 draining southeast from Donald Duck crater. The flanks of this gully show signs of 470 outgassing activity and the deposits are hydrothermally altered as a result. The deposits 471 at this locality are yellow-grey in colour and contain few large lapilli fragments. The two

472 sites chosen on the north-eastern wall of the volcanic amphitheatre (SP06 and SP08) 473 were selected due to their differences in alteration. Further, these sites are both located 474 within the 2004 landslide deposit that has an anonymously low gas flux (Bloomberg et 475 al., 2014). Site SP06 contains a hydrothermally altered grey/yellow-coloured ash/lapilli 476 debris flow deposit that contains large (~ 50 mm) angular lapilli fragments below a 477 laminated black/grey-coloured ash/lapilli deposit. Site SP08 comprises a grey-coloured 478 ash/lapilli debris flow deposit that contains large (~50 mm) angular lapilli fragments. 479 To measure characteristic reworked fluvial deposits not affected by hydrothermal 480 alteration, we selected two sites located far from the crater lake and the volcanic 481 amphitheatre rim (SP03 and SP05). Site SP03 contains a grey-coloured ash/lapilli 482 deposit that is overlain by a similar deposit that contains large (\sim 50 mm) angular lapilli 483 fragments. Site SP05 consists of a brown-coloured ash/lapilli deposit.

484 A PL-300 soil permeameter (Umwelt-Geräte-Technik) was used to measure the 485 permeability of the layers of unlithified material (for more information see Mayer et al., 486 2016b). Samples were first taken from the exposed stratigraphy using stainless steel 487 cylinders (of diameter 72 mm and length 61 mm; see inset in Figure 6). The 488 permeability could then be obtained using Darcy's law by measuring the volumetric flow 489 rate of air through the sample. An internal vacuum pump produces the inflow of air 490 through the sample, which is defined over a calibrated throat in the apparatus. The 491 pressure difference over the sample is recorded by a sensor, which provides the pressure 492 gradient of that flow. Comparison of the pressure gradient, in respect to a second 493 gradient over the calibrated permeability of the internal throat, enables the 494 determination of the volumetric flow rate of air through the sample and consequently 495 the estimation of its permeability. The permeability of at least two samples from each 496 distinguishable layer was measured (Table A3). We also measured the temperature of 497 each sample. The samples were then weighed and wrapped for transport.

Once back in the laboratory, the samples were oven-dried and their dry weight measured. The dry bulk density ρ_b could then be determined using the dry weight and the volume of the stainless steel cylinder. The solid density ρ_s (the density of the powder) was then measured using a helium pycnometer (Ultrapyc 1200e Quantachrome) and the total porosity determined using Equation 2. Finally, the water content of each sample was calculated using the *in-situ* and the oven-dry weights.

504

505 5.2 Laboratory materials and methods for the unlithified materials

506 Two unlithified samples were collected for permeability measurement in the 507 laboratory. The first was a poorly sorted unlithified tephra deposit, the most abundant 508 material of the crater fill (Figures 2 and 3g). The deposit (WI27; Figure 4i) was collected 509 at a depth of one metre at the locality indicated in Figure 1. The deposit was first oven-510 dried. A portion of the deposit was then poured into the rubber jacket and permeability 511 was measured using the above-described procedure. Due to the size of our rubber 512 jackets (inner diameter of 20 mm), large lapilli were first removed. The ash/lapilli 513 sample (WI27; Figure 4i) comprises mainly amorphous silica (59 wt.%), plagioclase (15 514 wt.%), pyroxene (9 wt.%), alunite (8 wt.%), and cristobalite (7 wt.%) (Table 1). The 515 porosity was measured by determining the dry bulk density of the deposit within the 516 jacket and the solid density (using the helium pycnometer); the total porosity could then 517 be calculated using Equation 2. The unlithified ash deposit collected (WI28; Figure 4j) 518 formed part of the blanket of fine ash that covers large parts of the crater floor near the 519 crater lake (Figures 2 and 3h-i). The fine ash sample (WI27; Figure 4i), collected from 520 Donald Duck crater (see Figure 1 for the collection locality), comprises mainly 521 amorphous silica (52 wt.%), pyroxene (20 wt.%), plagioclase (18 wt.%), cristobalite (7 522 wt.%), and gypsum (3 wt.%) (Table 1). In order to measure the *in-situ* permeability, we 523 carefully wrapped the sample upon collection to preserve the *in-situ* water content 524 (which was calculated to be ~ 0.25 by measuring the *in-situ* weight and oven-dry weight

525 of a portion of the deposit). Once back in the laboratory, cylindrical samples (20 mm in 526 diameter and nominally 40 mm in length) were taken by pushing a cylindrical metal 527 sleeve into the sample. These cylindrical samples were then quickly and carefully 528 jacketed (using a rubber jacket) and loaded into the permeameter. Due to the low 529 permeability of the ash sample, the pressure decay method (*Brace et al.*, 1968) was used 530 to determine their permeability. To do this, an initial increase (0.5 MPa) of the upstream 531 pressure was applied to the sample and the fluid inlet closed. The permeability was then 532 derived using the temporal decay of the upstream pressure. Owing to the delicate nature 533 of WI27 and WI28, permeability measurements were conducted at confining pressures 534 of 0.3 and 0.5 MPa, respectively.

535

536 **6. Results**

537

538 6.1 Field porosity and permeability of the lithified materials

539 Permeability as a function of dry bulk density for the 115 hand-sized sample 540 blocks is shown in Figure 7a (Tables A1 and A2). We find that the dry bulk density and 541 permeability of the samples varies between 1000 and 2000 kg/m³ and between $\sim 10^{-15}$ 542 and $\sim 10^{-11}$ m², respectively (Figure 7a). When dry bulk density is converted to total 543 porosity we find values between ~ 0.1 and ~ 0.6 (Figure 7b). We find no discernible 544 trends in the bulk density-permeability (Figure 7a) or porosity-permeability (Figure 7b) 545 data, although we note that the lava and lava breccia samples (black circles) are 546 typically denser/less porous than the ash tuffs (white circles) (Figure 7).

547

548 6.2 Field porosity and permeability of the unlithified materials

The calculated total porosity of the unlithified deposits is plotted as a function of their stratigraphic depth in Figure 8a. We find that porosity ranges from ~0.25 to ~0.75. There is no strong correlation between total porosity and the depth of the deposit (up to 552 a maximum depth of \sim 140 cm; Figure 8a). There is however a strong correlation 553 between water content (calculated using the *in-situ* and dry weight of the sample) and 554 depth (Figure 8b): water content increases linearly with depth. As a result, the air-filled 555 porosity in the deposits decreases as depth increases (Figure 8c). Based on these data 556 (Table A3), the water table within the crater-fill deposits at Whakaari is at a depth of 557 about 130-140 cm. We also note that the temperature of the deposit increases with 558 increasing depth, from \sim 22 °C at the surface to \sim 50-70 °C at a depth of \sim 100 cm (see 559 inset in Figure 8c). The data of Figure 8 are available in Table A3.

Since the gas moving through the sample can only travel through the air-filled porosity on the timescale of the measurement, we have chosen to plot permeability as a function of air-filled porosity (Figure 9a). We see that, generally, permeability increases with increasing air-filled porosity. As a result, and although there is a lot of scatter, permeability is seen to decrease with depth (Figure 9b). The data of Figure 9 are available in Table A3.

566

567 6.3 Laboratory porosity and permeability of the lithified materials

568 Permeability as a function of connected porosity for the 136 20 mm-diameter 569 samples is shown in Figure 10 (Tables A4-A6). We find that the connected porosity and 570 permeability of the samples varies between ~0.1 and ~0.7 and between ~ 10^{-15} and ~ 10^{-15} 571 ¹¹ m², respectively (Figure 10). If one considers the lava and lava breccias and the ash 572 tuffs separately, we find that, for both rock types, permeability increases as porosity 573 increases (Figure 10). In detail, increases in lava permeability are large as connected 574 porosity is increased when the porosity is low (less than ~ 0.15) and small as connected 575 porosity is increased when the porosity is high (greater than ~ 0.15) (Figure 10). All of 576 the lava breccia samples contain a porosity > 0.2 and therefore form part of the latter 577 trend. We also note that the jarosite (orange circles) and sulphur (green circles) crusts 578 do not plot within the broad porosity-permeability trend defined by the ash tuff 579 samples: the jarosite crust samples in particular were considerably more permeable 580 than ash tuffs of a similar porosity (Figure 10; Table A6). The permeability of the 581 prepared sulphur flow samples was too low for measurement in our benchtop 582 permeameter (permeability < 10^{-18} m²) (Table A6).

583

584 6.3.1 Influence of macrocracks on the permeability of lava

The presence of a macrocrack (parallel to the direction of flow) in samples of lava serves to greatly increase their porosity and permeability (Table A4). Porosity increased from 0.036 to between 0.092 and 0.112 when a macrocrack was present. Permeability increased from 1.6×10^{-16} m² (macrocrack-free sample) to 1.4×10^{-14} and 1.1×10^{-13} m² for the macrocracked samples containing porosities of 0.092 and 0.112, respectively (Table A4). We use here the two-dimensional model proposed by *Heap and Kennedy* (2016) to determine their fracture permeabilities k_f :

592

$$k_f = \frac{Ak_e - A_{intact} \cdot k_0}{A_{fracture}} \quad (3)$$

593

Where *A*, $A_{fracture}$ and A_{intact} , are the areas of the sample end face, the fracture, and the intact portion of the sample end face, respectively, and k_e and k_0 are the equivalent (i.e., the permeability of the fractured sample) and intact permeabilities, respectively. Assuming a constant throughgoing fracture width of 0.5 mm, we calculate fracture permeabilities of 4.3×10^{-13} and 3.3×10^{-12} m² for the macrocracked samples containing porosities of 0.092 and 0.112, respectively (Table A4).

600

601 6.3.2 Influence of gas elutriation pipes on the permeability of ash tuff

To assess the influence of gas elutriation pipes on the permeability of the ash tuffs, permeability was measured on samples of WI21b containing gas elutriation pipes orientated either parallel or perpendicular to the direction of fluid flow (Figure 11; Table A5). Although the sample with the highest permeability $(3.1 \times 10^{-15} \text{ m}^2)$ contains gas elutriation pipes parallel to flow, we note that two similarly prepared samples have permeability values ~10⁻¹⁶ m² (Figure 11; Table A5). It is difficult therefore to draw firm conclusions as to the influence of gas elutriation pipes on the permeability of the ash tuff from these data.

610

611 6.3.3 Influence of confining pressure on permeability

612 The influence of confining pressure (up to 30 MPa) on the permeability of a lava 613 sample (WI20; without macrocracks) and an ash tuff sample (WI21b; parallel gas 614 elutriation pipes) is presented in Figure 12 (data available in Table A7). Assuming a 615 constant bulk density for WI20 and WI21 of 2500 and 1500 kg/m³, respectively, the 616 depth at a pressure of 30 MPa was estimated using $P = \rho g z$ to be ~1.2 and ~2 km for 617 WI20 and WI21, respectively. The reduction of permeability with increasing pressure is 618 much higher for the lava than for the ash tuff sample (Figure 12). The permeability of 619 the ash tuff was reduced from 7.6 \times 10⁻¹⁵ m² at a confining pressure of 1 MPa down to 620 4.7×10^{-16} m² at 30 MPa. In the same pressure range, the permeability of the lava 621 deposit was reduced from 1.0×10^{-16} to 2.4×10^{-17} m². In both cases, the largest 622 permeability decrease was seen as the confining pressure was increased from 1 to 2 623 MPa. Following this initial decrease, the permeability decreased more-or-less 624 monotonously with increasing confining pressure (Figure 12).

625

626 6.3.4 Pore throat diameter of lava and ash tuffs

The pore throat diameter distributions, as measured by mercury injection, for the lava sample (WI20) and the four ash tuffs (WI21, WI22, WI23, and WI24) are presented in Figure 13. For the lava we find that only 5% of the pore throats have a diameter larger than 0.2 mm; the majority of the pore throats (95%) are between 0.2 and 0.004 mm in diameter. The most abundant pore throat diameter for WI20 is 0.006 632 mm. The pore throat diameter distributions for the ash tuffs are very different to that of 633 the lava: the ash tuffs contain a much larger proportion of larger pore throats (Figure 634 13). Indeed, pore throats greater than 100 μm were measured in the ash tuffs. The pore 635 throat diameter distributions for the different ash tuffs are quite similar, although we 636 note that WI24 contains a lower proportion of small (< 0.04 mm) pore throats than the 637 other ash tuffs. The most abundant pore throat diameters are 0.1, 0.006, 0.05, and 0.04 638 mm for WI21, WI22, WI23, and WI24, respectively (Figure 13).

639

640 6.4 Laboratory porosity and permeability of the unlithified materials

The total porosity of the unlithified ash/lapilli (dried) and the fine ash (dried) were calculated to be 0.54 and 0.45, respectively. Since the *in-situ* water content of the fine ash is ~0.25, the air-filled porosity of the *in-situ* deposit is estimated to be ~0.2. The permeability of the unlithified ash/lapilli (dried) and the fine ash (*in-situ* water content = ~0.25; air-filled porosity = ~0.2), as measured in the laboratory, were 3.36×10^{-12} and 4.50×10^{-19} m², respectively.

647

648 **7. Discussion**

649

650 7.1 Laboratory versus field measurements

651 The total porosity and permeability determined in the field is compared with 652 laboratory measurements on the samples cored from the same block in Figure 14. We 653 find that laboratory porosity is typically slightly higher than the porosity measured in 654 the field, but that there is no systematic variation with increasing porosity (Figure 14a). 655 It is likely that dry bulk density is overestimated using the field technique due to the 656 imbibition of water, an offset that could be corrected empirically (Farquharson et al., 657 2015). However, estimates of permeability using the TinyPerm II unit over- or 658 underestimated the permeability of the measured rocks by two or in some cases three

659 orders of magnitude (Figure 14b). For the most part, the permeability measured by the 660 TinyPerm II unit overestimated the permeability; this is perhaps due to the fact that the 661 permeability of some blocks was too low to be measured by the TinyPerm II (although 662 the unit gives a value regardless). Overestimations could also be due to an imperfect seal 663 between the rock and the nozzle due to an uneven rock surface and/or due to the 664 presence of a low-permeability skin (although, as noted above, we tried to remove low-665 permeability skins prior to measurement). It is clear from these data that, although the 666 TinyPerm II unit offers a quick and easy estimate of permeability, such estimates should 667 be groundtruthed by well-constrained laboratory data.

668

669 7.2 Porosity and permeability relationships in the lithified materials

670 Generally speaking, the permeability of porous media increases with increasing 671 connected porosity (e.g., Bourbié and Zinszner, 1985; Wadsworth et al., 2016). However, 672 the link between porosity and permeability is not straightforward, as permeability does 673 not strictly rely on porosity, but on the connectivity and geometry of the porosity (crack 674 porosity versus pore porosity, pore and crack geometries, pore and crack tortuosity, 675 amongst others). In other words, low-porosity rocks can have a high permeability, and 676 vice versa. Volcanic rocks in particular display a wide range of microstructure and 677 laboratory studies have exposed porosity-permeability relationships for a variety of 678 volcanic rocks (Klug and Cashman, 1996; Saar and Manga, 1999; Rust and Cashman, 679 2004; Mueller et al., 2005; Wright et al., 2009; Farquharson et al., 2015; Kennedy et al., 680 2016; Kushnir et al., 2016; Heap and Kennedy, 2016). Few laboratory studies however 681 have measured the porosity and permeability of hydrothermally altered volcanic rocks 682 (e.g., Siratovich et al., 2014; Mayer et al., 2016a; 2016b). Our study has shown that the 683 porosity-permeability relationships for the hydrothermally altered materials that form 684 Whakaari are complex (Figure 10). Due to the limitations of TinyPerm field

permeameter (Figure 14b), in the following discussion we will focus solely onlaboratory measurements of porosity and permeability (Figure 10).

687 We also highlight that the measurements of porosity and permeability provided 688 herein were determined using gas (helium and nitrogen, respectively). It is well known 689 that measurements of permeability will be influenced by the presence of clays (*Faulkner* 690 and Rutter, 2000; Tanikawa and Shimamoto 2009), which is the case for two of the 691 samples collected (WI23 and WI30; Table 1). The measurements of permeability 692 provided here will therefore overestimate the permeability of these samples to water, 693 since water will serve to swell the clays present within the rock and hence reduce the 694 permeability. However, it is unclear at present the influence of hydrothermal brines and 695 seawater on the permeability of clay-bearing rocks.

It is also important to note that laboratory measurements of permeability are scale-dependent (*Brace*, 1984; *Clauser*, 1992; *Neuman*, 1994; *Heap and Kennedy*, 2016; *Farquharson et al.*, 2016). Due to the small size of laboratory samples, measurements of permeability in the laboratory do not account for macroscopic features (such as macroscopic fractures or bedding). They will therefore under- or overestimate the equivalent permeability if the macroscopic feature provides a pathway or a barrier to fluid flow, respectively.

703

704 7.2.1 Porosity and permeability relationships in the lavas and lava breccias

We find that a single power law cannot describe the porosity-permeability trend
for the lavas and lava breccias (Figure 10). While porosity-permeability relationships for
volcanic rocks have been classically described using a single power law model (e.g., *Mueller et al.*, 2005), recent studies have invoked a double power law model
(*Farquharson et al.*, 2015; *Heap et al.*, 2015b; *Kushnir et al.*, 2016; *Heap and Kennedy*,
2016). The double power law model consists of two discrete power laws that intersect
at a so-called "porosity changepoint" *x**. The use of two power laws in these studies, as

712 opposed to one, has been statistically verified using Bayesian Information Criterion 713 (BIC) analysis (e.g., *Main et al.*, 1999). The physical meaning of the porosity changepoint 714 is thought to represent a change in microstructure. Low-porosity volcanic rocks often 715 contain a poorly connected or tortuous network of pores, and fluids are often obliged to 716 travel through narrow microcracks that connect the pore network (*Heap et al.*, 2014; 717 Farguharson et al., 2015; Kushnir et al., 2016; Heap and Kennedy, 2016). Moderate- to 718 high-porosity volcanic rocks, by contrast, often contain a well-connected network of 719 large pores and channels (Rust and Cashman, 2004; Wright et al., 2006; Farquharson et 720 al., 2015; Kennedy et al., 2016; Kushnir et al., 2016; Heap and Kennedy, 2016). The 721 porosity changepoint in these studies lies within a narrow range of porosity: between 722 0.15 and 0.2 (Farguharson et al., 2015; Heap et al., 2015b; Kushnir et al., 2016; Heap and 723 Kennedy, 2016). Applying the same BIC analysis to the lava and lava breccia data of this 724 study confirms that they are statistically better described by two power laws that 725 intersect at a changepoint porosity of ~ 0.14 (Figure 15), rather than a single power law. 726 The determined changepoint is similar to those found for extrusive andesites and 727 basaltic-andesites (Farguharson et al., 2015; Kushnir et al., 2016; Heap and Kennedy, 728 2016) and viscously densifying block-and-ash flow deposits (Heap et al., 2015b). The 729 physical meaning of the porosity changepoint in the lavas and lava breccias from 730 Whakaari appears consistent with that described by these previous studies. Lava sample 731 WI20—which contains a porosity of ~ 0.06 (i.e., below the changepoint)—contains few 732 pores that are connected by a tortuous network of microcracks (Figure 4a). Indeed, 733 mercury injection analysis shows that the majority of the pore throats (95%) are 734 between 0.2 and 0.004 mm in diameter (i.e., microcracks connect the porosity; Figure 735 13) and the application of modest confining pressures significantly reduced the 736 permeability, interpreted as a consequence of the closure of compliant microcracks 737 (Figure 12; see also Vinciguerra et al., 2005 and Nara et al., 2011). By contrast, sample 738 WI-F-96 contains a very high porosity ~ 0.65 (i.e., above the changepoint) and contains a

network of presumably well-connected channels that are visible with the naked eye(Figures A1 and A6).

741 We find that, despite the hydrothermal alteration of the lavas (Figures 4a, 4l, and 742 A6), their values of porosity and permeability are not dissimilar to those for unaltered lavas (e.g., Farquharson et al., 2015; Kushnir et al., 2016; Heap and Kennedy, 2016). 743 744 However, we stress that alteration must greatly modify the porosity structure of a 745 material to greatly modify porosity and permeability. This is typically not the case for 746 the studied altered lava samples (Figures 4a, 4l, and A6). For example, we find that pore-747 and fracture-filling precipitation is rare in the lavas collected (Figures A6-A7): a modest 748 volume of cristobalite (14 wt.%) and minor jarosite and gypsum precipitation is present 749 in sample WI20 (although the presence of pore-filling cristobalite may not decrease 750 permeability if associated with microporous diktytaxitic textures; *Kushnir et al.*, 2016) 751 and kaolinite (10 wt.%) is present in sample WI30 (although it is not clear whether such 752 clays are associated with mineral replacement or pore- or crack-filling precipitation). 753 Indeed, lava sample WI20 can be classified only as *moderately* altered, since less than 754 half of the original mineral phases have been altered or replaced (BS5930, 1999). We do 755 note however that hydrothermal alteration of the lava likely contributed to the growth 756 of the macrocracks that are commonly found within the lavas at Whakaari (e.g., Figure 757 4a); hydrothermal alteration has been previously shown to reduce material strength 758 (Pola et al., 2012; Frolova et al., 2014; Wyering et al., 2014). We show here that such 759 macrofractures can increase sample permeability by a three orders of magnitude (Table 760 A4; Figure 15), in accordance with previous studies on the influence of macrofractures 761 in volcanic rock (Nara et al., 2011; Heap and Kennedy, 2016). An example of such a 762 fracture in the lavas at Whakaari is provided in Figure 16a. A second noteworthy 763 observation is that macrocracks within the lavas can be efficiently sealed with 764 hydrothermal precipitates (see the example in Figure 16b). Andesite blocks ejected from 765 the 1978 crater also contained 5-10 mm wide veins of alunite, anhydrite, and Al-rich chlorite (*Hedenquist et al.*, 1993). Progressive precipitation within fractures will greatly
reduce their permeability (*Edmonds et al.*, 2003; *Griffiths et al.*, 2016).

The high permeability of the fractures within low-permeability lava ($\sim 10^{-13}$ to $\sim 10^{-12}$ m²) coupled with the presence of fracture-filling precipitation suggests that such fractures are preferentially used as pathways for hydrothermal fluids. Therefore, high permeability fractured (altered) lavas could be modified to low-permeability lavas containing sealed fractures over time, providing that the fluid temperature and composition (including pH) support mineral precipitation.

774

775 7.2.2 Porosity and permeability relationships in the tuffs and crusts

776 The porosity-permeability relationship for the ash tuffs is considerably more 777 scattered than that for the lavas and lava breccias, although there is a general trend of 778 decreasing permeability with decreasing porosity (Figure 15). We highlight that the 779 observation of a trend in these data is only made possible by the large number of 780 datapoints (n = \sim 100; Tables A5-A6), a prerequisite for understanding relationships in 781 rocks with variable microstructures due to variable alteration styles and intensities. We 782 observe no porosity changepoint in the ash tuff data. The absence of two distinct 783 porosity/permeability trends is likely due to the absence of two distinct microstructural 784 groups in the ash tuff samples (i.e., microcracks and few pores versus a well-connected 785 pore network). If microcracks were present in all of the ash tuff samples, it would serve 786 to increase the permeability of the low-permeability samples, but would not 787 significantly change the permeability of the high-permeability samples (see also *Heap* 788 and Kennedy, 2016), thus potentially creating a changepoint. Microcracks were not 789 observed during our SEM analysis of the ash tuff samples (Figures 4b-f) and mercury 790 injection analysis showed that pore throats can be as large as $\sim 100 \ \mu m$ (i.e., pores; 791 Figure 13). The modest decrease in permeability as confining pressure is increased is 792 further evidence of the absence of microcracks in the ash tuff samples (Figure 12).

793 However, we highlight that a changepoint can exist for granular, microcrack-free 794 materials (Bourbié and Zinszner, 1985; Heap et al., 2015b). It is possible therefore that a 795 microstructural changepoint could exist for the ash tuffs of this study at a lower porosity 796 (~0.1-0.15), in accordance with those found for sandstone (*Bourbié and Zinszner*, 1985) 797 and welded pyroclastic deposits (*Heap et al.*, 2015b). This assertion is supported by the 798 very low porosity and permeability of the highly cemented sulphur flow samples (WI29; 799 Table A6) which, if plotted on Figure 15, would presumably not follow the single power 800 law trend defined by the measured ash tuff samples. However, based on our data 801 (Figure 15), low-porosity and low-permeability ash tuffs at Whakaari are likely rare and 802 potential restricted to vent-proximal deposits cemented by sulphur.

803 Despite the trend of decreasing permeability with decreasing porosity, the 804 permeability of the ash tuff can vary by up to five orders of magnitude for a single value 805 of porosity (Figure 15). This is due to the considerable microstructural variability 806 between the samples (particle size, pore size, amongst others; Figures A8-11). If we first 807 consider alteration style, we find that white-coloured ash tuff samples (their white 808 colour is indicative of alunite, a sulphate that is found as a replacement mineral and as a 809 pore-filling precipitate; Figure 5; *Heap et al.*, 2015a; *Mayer et al.*, 2015) are typically of 810 lower porosity and permeability (white circles in Figure 15) than grey-, brown-, and 811 red/purple-coloured ash tuff samples (samples that do not contain alunite; grey circles 812 in Figure 15). It is possible that porosity and permeability were reduced in these 813 samples due to the precipitation of pore-filling alunite, a consequence of their exposure 814 to acid-sulphate fluids. Alunite precipitates from low-pH solutions (Brown, 1978; Ece et 815 al., 2008), typically between 2.5 and 3.0 (Brown, 1978), and requires the formation of 816 sulphuric acid either by (1) atmospheric oxidation of iron sulphides (supergene 817 environment), (2) atmospheric oxidation near the water table of H_2S from deeper 818 boiling fluids (steam-heated hydrothermal environment) and (3) disportionation of SO₂ 819 to H₂SO₄ and H₂S from a condensing magmatic vapour plume (magmatic hydrothermal

820 environment) (Rye et al., 2002; Mutlu et al., 2005; Zimbelman et al., 2005; Ece et al., 821 2008; *Pirajno*, 2009). Therefore, progressive alunite precipitation could reduce porosity 822 and permeability in accordance with the porosity-permeability trend shown in Figure 823 15, although we note that tuffs without alunite alteration can also be of low porosity and 824 permeability. While the observation that the density of rocks within the Biga Peninsula 825 (Turkey) increases with alunite content (*Ece et al.*, 2008) supports such a hypothesis, 826 firm conclusions cannot be drawn without undertaking laboratory-controlled 827 precipitation experiments.

828 We further note that some alunite-bearing samples contain very small pores (i.e., 829 pores that cannot be seen with the naked eye; WI-F-3, WI-F-52, WI-F-110, and WI-F-830 112; Figures A8-11) and that these samples all have very low permeabilities of $\sim 10^{-17}$ 831 m^2 (Table A5). It is likely that the pore radii in these samples have been dramatically 832 reduced by pervasive alunite precipitation, explaining their low permeability (although 833 a high density of very small pores allows the rock to maintain a high porosity). Such low-834 permeability layers can also exist as thin (\sim 2-4 mm) layers (as is the case for WI-F-4; 835 Figure A8). The thin layer in samples of WI-F-4, orientated perpendicular to the 836 measurement of permeability, dramatically reduced sample permeability: the 837 permeability of these samples is also $\sim 10^{-17}$ m² (Table A5).

838 Therefore, shallow tuffs that exist within pathways for hydrothermal fluids may 839 be of lower porosity and permeability than those shielded from acid-sulphate fluids, 840 providing that the fluid temperature and composition (including pH) support mineral 841 precipitation. Near active vents and fumaroles, where the pH may preclude alunite 842 precipitation (pH can be close to unity; Giggenbach et al., 1993), the cementation of 843 tephra deposits and tuffs by sulphur can effectively destroy porosity and permeability 844 (as is the case for the sulphur flow sample WI29; Table A6; see also Harris and 845 Maciejewski, 2000; Christenson et al., 2016).

846 Macroscopic textures add another degree of complexity. We observe gas 847 elutriation pipes (e.g., sample WI21; Figures A8-11) and bedding (e.g., samples WI22 848 and WI23; Figures A8-11) in some of the samples collected. Although the influence of 849 bedding-perpendicular gas elutriation pipes on permeability is unclear with the 850 available data (Figure 11), we anticipate that the observed bedding will promote a 851 permeability anisotropy within the ash tuff deposits (not tested systematically here due 852 to the limited size of the blocks collected), especially if adjacent interbedded layers or 853 laminations have disparate values of porosity and permeability (e.g., WI-F-4; Figure A8). 854 Bedding-induced permeability anisotropy will favour the lateral movement of fluids 855 over the vertical movement of fluids.

856 The porosity and permeability of the crust (jarosite and sulphur) samples 857 (Figure A12) is distinct from the ash tuffs (Figure 15; Tables A5-A6). The jarosite crust 858 samples in particular are much more permeable (about 5 orders of magnitude) than ash 859 tuffs of a similar porosity. In terms of porosity-permeability, the crust samples are 860 similar to porous sandstones. For example, the permeability of Berea sandstone 861 (porosity = 0.21) is about 5.0 \times 10⁻¹² m² (*Zhu and Wong, 1997*). The higher permeability 862 of the crust samples and the ash tuff samples can be explained by their larger pore size 863 compared to the ash tuffs (Figure 4). The presence of larger pores is likely due to the 864 fact that these surficial deposits are yet to undergo compaction as a result of burial by 865 more recent tephra deposits.

866

867 7.3 Porosity and permeability relationships in the unlithified materials

The outgassing of magmatic volatiles and the movement of hydrothermal fluids through and within the shallow crater floor must rely on the permeability of the surficial, unlithified ash/lapilli deposits. When dry, the unlithified crater-fill deposits have a high permeability of $\sim 10^{-12}$ m² (measured in the laboratory). Under *in-situ* (i.e., partially-saturated) conditions, we measured permeabilities between $\sim 10^{-15}$ and $\sim 10^{-12}$ 873 m^2 (Figure 9; Table A3) for the unlithified materials of the crater floor, values not 874 dissimilar to the ash tuffs (Figure 15). Samples at or very close to complete saturation (> 875 1 m depth) had permeabilities too low to be measured by the PL-300 soil permeameter 876 (Table A3). We find that permeability decreases with the available air-filled porosity and 877 with depth (Figure 9). It is important to note that the *in-situ* values of field permeability 878 for the unlithified deposits are relevant for gases (e.g., CO_2) moving through deposits 879 partially saturated with aqueous fluids; these values likely therefore considerably 880 underestimate the permeability of these deposits to aqueous solutions.

881 To understand the role of texture on the permeability of the unlithified deposits, 882 we plot those deposits containing large lapilli fragments as squares whilst deposits 883 without large lapilli fragments are plotted as circles (Figure 17). We find that there is no 884 correlation between the presence/absence of large lapilli and porosity-permeability 885 (Figure 17). We also use these data to assess the impact of alteration on the porosity and 886 permeability values of these unlithified deposits. The red-coloured deposits-887 interpreted here as high alteration layers—are plotted as red symbols, the highly 888 altered deposit at SP07 (Figure 6) is plotted as a yellow symbol, and the largely 889 unaltered deposits of trench SP05 and SP08 (Figure 6) are shown as grey symbols in 890 Figure 17. In highlighting these data we see that there is no obvious correlation between 891 alteration and porosity and permeability (Figure 17). It follows that, since we see no 892 evidence of compaction (Table A3), permeability to gas in these deposits simply 893 decreases with depth due to the increase in water saturation with depth (Figures 8 and 894 9). To modify permeability of a material, the alteration must modify the structure of the 895 porosity (e.g., through dissolution, precipitation, alteration-induced cracking). However, 896 if the deposit remains unlithified, the alteration must have simply resulted in 897 devitrification, oxidation, or mineral replacement of the ash and lapilli particles; there 898 was therefore no change to the interstitial void space, and therefore no change to the 899 porosity or permeability. However, permeability may be reduced if the alteration results

900 in the formation of clay minerals. Therefore, despite the absence of a correlation
901 between alteration and porosity/permeability in our data (Figure 17), we do not
902 preclude here alteration-induced changes to porosity/permeability in the unlithified
903 deposits at Whakaari.

904 Values of permeability between $\sim 10^{.15}$ m² and $\sim 10^{.12}$ m² were measured for the 905 unlithified crater-fill deposits (Figure 9; Table A3). Therefore, the circulation and 906 passage of fluids within and through the deposits of the crater floor should be largely 907 unimpeded. However, gases may struggle to quickly negotiate through deeper deposits 908 that are close to complete water saturation. Indeed, samples at or very close to complete 909 saturation (> 1 m depth) had permeabilities too low to be measured by the PL-300 soil 910 permeameter (Table A3). The movement of fluid (both gases and aqueous solutions) 911 will be strongly inhibited by the layers of partially saturated fine ash however (such as 912 that near Donald Duck and Noisy Nellie craters; Figures 3h-i), which has an in-situ 913 permeability of $\sim 10^{-19}$ m² (measured in the laboratory). Blankets of fine ash within the 914 unlithified crater floor deposits will create a permeability anisotropy and are likely to 915 strongly inhibit the vertical movement of hydrothermal fluids.

916

917 8. Conclusions and implications for Whakaari (White Island volcano)

918 Understanding the eruptive behaviour of, and modelling unrest at, Whakaari 919 rests on a detailed comprehension of the permeability of the materials that form and 920 exist within the crater. We find that the assembled products at Whakaari vary 921 considerably in terms of porosity and permeability: porosity ranges from ~ 0.01 up to 922 ~0.7 and permeability spans eight orders of magnitude (from $\sim 10^{-19}$ to $\sim 10^{-11}$ m²). This 923 variability is due to the variable rock types forming the flanks of the volcano and filling 924 the craters (tuffs, lavas, lava breccias, and unlithified tephra), their varied 925 microstructures, and their varied hydrothermal alteration. As a result, the spatial 926 distribution of the assembled volcanic materials is of paramount importance in
927 deciphering fluid flow in active volcanic hydrothermal systems. To this end, we have
928 constructed a cartoon cross section in which we capture the salient features of our
929 study, presented as Figure 18.

930 The chief lithologies at Whakaari are tuffs, lavas and lava breccias, and crater-931 filling unlithified tephra (Figures 2, 3, and 18). The crater-filling tephra has a relatively 932 high permeability ($\sim 10^{-15}$ to $\sim 10^{-12}$ m²), and is therefore unlikely to impede the 933 movement of fluids within the shallow crater (Figure 18, item 1), as evidenced by crater 934 floor gas flux measurements (*Bloomberg et al.*, 2014). Permeability in these unlithified 935 deposits may also be enhanced by vertical gas elutriation pipes (Figure 4c; Figure 18, 936 item 2). However, we highlight that bedding-induced permeability anisotropy (Figures 937 A8-11; Figure 18, item 3) and well-sorted layers of fine ash with a low permeability 938 (WI28 $\sim 10^{-19}$ m²; Figure 3h and 3i; Figure 18, item 4) may impede and prevent vertical 939 fluid movement, respectively. We also note that permeability to gases will likely 940 decrease with depth (in the first few meters) in the unlithified crater-fill due to the 941 increase in water content, and therefore decrease in air-filled porosity, as depth 942 increases (Figures 8 and 9). While mineralogical transformations are unlikely to 943 influence porosity and permeability (Figure 17), cementation or clay formation will 944 likely result in reductions in tephra permeability. In particular, cementation of near-vent 945 tephra with sulphur (Figure 3e and 3f), where the pH is too high for alunite precipitation 946 (Giggenbach et al., 2003), results in the destruction of porosity and permeability, both 947 horizontally as subcrater layers (Christenson et al., 2016) and vertically as fumaroles 948 (sulphur flow sample WI29 contains a porosity of ~0.01 and a permeability < 10^{-18} m²; 949 Figure 18, item 5). Near-vent cementation of tephra and tuffs with sulphur could 950 therefore lead to the clogging of active vents or time variable prevention of lateral 951 and/or vertical fluid movement (see also Harris and Maciejewski, 2000; Mayer et al., 952 2016b; Christenson et al., 2016).

953 The tuffs—formed by the consolidation and cementation of tephra deposits— 954 are typically porous (porosity = 0.3-0.7), although their permeability can range from 955 $\sim 10^{-17}$ to $\sim 10^{-12}$ m² (Figure 15). Tuffs are found forming the crater wall (although the 956 dominant lithology is lava/lava breccia; Figures 2, 3, and 18) and presumably comprises 957 the deep main crater fill (Figure 18), as can be seen in ballistics from recent eruptions. 958 As for the crater-fill tephra, bedding-induced permeability anisotropy and well-sorted 959 layers of fine ash tuff with a very low permeability may impede or restrict the vertical 960 movement of fluids (Figure 18, item 6) and vertical gas elutriation pipes may assist 961 vertical fluid movement (Figure 18, item 4). A notable observation is that the white-962 coloured ash tuffs (an alteration colour associated with alunite) contain the lowest 963 porosities and have the lowest permeabilities (Figure 15). Therefore, hydrothermal 964 alteration of the ash tuffs in shallow zones (alunite is formed through the oxidation of 965 H₂S from deeper boiling fluids) that host or have hosted acid-sulphate hydrothermal 966 fluids can result in reductions in porosity and permeability through the precipitation of 967 pore-filling alunite (Figure 18, item 7), providing that the fluid temperature and 968 composition (including pH) support mineral precipitation. Hydrothermal alteration 969 deeper in the crater could also result in modifications to permeability (e.g., dissolution, 970 clay formation, amongst others; Figure 18, item 8).

971 The crater walls chiefly comprise lavas and lava breccias (Figures 2 and 3). 972 Discontinuities at the crater margin serve as a passageway for hydrothermal fluids and 973 feed the numerous active fumaroles, boiling mud pools, and springs for acid streams 974 (Figures 2, 3, and 16a; Figure 18, item 9) (Bloomberg et al., 2014). The porosity of the 975 lavas and lava breccias at Whakaari ranges from ~ 0.05 up to ~ 0.7 , and permeability 976 ranges from $\sim 10^{-16}$ to $\sim 10^{-12}$ m² (Figure 15). Notably, we find that (1) the lava breccias 977 are significantly more permeable than the coherent lavas (Figure 15) and, (2) 978 macroscopic fractures within the lava samples (Figure 16a) increase sample 979 permeability by up to three orders of magnitude (Figure 15; Table A4). As a result, fluids

980 travelling through the crater walls (hydrothermal fluids and seawater; Figure 18) will 981 likely negotiate through a combination of fractures within the lava (Figure 18, item 10) 982 and the permeable lava breccia (Figure 18, item 11). The ingress of seawater into the 983 hydrothermal system will result in the mixing of hydrothermal fluids and seawater 984 (Figure 18, item 12). While the passage of hydrothermal fluids can weaken the lava and 985 promote the growth and widening of fractures through alteration (e.g., mineral 986 replacement and/or dissolution; Figure 16a), hydrothermal fluids can also seal fractures 987 through precipitation (Figure 16b; *Hedenquist et al.*, 1993). In zones where precipitation 988 dominates over dissolution (a function of the fluid temperature and composition, 989 including pH), the sealing of fractures will significantly reduce permeability (Griffiths et 990 al., 2016; Figure 18, item 13), transforming a zone of high-permeability into a zone of 991 low-permeability. The mixing of hydrothermal fluids and seawater can also result in 992 mineral precipitation (such as anhydrite) and reductions to permeability (Kawada and 993 *Yoshida*, 2010). Fracture pathways within the crater wall may also become clogged with 994 sulphur precipitation (Figure 18, item 14). Finally, our data have shown that an increase 995 in effective pressure (i.e., depth) will significantly reduce the permeability of the 996 microfractured lava (Figure 12). The permeability of macrofractures will also be 997 reduced at depth (*Nara et al.*, 2011). As a result, fluids may find it increasingly difficult 998 to find an escape route into the crater wall as depth is increased (Figure 18, item 15).

999 As outlined above, hydrothermal alteration typical of a volcanic hydrothermal 1000 system can result in increases (due to alteration-induced weakening and fracturing) and 1001 decreases (due to hydrothermal precipitation) to permeability. Importantly, a decrease 1002 in permeability, be it due to fracture sealing in the lava, pore-filling alunite precipitation, 1003 cementation by sulphur, and/or very low permeability layers (Figure 18), can result in 1004 pore pressure augmentation (Christenson et al., 2010; Heap and Wadsworth, 2016). An 1005 increase in pore pressure could jeopardise the stability of the volcanic slopes (*Day*, 1006 1996; Voight and Elsworth, 1997; Reid et al., 2001; Reid, 2004; Moon et al., 2009), result in seismicity (*Nishi et al.*, 1996; *Sherburn et al.*, 1998; *Chardot et al.*, 2015), and/or drive
the wide variety of eruptions observed at Whakaari (e.g., *Houghton and Nairn*, 1991; *Mayer et al.*, 2015). Indeed, an increase in pore pressure due to the hydrothermal
system was thought responsible for the unrest between 2002-2006 and 2007-2009
(*Fournier and Chardot*, 2012), although we note that lake level variations are likely to
have significantly modified the pore fluid pressure within the system during these
intervals (*Christenson et al.*, 2016).

1014 The reduction in permeability may also encourage changes to the preferred 1015 pathways for hydrothermal fluid circulation. This is exemplified by the numerous past 1016 and present vents, fumaroles, and craters that pepper the main crater floor and the base 1017 of the slopes of the volcanic amphitheatre at Whakaari (Figures 2 and 3) and highlights 1018 the constantly evolving and unpredictable nature of hydrothermal volcanic systems (see 1019 also Harris and Maciejewski, 2000). Changes to hydrothermal circulation will also 1020 expose new tephra and tuffs to alunite, clay, and sulphur alteration/precipitation. The 1021 exposure of new materials to porosity and permeability reducing alteration could 1022 therefore result in a reduction of the equivalent permeability of the system over time, 1023 thereby increasing the potential for pore pressure augmentation and the associated 1024 hazardous consequences.

1025 The primary goal of this contribution was to produce a very large dataset to 1026 inform future modelling efforts. The permeability of the materials within active volcanic 1027 hydrothermal systems is, for example, required to understand and accurately model the 1028 outgassing of magmatic volatiles from the magma-filled conduit (Collombet, 2009; 1029 *Collinson and Neuberg*, 2012), subsurface hydrothermal activity and therefore volcanic 1030 unrest (Hurwitz et al., 2007; Peltier et al., 2009; Todesco et al., 2010; Christenson et al., 1031 2010 Fournier and Chardot, 2012), gas monitoring (Bloomberg et al., 2014; Peiffer et al., 1032 2014), and volcano seismicity (Leet, 1988; Nishi et al., 1996; Sherburn et al., 1998; Bean 1033 et al., 2014; Chardot et al., 2015). For example, Fournier and Chardot (2012) show, using

1034 a thermo-poro-elastic model, that increases in pore pressure at depth were likely 1035 responsible for recent (2002-2006 and 2007-2009) episodes of ground deformation at 1036 Whakaari. The model of Fournier and Chardot (2012) assumes an isotropic value 1037 permeability of 10⁻¹⁵ m² and, while these authors concede that the assumption of a 1038 permeability isotropy is an oversimplification, our study—which shows that 1039 permeability spans eight orders of magnitude (from $\sim 10^{-19}$ to $\sim 10^{-11}$ m²) and is 1040 complicated by numerous spatial and temporal considerations (summarised in Figure 1041 18)—highlights the challenge presented for the construction of more complex models. 1042 Further, we stress that the construction of such models will require an improved 1043 understanding of the subsurface stratigraphy, hydrogeology, and geochemical 1044 composition of the hydrothermal fluids at Whakaari.

1045Our study provides the most comprehensive dataset for the porosity and1046permeability of the materials that comprise a volcanic hydrothermal system to date.1047Although our study highlights an extreme variability in these parameters, we anticipate1048that these data will allow for a better understanding of the behaviour and evolution of1049volcanic hydrothermal systems worldwide.

1050

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1078

1079 Author contributions

1080 M.J. Heap led the project and wrote the manuscript. Field work and sample 1081 collection was carried out by M.J. Heap, B.M. Kennedy, J.I. Farquharson, K. Mayer, M. 1082 Letham-Brake, B. Scheu, Y. Lavallée, and P. Siratovich. Laboratory measurements of 1083 permeability were collected and analysed by J.I. Farquharson, M.J. Heap, and T. Reuschlé. 1084 The permeability at different confining pressures was performed and analysed by T. 1085 Reuschlé. J. Ashworth and J.I. Farquharson collected and analysed the field data for the 1086 lithified rocks. K. Mayer and B. Scheu collected the field data for the unlithified deposits, 1087 which was analysed by K. Mayer. H.A. Gilg performed and analysed all of the XRPD data.

- 1088 M.J. Heap and B.M. Kennedy conducted the SEM work. J.I Farquharson performed the
- 1089 Bayesian Information Criterion (BIC) analysis. The cross section was constructed by
- 1090 B.M. Kennedy, with help from M.J. Heap. All authors had a hand in the interpretation of
- 1091 the data and the writing of the manuscript.
- 1092

1093 **Table 1**. X-ray powder diffraction (XRPD) analysis showing the quantitative bulk

1094 mineralogical composition of the nine lithified blocks and the two unlithified samples

- 1095 chosen for additional analyses. Values are in wt.%.
- 1096

Sample	WI20	WI21	WI22	WI23	WI24	WI25	WI26	WI27	WI28	WI29	WI30
Rock type	Lava	Ash tuff	Ash tuff	Ash tuff	Ash tuff	Jarosite crust	Sulphu r crust	Ash lapilli	Ash	Sulphur flow	Lava breccia
(K, Na)- Alunite	-	32 ± 3	6 ± 3	25 ± 3	1 ± 1	-	-	8 ± 2	-	1 ± 0	-
Jarosite	3 ± 1	-	-	4 ± 1	4 ± 1	25 ± 5	-	-	-	-	1 ± 0
Anhydrite	-	-	-	-	-	-	-	-	-	-	1 ± 0
α-Sulphur	-	-	-	-	-	-	46 ± 5	-	-	99 ± 0	-
Gypsum	4 ± 1	1 ± 1	1 ± 1	-	-	-	-	2 ± 1	3 ± 1	-	-
Amorphous phases (volcanic glass, opal- A)	14 ± 5	66 ± 6	90 ± 3	68 ± 4	92 ± 2	37 ± 5	44 ± 5	59 ± 5	52 ± 5	-	72 ± 4
Kaolinite	-	-	-	2 ± 2	-	-	-	-	-	-	10 ± 2
Cristobalite	17 ± 4	1 ± 1	3 ± 1	1±1	2 ± 1	4 ± 1	1±1	7 ± 1	7 ± 1	-	3 ± 2
Quartz	-	< 1	< 1	< 1	1 ± 1	1±1	-	-	-	-	1 ± 0
Pyroxene	10 ± 2	-	-	-	-	22 ± 4	4 ± 3	9 ± 2	20 ± 4	-	3 ± 1
Plagioclase	37 ± 3	-	-	-	-	10 ± 3	5 ± 3	15 ± 2	18 ± 3	-	8 ± 3
Hematite	1 ± 1	-	-	-	1 ± 1	-	-	-	-	-	1 ± 0
K-Feldspar	14 ± 2	-	-	-	-	-	-	-	-	-	-



Figure 1. Whakaari (White Island volcano). (a) Map of Whakaari showing the locations of the collection sites for the lithified materials. The collection areas for the hand-sized sample blocks are indicated by the red circles. The collection sites for the nine main blocks of this study (and the two unlithified materials WI27 and WI28) are indicated by the black stars. The inset shows a map of New Zealand showing the location of the Taupo Volcanic Zone (pink area) and Whakaari volcano (red triangle). (b) Aerial photograph of Whakaari taken looking east-southeast.



1111

Figure 2. Reconnaissance by land, sea, and air. Map of Whakaari (centre) surrounded by

1113 photographs of the volcanic island. The position from which each photograph was taken,

- 1114 and the direction of view, is indicated by the numbers and arrows. Prominent rock types
- 1115 are labelled on the photographs.
- 1116



1117

Figure 3. Photographs showing the sites at which the nine lithified blocks of this study

- 1119 (and the two unlithified materials) were collected. The location of the collection sites are
- indicated on the map in Figure 1.
- 1121
- 1122



1123

Figure 4. Photographs and scanning electron microscope (SEM) images of the nine main
blocks of this study (and the two unlithified materials). (a) Macrocracked and uncracked
lava WI20. (b-f) Ash tuff WI21, WI22, WI23, and WI24. (g) Jarosite crust WI25. (h)
Sulphur crust WI26. (i) Unlithified ash/lapilli WI27. (j) Unlithified ash WI28. (k) Sulphur
flow WI29. (l) Lava breccia WI30. Collection sites for each material are indicated in
Figure 1 and photographs of the collection sites are provided in Figure 3.



- 1130
- **Figure 5**. Optical microscope image showing pore-filling alunite precipitates in a sample
- 1132 of ash tuff (WI21). Image taken from *Heap et al.* (2015a).
- 1133
- 1134



Figure 6. Locality and photographs of each of the eight trenches dug for unlithified
material analysis (permeability, porosity, water content, and temperature). Numbers on
the pictures indicate the sample number (Table A3). Inset shows two examples taken
from trench SP01.



Figure 7. Field measurements on the lithified hand-sized sample blocks collected. (a)
Field permeability (using the TinyPerm II) as a function of field dry bulk density. (b)
Field permeability (using the TinyPerm II) as a function of total porosity. Data are
available in Tables A1 and A2.



Figure 8. Field measurements on the unlithified materials from the eight trenches
shown in Figure 6. (a) Depth as a function of total porosity. (b) Depth as a function of
water content. (c) Depth as a function of air-filled porosity. Inset shows a graph of depth
as a function of deposit temperature. Data are available in Table A3.



Figure 9. Field measurements on the unlithified materials from the eight trenches
shown in Figure 6. (a) Field permeability as a function of air-filled porosity. (b) Depth as
a function of field permeability. Data are available in Table A3.





Figure 10. Laboratory measurements of permeability as a function of connected porosity for all of the 20 mm-diameter cylindrical samples of this study, plotted on loglinear axes. Data are available in Tables A4, A5, and A6. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)





Figure 11. The influence of gas elutriation pipes on the permeability of ash tuff (WI21).

1172 Graph is a plot of permeability (measured in the laboratory; data available in Table A5)

- 1173 as a function of connected porosity.
- 1174



1177 Figure 12. The influence of confining pressure (depth) on the permeability of lava

(WI20) and ash tuff (WI21). Graph is a plot of permeability (measured in the laboratory;

1179 Table 4) as a function of confining pressure. Data are available in Table A7.

1180





Figure 13. The distribution of pore throat diameters for WI20, WI21, WI22, WI23, and

- 1184 WI23 determined through mercury injection tests.
- 1185
- 1186



Figure 14. Laboratory versus field measurements of porosity and permeability. (a) Field
total porosity as a function of laboratory total porosity. (b) The ratio of field to
laboratory permeability as a function of laboratory total porosity.



Figure 15. Laboratory measurements of permeability as a function of connected porosity for all of the 20 mm-diameter cylindrical samples of this study, plotted on loglog axes. The dashed lines indicate the two best-fit power law curves through the data, as determined using modified Bayesian Information Criterion (BIC) analysis (see text for details). Data are available in Tables A4, A5, and A6. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

1201



- 1203
- 1204 Figure 16. Macroscopic fractures in lavas at Whakaari. (a) Open fractures near the
- 1205 crater rim act as pathways for the subvertical flow of hydrothermal fluids. (b) Examples
- 1206 of sealed fractures that significantly reduce the permeability of the lava.
- 1207
- 1208



Figure 17. Field measurements on the unlithified materials from the eight trenches
shown in Figure 6. (a) Field permeability as a function of air-filled porosity. (b) Depth as
a function of field permeability. Graphs have been modified from Figure 9 to show
textural features (lapilli or no lapilli) and alteration intensity. Data are available in Table
A3.



- **Figure 18**. Cartoon cross section of Whakaari from west-northwest to east-southeast
- 1219 (not to scale) that captures the salient features of our study. See text for details.

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- 1222
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1512	Appendix
1513	The goal of our appendix is twofold. First, in the interests of transparency and
1514	future reinterpretation, we are keen to show photographs of each of the hand-sized
1515	lithified blocks collected (Figures A1-A5) and each of the 20 mm-diameter laboratory
1516	samples (Figures A6-A12). Second, we are keen to future studies to use our data. To this
1517	end, we offer all of the field and laboratory data in seven tables (Tables A1-A7).
1518	
1519	Appendix figure captions
1520	
1521	Figure A1. Photographs of the hand-sized sample lava and lava breccia blocks. Each
1522	square on the scale bar is 10 mm.
1523	
1524	Figure A2. Photographs of the hand-sized sample lithified ash tuff blocks. Each square
1525	on the scale bar is 10 mm.
1526	
1527	Figure A3. Photographs of the hand-sized sample lithified ash tuff blocks (continued).
1528	Each square on the scale bar is 10 mm.
1529	
1530	Figure A4. Photographs of the hand-sized sample lithified ash tuff blocks (continued).
1531	Each square on the scale bar is 10 mm.
1532	
1533	Figure A5. Photographs of the hand-sized sample lithified ash tuff blocks (continued).
1534	Each square on the scale bar is 10 mm.
1535	
1536	Figure A6. Photographs of the 20 mm-diameter lava and lava breccia laboratory
1537	samples. Missing samples: WI20_1 and WI20_B.

1538	Figure A7. Photographs of the 20 mm-diameter lava and lava breccia laboratory
1539	samples (continued).
1540	
1541	Figure A8. Photographs of the 20 mm-diameter lithified ash tuff laboratory samples.
1542	
1543	Figure A9. Photographs of the 20 mm-diameter lithified ash tuff laboratory samples
1544	(continued).
1545	
1546	Figure A10. Photographs of the 20 mm-diameter lithified ash tuff laboratory samples
1547	(continued).
1548	
1549	Figure A11. Photographs of the 20 mm-diameter lithified ash tuff laboratory samples
1550	(continued).
1551	
1552	Figure A12. Photographs of the 20 mm-diameter surficial laboratory samples (jarosite
1553	and sulphur crust, and sulphur flow). Missing samples: WI25_A, WI25_G, WI26_B, and
1554	WI26_D.
1555	

- **Table A1**. Summary of the field data for the lava and lava breccia blocks. Photographs of
- 1557 the blocks are provided in Figures A1-A2.

Sample	Bulk density	Skeletal	Total	TinyPerm	Notes
	(kg/m ³)	density	porosity	permeability	
		(kg/m³)		estimate	
				(m ²)	
WI-F-90	1478	2466	0.40	4.79×10^{-14}	
WI-F-91	1656	2313	0.28	2.73×10^{-14}	
WI-F-92	1724	2393	0.28	2.90×10^{-12}	
WI-F-96	1154	2745	0.58	9.65 × 10 ⁻¹³	Vesicular
WI-F-97	1901	2391	0.20	1.36×10^{-12}	Red/brown
					surface
					discolouration
WI-F-98	1581	2363	0.33	4.24×10^{-12}	Vesicular
WI-F-99	1812	2214	0.18	6.09 × 10 ⁻¹⁵	Red/yellow
					surface
					discolouration
WI-F-100	1821	2383	0.24	1.18×10^{-12}	
WI-F-101	1756	2376	0.26	2.16×10^{-14}	
WI-F-102	1874	2354	0.20	8.44 × 10 ⁻¹³	
WI-F-103	1784	2364	0.25	8.31 × 10 ⁻¹²	
WI-F-104	1872	2434	0.23	3.23 × 10 ⁻¹³	
WI-F-105	1860	2421	0.23	1.11×10^{-14}	
WI-F-106	2178	2394	0.09	1.78×10^{-15}	
WI-F-107	1871	2344	0.20	3.65×10^{-14}	
WI-F-111	1316	2426	0.46	3.83×10^{-14}	Red/yellow
					surface
					discolouration
WI-F-114	1637	2375	0.31	1.50×10^{-13}	White/yellow
					surface
					discolouration
WI-F-116	1973	2784	0.29	4.11×10^{-12}	Red surface
					discolouration
WI-F-119	1781	2411	0.26	2.31 × 10 ⁻¹²	Lava breccia

Table A2. Summary of the field data for the lithified ash tuff blocks. Photographs of the
blocks are provided in Figures A2-A5. Bulk dry density could not be determined for
those samples that floated; in this case, total porosity was calculated assuming a bulk
dry density of 1000 kg/m³ (values indicated with an asterisk).

Sample	Bulk dry	Skeletal	Total	TinyPerm	Notes
	density	density	porosity	permeability	
	(kg/m³)	(kg/m³)		estimate	
	1051		0.00	(m ²)	5 11 1
WI-F-1	1351	2228	0.39	5.30×10^{-13}	Bedded
WI-F-2	1370	2379	0.42	5.00×10^{-14}	
WI-F-3	1033	2257	0.54	3.86 × 10 ⁻¹⁴	
WI-F-4	1354	2348	0.42	8.57 × 10 ⁻¹⁵	
WI-F-5	1086	2289	0.53	2.35×10^{-13}	
WI-F-6	1189	2395	0.50	4.23×10^{-15}	
WI-F-8	1416	2103	0.33	2.57×10^{-14}	Bedded
WI-F-9	1285	2338	0.45	2.53×10^{-14}	
WI-F-10	1016	2167	0.53	1.32×10^{-13}	
WI-F-11	1472	2443	0.40	1.79 × 10 ⁻¹³	
WI-F-12	1219	2370	0.49	7.49 × 10 ⁻¹³	
WI-F-13	1260	2213	0.43	2.10 × 10 ⁻¹⁴	
WI-F-14	1182	2240	0.47	1.83 × 10 ⁻¹³	Bedded
WI-F-15	1250	2397	0.48	2.94 × 10 ⁻¹²	
WI-F-16	Floater	2212	0.55*	2.06 × 10 ⁻¹²	
WI-F-17	1469	2358	0.38	8.08 × 10 ⁻¹³	
WI-F-18	1433	2291	0.37	1.85×10^{-14}	Bedded
WI-F-19	1356	2259	0.40	1.70 × 10 ⁻¹³	
WI-F-20	1247	2308	0.46	7.42 × 10 ⁻¹³	
WI-F-21	1490	2330	0.36	2.61 × 10 ⁻¹²	
WI-F-23	1099	2290	0.52	1.49 × 10 ⁻¹²	
WI-F-24	1372	2260	0.39	3.30 × 10 ⁻¹⁴	Bedded
WI-F-25	1052	2224	0.53	5.56 × 10 ⁻¹³	
WI-F-26	1293	2428	0.47	5.26 × 10 ⁻¹⁴	
WI-F-27	1390	2273	0.39	4.65 × 10 ⁻¹³	
WI-F-28	1222	2363	0.48	2.45 × 10 ⁻¹²	
WI-F-29	1295	2420	0.46	8.75 × 10 ⁻¹³	
WI-F-30	1291	2264	0.43	1.25 × 10 ⁻¹²	
WI-F-31	1061	2211	0.52	9.46 × 10 ⁻¹⁴	
WI-F-32	1174	2298	0.49	1.50 × 10 ⁻¹³	
WI-F-33	1313	2330	0.44	9.27 × 10 ⁻¹⁴	
WI-F-34	1250	2329	0.46	2.58 × 10 ⁻¹³	
WI-F-35	1304	2296	0.43	4.49×10^{-14}	
WI-F-36	1363	2368	0.42	7.23 × 10 ⁻¹⁴	
WI-F-37	1475	2284	0.35	1.23 × 10 ⁻¹⁵	
WI-F-38	1611	2431	0.34	3.72 × 10 ⁻¹³	
WI-F-39	1253	2310	0.46	5.02 × 10 ⁻¹⁴	
WI-F-40	1478	2359	0.37	4.78 × 10 ⁻¹³	
WI-F-41	1623	2460	0.34	2.56 × 10 ⁻¹⁴	
WI-F-42	1193	2274	0.48	1.57×10^{-14}	
WI-F-43	1146	2265	0.49	1.13×10^{-12}	
WI-F-44	1059	2206	0.52	1.30×10^{-12}	
WI-F-45	1456	2320	0.37	1.33 × 10 ⁻¹²	
WI-F-46	Floater	2323	0.57*	6.51 × 10 ⁻¹³	
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WI-F-47	1239	2244	0.45	1.23 × 10 ⁻¹³	Bedded
WI-F-48	1137	2254	0.50	4.57 × 10 ⁻¹³	
WI-F-49	1338	2179	0.39	3.17 × 10 ⁻¹⁴	Lapilli
					fragments
WI-F-50	1937	2460	0.21	1.65 × 10 ⁻¹³	
WI-F-51	1285	2293	0.44	7.37 × 10 ⁻¹⁴	
WI-F-52	1656	2424	0.32	1.10×10^{-14}	
WI-F-53	1106	2306	0.52	1.62 × 10 ⁻¹³	
WI-F-54	1510	2458	0.39	9.04 × 10 ⁻¹³	
WI-F-55	1392	2363	0.41	1.91 × 10 ⁻¹²	
WI-F-56	1201	2166	0.45	2.83 × 10 ⁻¹³	
WI-F-57	1611	2437	0.34	9.25 × 10 ⁻¹⁶	Bedded
WI-F-58	1511	2343	0.35	1.23 × 10 ⁻¹⁵	
WI-F-59	Floater	2353	0.58*	1.26 × 10 ⁻¹³	
WI-F-60	1311	2256	0.42	3.24 × 10 ⁻¹⁵	
WI-F-62	1154	2092	0.45	3.54 × 10 ⁻¹²	
WI-F-63	1337	2271	0.14	4.08 × 10 ⁻¹³	
WI-F-65	1213	2259	0.46	8.64 × 10 ⁻¹⁴	Bedded
WI-F-66	1243	2256	0.45	1.39 × 10 ⁻¹³	
WI-F-67	1222	2301	0.47	9.38 × 10 ⁻¹³	
WI-F-68	1204	2263	0.47	2.46 × 10 ⁻¹³	Bedded
WI-F-69	1360	2525	0.46	3.52 × 10 ⁻¹⁴	
WI-F-70	1360	2339	0.42	6.15 × 10 ⁻¹²	
WI-F-71	1236	2305	0.46	9.47 × 10 ⁻¹³	
WI-F-72	1252	2324	0.46	3.23 × 10 ⁻¹³	
WI-F-73	1269	2258	0.44	2.44×10^{-13}	Yellow
					surface
					discolouration
WI-F-74	1046	2216	0.53	1.15 × 10 ⁻¹³	
WI-F-75	1643	2412	0.32	3.00×10^{-14}	
WI-F-76	1180	2159	0.45	1.73×10^{-14}	
WI-F-77	Floater	2237	0.55*	7.55 × 10 ⁻¹³	Bedded
WI-F-78	1238	2272	0.46	3.72×10^{-14}	
WI-F-79	1123	2207	0.49	9.40×10^{-14}	x
WI-F-80	1610	2385	0.32	9.28×10^{-15}	Lapilli
	1107	2225	0.47	4.22 10.12	fragments
WI-F-81	118/	2235	0.47	4.23×10^{-12}	T ·11·
WI-F-82	1421	2453	0.42	1.55×10^{-12}	Lapilli
	1101	2225	0.40	1 1 2 10-12	Tragments
WI-F-83	1131	2235	0.49	1.12×10^{-12}	Dedeutere
WI-F-84	1910	2515	0.24	3.17×10^{-13}	Red surface
	1(1)	22(0	0.22	2 22 10-15	uiscolouration
WI-F-85	1612	2360	0.32	3.33×10^{-13}	
	1554	2301	0.35	1.20×10^{-13}	Lopilli
VV I-F-0/	1400	2395	0.41	2.50×10^{-11}	fragmonts
WIE 88	12/1	2729	0.51	2 77 × 10-13	Il aginents
WI E 90	1341	2730	0.31	3.77×10^{-10}	Lapilli
VV 1-F-07	1407	2374	0.41	1.55 × 10 **	fragments
WIE02	1200	2147	0.40	2.20×10^{-14}	Boddod
	1270	2147	0.40	5.50×10^{14}	Lapilli
VV 1-F-74	1311	2200	0.32	0.43 × 10 10	fragments
W/I_F_05	1525	2410	0.37	3 21 ~ 10-14	Sulphur
WI_F_108	1084	2410	0.57	5.21×10^{-1} 5.90 x 10-15	Sulphur
WI-F-100	1290	2555	0.37	2 93 x 10-14	Sulphur
WI_F_110	1488	2333	0.40	6.85 x 10-15	Red surface
VV1-1-11U	1100	27/1	0.40	0.03 × 10	incu sui late

					discolouration
WI-F-112	1549	2412	0.36	4.95 × 10 ⁻¹³	Sulphur
WI-F-113	1129	2451	0.54	5.83 × 10 ⁻¹⁴	Sulphur
WI-F-115	1190	2269	0.48	3.65 × 10 ⁻¹³	Sulphur
WI-F-117	1149	2593	0.56	8.15 × 10 ⁻¹³	Sulphur
WI-F-118	1100	2245	0.51	1.10 × 10 ⁻¹²	

Table A3. Summary of the field data for the unlithified deposits.

Trench	Sample	Depth (cm)	Temperature (°C)	Dry bulk density (kg/m³)	Skeletal density (kg/m³)	Total porosity	Water saturation	Air-filled porosity	Field permeability estimate (m ²)
SP01	01-A	10	22.4	1324	2220	0.40	0.18	0.22	4.88 × 10 ⁻¹³
SP01	01-B	10	22.4	1414	2220	0.36	0.18	0.18	2.72 × 10 ⁻¹³
SP01	02-A	30	30.0	1359	2180	0.38	0.25	0.13	4.69 × 10 ⁻¹⁵
SP01	02-B	30	30.0	1350	2180	0.38	0.25	0.13	3.94 × 10 ⁻¹⁵
SP01	03-A	50	30.5	867	2100	0.59	0.31	0.28	8.25 × 10 ⁻¹⁵
SP01	03-B	50	30.5	803	2100	0.62	0.31	0.31	2.44×10^{-14}
SP01	04-A	85	41.3	791	1950	0.59	0.51	0.08	1.69 × 10 ⁻¹⁵
SP01	04-A	85	41.3	789	1950	0.60	0.51	0.09	5.63 × 10 ⁻¹⁶
SP01	05-A	100	54.0	894	2000	0.55	0.48	0.07	1.88×10^{-15}
SP01	05-B	100	54.0	884	2000	0.56	0.48	0.08	1.31 × 10 ⁻¹⁵
SP01	06-A	120	51.7	826	1870	0.56	0.47	0.09	6.00 × 10 ⁻¹⁵
SP01	06-B	120	51.7	783	1870	0.58	0.47	0.11	9.57 × 10 ⁻¹⁵
SP02	01-A	115	27.8	528	2080	0.75	0.69	0.06	1.97×10^{-14}
SP02	01-B	115	27.8	532	2080	0.74	0.69	0.05	1.50×10^{-14}
SP02	02-A	135	32.2	746	1650	0.55	0.57	0.00	Out of range
SP02	02-B	135	32.2	734	1650	0.55	0.57	0.00	Out of range
SP03	01-A	10	20.0	1246	2130	0.41	0.15	0.26	2.06×10^{-14}
SP03	01-B	10	20.0	942	2130	0.56	0.15	0.41	6.00×10^{-14}
SP04	01-A	25	26.8	1255	1720	0.27	0.18	0.09	3.00×10^{-15}
SP04	01-B	25	26.8	1318	1720	0.23	0.18	0.05	1.88×10^{-15}
SP04	02-A	40	34.1	1080	1610	0.33	0.17	0.16	7.50×10^{-15}
SP04	02-B	40	34.1	1103	1610	0.31	0.17	0.14	1.88×10^{-15}
SP05	01-A	10	20.0	1375	2150	0.36	0.07	0.29	4.50×10^{-14}
SP05	01-B	10	20.0	1487	2150	0.31	0.07	0.24	3.62 × 10 ⁻¹⁵
SP06	01-A	60	64.0	1145	1670	0.31	0.25	0.06	Out of range
SP06	01-B	60	64.0	1109	1670	0.34	0.25	0.09	Out of range
SP06	02-A	90	70.0	827	1980	0.58	0.46	0.12	5.35×10^{-14}
SP06	02-B	90	70.0	853	1980	0.57	0.46	0.11	7.35×10^{-14}
SP07	01	-	97.8	1091	1880	0.42	0.26	0.16	3.25×10^{-14}
SP08	01	50	22.8	1142	1930	0.41	0.33	0.08	5.25×10^{-14}

Table A4. Summary of the laboratory data for the 36 lava and lava breccia samples.1571Permeability (nitrogen) was measured under a confining pressure of 1 MPa unless1572otherwise stated. Photographs of the samples are provided in Figures A6 and A7.1573Fracture permeability k_f is provided for samples WI20_D and WI20_E using the1574approach outlined in *Heap and Kennedy* (2016) (see text for details).

Sample	Dry bulk	Connected	Permeability	Notes
	density (kg/m ³)	porosity	(m ²)	
WI20_1	2525	0.05	1.58 × 10 ⁻¹⁶	Pc = 2 MPa
WI20_A	2542	0.06	7.05 × 10 ⁻¹⁷	
WI20_B	2512	0.05	-	
WI20_C	2517	0.07	< 10 ⁻¹⁸	
WI20_D	2382	0.13	1.38 × 10 ⁻¹⁴	Macrocracks
			$(k_f = 4.3 \times 10^{-13})$	parallel to flow;
				Pc = 2 MPa
WI20_E	1419	0.13	1.04×10^{-13}	Macrocracks
			$(k_f = 3.3 \times 10^{-12})$	parallel to flow;
				Pc = 2 MPa
WI30_1	1564	0.33	1.57 × 10 ⁻¹³	Breccia
WI30_2	1579	0.33	1.37 × 10 ⁻¹³	Breccia
WI30_3	1658	0.30	4.93 × 10 ⁻¹³	Breccia
WI30_4	1828	0.24	9.93 × 10 ⁻¹⁴	Breccia
WI30_5	1551	0.34	1.56 × 10 ⁻¹³	Breccia
WI30_6	1612	0.31	4.24 × 10 ⁻¹³	Breccia
WI30_7	1929	0.21	9.38 × 10 ⁻¹⁴	Breccia
WI30_8	1587	0.32	1.92 × 10 ⁻¹³	Breccia
WI-F-90_1	1467	0.45	2.66 × 10 ⁻¹³	
WI-F-90_2	1463	0.42	3.24 × 10 ⁻¹³	
WI-F-91	1575	0.35	1.65 × 10 ⁻¹²	
WI-F-92	1560	0.34	2.84 × 10 ⁻¹³	
WI-F-96_1	1000	0.63	5.77 × 10 ⁻¹³	Vesicular
WI-F-96_2	825	0.69	1.48×10^{-12}	Vesicular
WI-F-98	1409	0.41	8.37 × 10 ⁻¹³	Breccia
WI-F-99	1831	0.14	2.79 × 10 ⁻¹⁴	Pale
				discolouration
WI-F-101	1613	0.32	3.23 × 10 ⁻¹³	Breccia
WI-F-102	1808	0.22	5.55 × 10 ⁻¹³	Breccia
WI-F-103	1690	0.30	6.97 × 10 ⁻¹³	Breccia
WI-F-105_1	1894	0.18	1.11 × 10 ⁻¹⁴	
WI-F-105_2	1637	0.30	1.28 × 10 ⁻¹⁴	
WI-F-105_3	1863	0.21	2.07 × 10 ⁻¹³	
WI-F-106	2272	0.04	< 10 ⁻¹⁸	Permeability too
				low for benchtop
				measurement
WI-F-107	1728	0.29	6.69 × 10 ⁻¹³	
WI-F-111_1	1290	0.49	2.90 × 10 ⁻¹³	
WI-F-111_2	1260	0.51	3.34 × 10 ⁻¹³	
WI-F-114	2082	0.07	7.94 × 10 ⁻¹⁷	Pale
				discolouration
WI-F-116_1	1895	0.46	2.51 × 10 ⁻¹³	Orange
				discolouration

WI-F-116_2	1711	0.49	2.78 × 10 ⁻¹³	Orange
				discolouration
WI-F-116_3	2140	0.39	1.77 × 10 ⁻¹⁴	Orange
				discolouration

Table A5. Summary of the laboratory data for the 99 lithified ash tuff samples.
Permeability (nitrogen) was measured under a confining pressure of 1 MPa unless
otherwise stated. Photographs of the samples are provided in Figures A8-A11.

Sample	Dry bulk	Connected	Permeability	Notes
	density (kg/m ³)		(m²)	Alunita
WI-F-2_1	1300	0.45	2.18×10^{-16}	Alunite
WI-F-2_2	138/	0.44	1.70×10^{-16}	Alunite
WI-F-3_1	964	0.58	1.00×10^{-16}	Alunite; small
WI-E-3 2	980	0.57	6 5 2 × 10-17	Alunite: small
WI-I-5_2	500	0.37	0.55 × 10 1/	pores
WI-F-4_1	1340	0.42	1.94 × 10 ⁻¹⁵	Thin (2-3 mm)
_				low-porosity
				layer
WI-F-4_2	1306	0.45	4.62 × 10 ⁻¹⁶	Thin (2-3 mm)
				low-porosity
	1005	0.44		layer
WI-F-4_3	1325	0.44	7.88×10^{-18}	Thin (2-3 mm)
				low-porosity
WI-F-4 4	1335	0.43	8 89 x 10-17	Thin (2-3 mm)
	1000	0.15	0.07 ~ 10	low-porosity
				layer
WI-F-13_1	1134	0.51	4.96 × 10 ⁻¹⁶	Alunite
WI-F-13_2	1164	0.49	1.52 × 10 ⁻¹⁶	Alunite
WI-F-14	1115	0.51	2.82 × 10 ⁻¹⁵	Alunite
WI-F-18	1299	0.45	8.03 × 10 ⁻¹⁴	Alunite
WI-F-20_1	1125	0.51	1.22 × 10 ⁻¹³	
WI-F-20_2	1117	0.52	1.28 × 10 ⁻¹³	
WI-F-20_3	1147	0.51	1.36 × 10 ⁻¹³	
WI-F-25_1	898	0.61	3.81 × 10 ⁻¹³	
WI-F-25_2	943	0.61	3.18 × 10 ⁻¹³	
WI-F-25_3	922	0.61	2.32 × 10 ⁻¹³	
WI-F-27_1	1290	0.46	1.55 × 10 ⁻¹⁵	Alunite
WI-F-27_2	1347	0.43	1.38 × 10 ⁻¹⁵	Alunite
WI-F-27_3	1391	0.42	4.82 × 10 ⁻¹⁵	Alunite
WI-F-34_1	1098	0.53	3.01 × 10 ⁻¹³	
WI-F-34_2	1124	0.51	3.52 × 10 ⁻¹⁴	
WI-F-34_3	1134	0.52	4.44 × 10 ⁻¹³	
WI-F-35_1	1216	0.48	9.97 × 10 ⁻¹⁴	
WI-F-35_2	1189	0.49	3.76 × 10 ⁻¹⁴	
WI-F-37_1	1393	0.40	2.00×10^{-14}	
WI-F-37_2	1379	0.64	1.08 × 10 ⁻¹⁴	
WI-F-42_1	1190	0.50	7.32 × 10 ⁻¹⁶	
WI-F-42_2	1200	0.49	6.26 × 10 ⁻¹⁴	
WI-F-44_1	911	0.59	4.74 × 10 ⁻¹³	Alunite
WI-F-44_2	905	0.59	5.19 × 10 ⁻¹³	Alunite
WI-F-44_3	1059	0.54	2.59 × 10 ⁻¹³	Alunite
WI-F-46_1	803	0.66	2.73 × 10 ⁻¹³	Alunite
WI-F-46_2	825	0.65	3.87 × 10 ⁻¹³	Alunite

WI-F-48	1001	0.57	5.77 × 10 ⁻¹³	
WI-F-52_1	1493	0.40	2.40×10^{-16}	Alunite; small
WI-F-52_2	1506	0.38	8.02 × 10 ⁻¹⁶	Alunite; small
WI-F-52_3	1498	0.39	9.40 × 10 ⁻¹⁶	Alunite; small
WI-F-54_1	1553	0.34	2.02 × 10 ⁻¹⁵	Alunite; small
WI-F-54_2	1474	0.38	5.85 × 10 ⁻¹⁵	Alunite; small
WI-F-55 1	1269	0.46	9.42 × 10 ⁻¹³	pores
	1242	0.47	1.26 × 10 ⁻¹²	
 WI-F-56 1	1107	0.49	1.48 × 10 ⁻¹³	
WI-F-56 2	1122	0.49	2.07×10^{-13}	
WI-F-56.3	1102	0.50	9.66 × 10-14	
WI-F-59	827	0.67	1 12 x 10-13	Alunite
WI-F-60 1	1236	0.07	1.12×10^{-14}	Alunite
WI-F-60 2	1101	0.17	2.40×10^{-15}	Alunite
WI-F-63_1	1202	0.50	2.93 × 10 ⁻¹⁵	Alunite
WI-F-63 2	1202	0.50	4.31 × 10 ⁻¹⁵	Alunite
	1225	0.50	5.05×10^{13}	Alunite
WI E 66	1102	0.30	1.29×10^{12}	
WI-F-00	1192	0.40	3.00 × 10 ⁻¹³	
WI-F-0/_1	1108	0.55	4.98 × 10 ⁻¹³	
WI-F-07_2	1000	0.55	6.55×10^{-13}	
WI-F-67_3	1086	0.55	7.09 × 10-13	Alexantes
WI-F-68_1	1078	0.54	3.54×10^{-14}	Alunite
WI-F-68_2	1076	0.54	Broke	Alunite
WI-F-70_1	1215	0.50	1.51×10^{-12}	
WI-F-70_2	1225	0.50	6.26 × 10 ⁻¹³	
WI-F-70_3	1201	0.51	1.11×10^{-12}	A1 11
WI-F-73	1264	0.49	3.60 × 10-14	Alunite
WI-F-74	967	0.60	1.51 × 10 ⁻¹²	
WI-F-75	1498	0.36	1.18 × 10 ⁻¹⁵	
WI-F-76	1074	0.52	1.90 × 10 ⁻¹⁴	
WI-F-78_1	1264	0.49	3.61 × 10 ⁻¹⁵	Alunite
WI-F-78_2	1175	0.52	4.09 × 10 ⁻¹⁵	Alunite
WI-F-79_1	1067	0.56	2.66 × 10 ⁻¹³	Alunite
WI-F-79_2	988	0.57	3.24 × 10 ⁻¹³	Alunite
WI-F-95_1	1559	0.38	7.38 × 10 ⁻¹⁴	
WI-F-95_2	1269	0.50	3.27 × 10 ⁻¹³	
WI-F-110_1	1521	0.41	8.62×10^{-18}	Alunite; small pores
WI-F-110_2	1474	0.44	2.85 × 10 ⁻¹⁶	Alunite; small pores
WI-F-110_3	1531	0.42	6.82 × 10 ⁻¹⁷	Alunite; small pores
WI-F-112_1	1486	0.42	2.38 × 10 ⁻¹⁶	Alunite; small pores
WI-F-112_2	1505	0.42	4.68 × 10 ⁻¹⁶	Alunite; small pores
WI-F-112_3	1445	0.44	4.30 × 10 ⁻¹⁶	Alunite; small pores
WI-F-115_1	1133	0.55	4.33 × 10 ⁻¹⁵	<u> </u>

WI-F-115_2	1145	0.55	1.40×10^{-16}	
WI-F-118_1	1048	0.56	1.82 × 10 ⁻¹³	
WI-F-118_2	994	0.55	2.21 × 10 ⁻¹³	
WI21b_A	1546	0.37	3.09 × 10 ⁻¹⁵	Alunite;
				Elutriation pipes
				parallel; Pc = 2
				МРа
WI21b_C	1410	0.44	1.80×10^{-16}	Alunite;
				Elutriation pipes
				parallel
WI21b_D	1511	0.38	6.27 × 10 ⁻¹⁶	Alunite;
				Elutriation pipes
	4.440	0.44		parallel
WI21b_E	1412	0.44	1.27×10^{-16}	Alunite;
				Elutriation pipes
	12(2	0.46	1.00 10.10	parallel
WIZID_H	1363	0.46	1.30×10^{-16}	Alunite;
				Elutriation pipes
WI21h I	1462	0.42	7 27 × 10-16	Alunitor
VV1210_1	1402	0.42	7.27 × 10-10	Flutriation pipes
				nernendicular:
				Pc = 2 MPa
WI21h I	1195	0.52	1 00 × 10 -16	Alunite
((1210_)	1155	0.52	1.07 ~ 10	Elutriation pipes
				perpendicular
WI21_K	1639	0.30	1.47 × 10 ⁻¹⁶	Alunite
WI21_20	1282	0.50	1.26 × 10-16	Alunite
WI22_A	1125	0.52	3.14 × 10 ⁻¹⁵	Pc = 2 MPa
WI22_B	1148	0.51	2.29 × 10 ⁻¹⁵	
WI22_17	1078	0.50	6.71 × 10 ⁻¹⁴	Diameter = 25
				mm
WI22_18	1178	0.49	1.55 × 10 ⁻¹³	Diameter = 25
				mm
WI22_22	1163	0.48	1.50×10^{-16}	
WI23_A	1311	0.48	1.21 × 10 ⁻¹⁵	Alunite; Pc = 2
				МРа
WI23_B	1343	0.45	1.22 × 10 ⁻¹⁶	Alunite
WI24_A	1144	0.52	3.08 × 10 ⁻¹³	Pc = 2 MPa
WI24_B	1144	0.52	2.03×10^{-14}	

Table A6. Summary of the laboratory data for the surficial samples (13 jarosite and
sulphur crusts, and 4 ash tuff samples). All values of permeability (nitrogen) were
measured under a confining pressure of 1 MPa. Photographs of the samples are
provided in Figure A12.

Sample	Bulk density	Connected	Permeability	Notes
	(kg/m ³)	porosity	(m ²)	
WI25_A	2017	0.31	2.63 × 10 ⁻¹²	
WI25_B	2104	0.32	3.47 × 10 ⁻¹²	
WI25_C	2128	0.32	2.34 × 10 ⁻¹²	
WI25_D	2133	0.30	1.89 × 10 ⁻¹²	
WI25_E	2074	0.31	3.84 × 10 ⁻¹²	
WI25_F	2041	0.32	5.12 × 10 ⁻¹²	
WI25_G	2171	0.32	3.04 × 10 ⁻¹²	
WI25_H	2079	0.32	5.92 × 10 ⁻¹²	
WI25_I	1915	0.37	6.05 × 10 ⁻¹²	
WI26_A	1194	0.49	1.11 × 10 ⁻¹²	
WI26_B	1474	0.40	3.99 × 10 ⁻¹³	Very high
				sulphur
				content
WI26_C	1429	0.46	1.75 × 10 ⁻¹²	High sulphur
				content
WI26_D	1314	0.37	7.81 × 10 ⁻¹³	
WI29_1	1986	0.01	< 10 ⁻¹⁸	Permeability
				too low for
				benchtop
				measurement
WI29_2	1984	0.01	< 10 ⁻¹⁸	Permeability
				too low for
				benchtop
				measurement
WI29_3	1990	0.01	< 10 ⁻¹⁸	Permeability
				too low for
				benchtop
				measurement
WI29_4	1977	0.02	< 10 ⁻¹⁸	Permeability
				too low for
				benchtop
				measurement

- **Table A7**. Summary of the laboratory permeability data for the two samples (one lavasample WI20 and one ash tuff sample WI21b) measured under different confining
- pressures (from 1 to 30 MPa). The porosity was 0.05 and 0.37 for the samples WI20 and
- 1593 WI21b, respectively.
- 1594

Sample	Confining pressure (MPa)	Permeability (m ²)
WI20	1	1.03×10^{-16}
WI20	2	7.63 × 10 ⁻¹⁷
WI20	5	6.63 × 10 ⁻¹⁷
WI20	10	5.32 × 10 ⁻¹⁷
WI20	15	4.36 × 10 ⁻¹⁷
WI20	20	3.62 × 10 ⁻¹⁷
WI20	25	3.00×10^{-17}
WI20	30	2.39 × 10 ⁻¹⁷
WI21b	1	7.64 × 10 ⁻¹⁵
WI21b	2	6.08 × 10 ⁻¹⁵
WI21b	5	5.55 × 10 ⁻¹⁵
WI21b	10	5.21 × 10 ⁻¹⁵
WI21b	15	5.05 × 10 ⁻¹⁵
WI21b	20	4.94×10^{-15}
WI21b	25	4.76 × 10 ⁻¹⁵
WI21b	30	4.73 × 10 ⁻¹⁵