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1 Permeability of volcanic rocks to gas and water

2

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6

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8

9 **Highlights**

10

- 11 • Gas permeability is a factor of 2-5 higher than water permeability in two volcanic rocks.
- 12 • The average pore radius (assuming tube-shaped pores) used by gas, calculated using the
13 Klinkenberg slip factor, is ~0.1-0.5 μm .
- 14 • We speculate that the radius of these microstructural elements is restricted or that they are
15 inaccessible to water due to water adsorption.
- 16 • A difference between water and gas permeability has important implications for fluid flow in
17 volcanic systems.

18

19 **Abstract**

20 The phase (gas or liquid) of the fluids within a porous volcanic system varies in both time and
21 space. Laboratory experiments have shown that gas and water permeabilities can differ for the same
22 rock sample, but experiments are biased towards rocks that contain minerals that are expected react
23 with the pore fluid (such as the reaction between liquid water and clay). We present here the first
24 study that systematically compares the gas and water permeability of volcanic rocks. Our data show
25 that permeabilities to argon gas and deionised water can differ by a factor between two and five in
26 two volcanic rocks (basalt and andesite) over a confining pressure range from 2 to 50 MPa. We
27 suggest here that the microstructural elements that offer the shortest route through the sample—
28 estimated to have an average radius $\sim 0.1\text{-}0.5\ \mu\text{m}$ using the Klinkenberg slip factor—are accessible
29 to gas, but restricted or inaccessible to water. We speculate that water adsorption on the surface of
30 these thin microstructural elements, assumed here to be tortuous/rough microcracks, reduces their
31 effective radius and/or prevents access. These data have important implications for fluid flow and
32 therefore the distribution and build-up of pore pressure within volcanic systems.

33

34 **Keywords:** permeability; Klinkenberg; volcano; pore fluid; microstructure

35 **1. Introduction**

36 Permeability is a measure of the ability of a material to transmit fluids [*Guéguen and*
37 *Palciauskas, 1994*]. The permeability of volcanic rocks therefore largely controls the movement of
38 fluids and the distribution of pore pressure in a volcanic edifice. The build-up of pore pressure within
39 a volcanic system is thought to promote explosive volcanism [e.g., *Eichelberger et al., 1986; Sparks,*
40 *1997; Melnik et al., 2005; Farquharson et al., 2017*] and flank collapse [e.g., *Reid, 2004*] and, as such,
41 many experimental and theoretical studies have been devoted to better understanding the
42 permeability of volcanic materials [e.g., *Eichelberger et al., 1986; Saar and Manga, 1999; Blower,*
43 *2001; Rust and Cashman, 2004; Mueller et al., 2005; Costa, 2006; Mueller et al., 2008; Wright et al.,*
44 *2009; Degruyter et al., 2010; Lavallée et al., 2013; Gaunt et al., 2014; Farquharson et al., 2015, 2016;*
45 *Heap and Kennedy, 2016; Wadsworth et al., 2016; Lindoo et al., 2016; Heap et al., 2016, 2017; Burgisser*
46 *et al., 2017; Kushnir et al., 2017a; Colombier et al., 2017; Vasseur and Wadsworth, 2017; Kushnir et al.,*
47 *2017b*]. These studies, and many others, have shown that the permeability of volcanic materials is
48 not only an increasing function of porosity (a scalar), but also highlight the importance of a wealth of
49 additional parameters, such as porosity type (pores and microcracks), pore geometry (size, shape,
50 and preferred orientation), porosity connectivity, and alteration, amongst others.

51 The constitutive equation that describes fluid flow through a porous medium is Darcy's law
52 [*Darcy, 1856*]. The equation relates a volumetric flow rate to a fluid pressure gradient using a
53 coefficient called the permeability. Permeability is a property of the medium and is therefore
54 independent of the fluid used in its determination. Darcy's law is valid for all porous media as long as
55 the volumetric flow rate is linearly proportional to the fluid pressure gradient, i.e. the flow is laminar.
56 For instances of nonlaminar flow, auxiliary corrections are required to derive the "true" (Darcian)
57 permeability of a medium from pressure and flow rate data. For example, when measuring materials
58 with very high permeabilities and/or using pore fluids with very low viscosities, fluid flow can be
59 turbulent. In this regime, a new parameter is introduced in order to account for inertial forces: this

60 is known as the Forchheimer correction [*Forchheimer*, 1901]. Another instance where the
61 relationship between volumetric flow rate and fluid pressure gradient is nonlinear is specific to
62 measurements using gas. When the mean free path of the molecules/atoms approaches the
63 characteristic pore or aperture size, interactions between the gas molecules/atoms and the pore
64 walls serve to reduce resistance to flow, a phenomenon known as “slip flow” or “gas slippage”. In this
65 case, the apparent permeability is corrected using the Klinkenberg correction [*Klinkenberg*, 1941].

66 Most of the experimental studies designed to measure the permeability of volcanic rocks use
67 an inert gas as the pore fluid; only a handful of studies have used water [e.g., *Kolzenburg et al.*, 2012;
68 *Kendrick et al.*, 2013; *Heap et al.*, 2014a, b; *Gaunt et al.*, 2014]. To the knowledge of the authors, there
69 are no studies that compare gas and water permeabilities in volcanic materials. Laboratory
70 measurements have shown that permeabilities to liquid water and gas are essentially equal for
71 granite [*Brace et al.*, 1968; *Zhang et al.*, 2000]. A difference between permeabilities to gas and water
72 has been observed for rocks containing mineral constituents that can react with the pore fluid, such
73 as the reaction between liquid water and clay [*Faulkner and Rutter*, 2000, 2003; *Tanikawa and*
74 *Shimamoto* 2006; *Davy et al.*, 2007; *Tanikawa and Shimamoto* 2009; *Behnsen and Faulkner*, 2011] or
75 the reaction between CO₂-enriched water and calcite [*Noiriel et al.*, 2004; *Luquot and Gouze*, 2009].
76 These studies have shown, for example, that permeabilities to gas and liquid water in clay-rich rocks
77 can differ by as much as an order of magnitude [e.g., *Faulkner and Rutter*, 2000]. A higher
78 permeability to gas in clay-rich rocks is explained by the expansion of clay minerals in the presence
79 of water due to water adsorption – the swollen clay minerals effectively constrict pore throats thus
80 reducing permeability [*Faulkner and Rutter*, 2003].

81 It is often assumed, after the appropriate corrections have been applied, that permeabilities
82 to gas and water are equivalent for materials for which significant physicochemical reactions are not
83 expected. However, a lack of data comparing the gas and water permeabilities of a range of materials
84 obscures a complete understanding. For example, the complex microstructure presented by volcanic

85 rocks—often a complex network of pores and microcracks [*Heap et al., 2014a; Farquharson et al.,*
86 *2015; Colombier et al., 2017*—may yield a difference in permeabilities to gas and water in the
87 absence of significant physicochemical reactions. The phase (gas or liquid) of the fluids within a
88 porous volcanic system varies in time and space [e.g., *Giggenbach and Soto, 1992; Taran et al., 1998;*
89 *Chiodini et al., 2000; Byrdina et al., 2014*], exemplified by the occurrence of phreatic and
90 phreatomagmatic eruptions that are driven by the boiling of liquid water [e.g., *Houghton and Nairn,*
91 *1991; Barberi et al., 1992; Mayer et al., 2015; Montanaro et al., 2016*]. As a result, quantifying potential
92 differences in the permeability to gas and water in volcanic rocks emerges as an important, yet
93 unexplored, avenue for research. With this in mind, we present herein a study in which we measured
94 the gas and water permeabilities of samples of andesite and basalt.

95

96 **2. Description of experimental materials**

97 For the purpose of this study, we selected a suite of variably porous andesites from Volcán de
98 Colima (Mexico) and a well-studied basalt from Mt Etna (Italy). Volcán de Colima is an active andesitic
99 stratovolcano located at the western end of the Trans-Mexican Volcanic Belt in Mexico [*Varley and*
100 *Komorowski, 2018*] and Mt Etna is an active basaltic stratovolcano located on the east coast of the
101 island of Sicily (Italy) [*Allard et al., 2006*].

102 The five andesite blocks used in this study (A5, B4, B5, C8, and LAH4) have been used in a
103 number of recent studies focussed on the mechanical behaviour of andesite [e.g., *Kendrick et al., 2013;*
104 *Heap et al., 2014a; 2015*]. Using the classification scheme of *Farquharson et al. [2015]*, sample B5 can
105 be classified as an “altered lava” (B5 displays high-temperature alteration, as evidenced by the
106 presence of cristobalite; Figure 1a), while the remaining blocks can be classified as “lava”. The
107 andesites all have a porphyritic texture consisting of a glassy groundmass (with abundant microlites)
108 that hosts pores and a phenocryst cargo (Figure 1). The porosity within these andesites comprises a
109 combination of microcracks and pores (Figure 1) [*Kendrick et al., 2013; Heap et al., 2014a; 2015*]. As

110 evident in the scanning electron microscope (SEM) images of Figure 1, the andesites contain a
111 microcrack network that is both pervasive and tortuous. Indeed, microcrack densities range between
112 35 and 45 mm⁻¹ [Heap *et al.*, 2014a]. The microcracks are typically thin (no more than a few microns)
113 and are usually no longer than 0.5 mm in length (Figure 1). The andesites also contain high pore
114 number densities, between 3.3 and 8.1 mm⁻² [Heap *et al.*, 2014a]. The pores are often far from
115 spherical (Figure 1) and are, in the case of the lowest porosity sample (B5), coated with cristobalite
116 crystals (Figure 1a). All of the andesites show a wide range of pore diameters, from 1-2 mm to a few
117 tens of microns [Heap *et al.*, 2014a].

118 The basalt from Mt Etna has porphyritic texture consisting of a completely crystallised
119 groundmass containing pores and a phenocryst cargo (Figure 2). This basalt has been the focus of a
120 number of studies in the last ten years [e.g., Vinciguerra *et al.*, 2005; Stanchits *et al.*, 2006; Heap *et al.*,
121 2011; Zhu *et al.*, 2016]. The porosity within the basalt comprises a combination of microcracks and
122 pores (Figure 2). Qualitatively, the microcracks in the basalt are typically much longer (often several
123 mm) than those in the andesites (Figures 1 and 2). The microcracks within the basalt often traverse
124 through both the crystallised groundmass and the large phenocrysts present within the sample
125 (Figure 2a). The pores within the basalt are not distributed throughout the sample, but are present
126 in pockets (Figure 2b). The pores within these porous pockets represent the volume between
127 microlites where the groundmass is absent, a texture termed diktytaxitic [see, for example, Kushnir
128 *et al.*, 2016]. The pores are typically less than 100 microns in diameter (Figure 2b).

129

130 **2. Experimental methods**

131 Cylindrical samples (20 mm in diameter and precision-ground to a nominal length of 40 mm)
132 were prepared from the five blocks of andesite (A5, B4, B5, C8, and LAH4) and the block of basalt.
133 These samples were then dried in a vacuum oven at 40 °C for at least 48 h. Before measuring their
134 permeability, the prepared 20 mm-diameter samples were first investigated in terms of their

135 connected porosity (using helium pycnometry) and specific surface area (using Brunauer, Emmett,
136 and Teller (BET) gas adsorption measurements).

137 The connected porosity of each sample was measured using the skeletal (connected) volume
138 provided by a helium pycnometer (Micromeritics AccuPyc II 1340) (values are an average of 20
139 measurements; precision $\pm 0.00005 \text{ cm}^3$) and the bulk volume determined using the sample
140 dimensions (precision $\pm 0.005 \text{ mm}$). The specific surface area was determined using BET nitrogen or
141 krypton adsorption measurements [Brunauer *et al.*, 1938]. The samples were placed a vacuum sealed
142 vessel (itself placed in a liquid nitrogen bath at a temperature of $-196.15 \text{ }^\circ\text{C}$) and the specific surface
143 area was calculated by determining the amount of adsorbate gas needed to create a monomolecular
144 layer on the connected surface inside the sample. The absorbent gas used for the basalt was nitrogen.
145 Krypton gas was used for the andesite samples due to their low specific surface areas (krypton is
146 better suited for samples with low specific surface areas due to its smaller molecular size). More
147 details on the BET method and theory employed here can be found in *Brunauer et al.* [1938] and
148 *Kushnir et al.* [2016].

149 To assess their pore throat structure, mercury injection porosimetry was performed on
150 pieces ($\sim 5 \text{ g}$) of two of the andesite blocks (B5 and C8) and the basalt using the Micromeritics
151 Autopore IV 9500 at the University of Aberdeen (Scotland). The evacuation pressure and evacuation
152 time were $50 \text{ }\mu\text{mHg}$ and 5 min , respectively, and the mercury filling pressure and equilibration time
153 were $0.52 \text{ pounds per square inch absolute (psia)}$ and 10 s , respectively. The pressure range was 0.1
154 to $60,000 \text{ psia}$ (i.e. up to a pressure of about 400 MPa). Mercury injection data permit the calculation
155 of the pore throat size distribution within a particular sample. The mercury injection data were
156 corrected for the “low pressure correction” recommended by ASTM International [ASTM D4404-10].

157 The permeabilities to gas (argon) and water (deionised water) were then measured on the
158 prepared cylindrical samples. We first performed a suite of gas and water permeability
159 measurements on the variably porous andesite samples at a confining pressure of 2 MPa . We then

160 performed gas and water permeability measurements at a range of confining pressures (from 1 to 50
161 MPa) on a sample of andesite (B5) and a basalt sample. To avoid problems associated with potential
162 permanent microstructural changes following exposure to 50 MPa, we used different samples (cored
163 from the same block) for the gas and water permeability measurements in these latter experiments.
164 The pairs of samples for these experiments were selected based on their almost identical initial
165 permeabilities (measured with inert gas at a confining pressure of 1 MPa).

166 Samples to be measured with water were first vacuum-saturated with deionised water. The
167 saturation procedure consisted of three steps:

- 168 (1) the samples were vacuum-dried at 40 °C for at least 48 h,
- 169 (2) the samples were then immediately placed inside a belljar which was then vacuumed for
170 at least 12 h and, finally,
- 171 (3) degassed (using a Venturi siphon with municipal water as the motive fluid), deionised
172 water was introduced into the belljar (whilst under vacuum).

173 Great care was taken to ensure that the water was fully degassed and that the samples were fully
174 saturated. Once prepared, the samples were jacketed in a viton sleeve and placed inside a hydrostatic
175 pressure vessel. The confining pressure (P_c) was then increased to 2 MPa (for the set of
176 measurements on the variably porous andesite sample suite) or 1 MPa (for the measurements to be
177 performed at different confining pressures). The samples were left overnight at this pressure to allow
178 for microstructural equilibration. All measurements of water permeability were performed using the
179 steady-state flow method. Following microstructural equilibrium, a pressure gradient was imposed
180 across the sample and the flow rate measured using an electronic balance (with a precision ± 0.0005
181 g). Once steady-state flow had been established, the water permeability k_{water} was determined using
182 Darcy's relation:

183

$$184 \quad \frac{Q}{A} = \frac{k_{water}}{\eta L} (P_{up} - P_{down}), \quad (1)$$

185

186 where Q is the volumetric flow rate, A is the cross-sectional area of the sample, P_{up} and P_{down} represent
187 the upstream and downstream pressure, respectively (where P_{down} is the atmospheric pressure), L is
188 the length of the sample, k_{water} is the permeability to water, and η is the viscosity of the pore fluid
189 (taken here as 1.008×10^{-3} Pa.s). A pressure gradient (i.e. $P_{up} - P_{down}$) of 0.5 MPa was used for all of
190 the steady-state measurements using water as the pore fluid (i.e. the mean pore fluid pressure, P_m ,
191 was 0.35 MPa, where $P_m = (P_{up} + P_{down})/2$).

192 Gas (argon) permeability was measured using either the steady-state method (for the
193 andesites) or the pulse-decay method (for the basalt). The method chosen was based on the
194 permeability of the sample: the steady-state method is better suited for high-permeability samples
195 and the pulse-decay method is better suited for low-permeability samples (we note that there is no
196 experimental bias between the methods: the permeability values of samples of intermediate
197 permeability measured using both techniques in our laboratory are essentially identical). For the
198 steady-state method, a pressure gradient was imposed across the sample (following microstructural
199 equilibrium) and the outlet flow rate was measured using one of three Bronkhorst flowmeters. The
200 choice of flowmeter depended on the permeability of the sample and therefore volumetric flow rate
201 (the volumetric flow rate range for the three flowmeters: 1, 3, and 125 ml/min). Since the pore fluid
202 is compressible, the raw permeability to gas k_{gas_raw} is expressed as [Scheidegger, 1947]:

203

204
$$\frac{Q}{A} = \frac{k_{gas_raw}}{\eta L} \frac{(P_{up})^2 - (P_{down})^2}{2P_{down}}, \quad (2)$$

205

206 where η , the viscosity of the pore fluid, was taken as 2.21×10^{-5} Pa.s. Steady-state volumetric flow
207 rate Q measurements were taken under several pore pressure gradients (i.e. $P_{up} - P_{down}$, where
208 P_{down} is the atmospheric pressure) to check whether any auxiliary corrections were required. The

209 magnitude of pore pressure gradients used for the gas steady-state measurements varied depending
 210 on the permeability of the sample, but never exceeded 0.5 MPa (i.e. $P_m \leq 0.35$ MPa). We first plot
 211 $1/k_{gas_raw}$ as a function of Q to check whether the Forchheimer correction is required. The
 212 correction is necessary if these data can be well described by a positive linear slope. The
 213 Forchheimer-corrected permeability is taken as the inverse of the y -intercept of the best-fit linear
 214 regression in the plot of $1/k_{gas_raw}$ as a function of Q . An example of data that required the
 215 Forchheimer correction is shown in Figure 3a (for sample LAH4_7). If the Forchheimer correction is
 216 not required, we then check whether the Klinkenberg correction is required. To do so, we plot
 217 k_{gas_raw} as a function of the reciprocal mean pressure, $1/P_m$. The Klinkenberg correction is required
 218 if these data can be well described by a positive linear slope and, if true, the Klinkenberg-corrected
 219 permeability can be taken as the y -intercept of the best-fit linear regression in the plot of k_{gas_raw} as
 220 a function of $1/P_m$. The Forchheimer correction was required for the high-porosity andesites (Table
 221 1) and the Klinkenberg correction was required for the low-porosity andesites (Table 1).

222 We used the pulse decay method [Brace *et al.*, 1968] to measure the gas permeability of the
 223 basalt sample. Following microstructural equilibrium at the target confining pressure, the decay of
 224 an initial pore pressure differential ($P_{up} - P_{down} = 0.5$ MPa, where P_{down} is the atmospheric pressure;
 225 i.e. $P_m = 0.35$ MPa at the start of the experiment and decayed to the atmospheric pressure with time)
 226 was monitored using a pressure transducer following the closure of the upstream pressure inlet. The
 227 gas permeability k_{gas_raw} was then determined using the following relation:

228

$$229 \quad k_{gas_raw} = 2 \frac{\eta L}{A} \frac{V_{up}}{P_{up}^2 - P_{down}^2} \frac{dP_{up}}{dt}, \quad (3)$$

230

231 where V_{up} is the volume of the upstream pore pressure circuit ($= 7.8 \times 10^{-6}$ m³) and t is time. As before,
 232 we checked whether these data required any auxiliary corrections (the Forchheimer or Klinkenberg

233 correction). We found that the Klinkenberg correction was required for all of the basalt
234 measurements (Table 1). An example of data that required the Klinkenberg correction is shown in
235 Figure 3b (for sample EB_3).

236 Each permeability measurement was remeasured the following day. The sample was only
237 removed, or subjected to the next confining pressure increment, if the retrieved permeability was
238 the same as for the previous day. All the measurements in this study were performed under ambient
239 laboratory temperatures.

240

241 **3. Results**

242 3.1 Connected porosity, specific surface area, and pore throat size distribution

243 The connected porosities of the andesite samples ranged from 0.08 to 0.27 and the connected
244 porosities of the basalt samples were measured to be 0.04 (Table 1).

245 The specific surface area of the andesites varied from 15 to 100 m² kg⁻¹ (Table 1). We note
246 that the specific surface area does not correlate with connected porosity: the andesite with the
247 highest specific surface area (B5; Figure 1a) contains one of the lowest porosities (Table 1). The
248 specific surface of the basalt block was measured to be 126 m² kg⁻¹ (Table 1).

249 Mercury porosimetry, which provides the pore throat size distribution, was performed on a
250 low-porosity (B5) and a high-porosity (C8) andesite (Figure 4a) and the basalt (Figure 4b). About
251 50% of the porosity in andesite sample C8 (porosity = 0.144) is connected by pore throats with a
252 radius $\geq 5 \mu\text{m}$. Pore throats with a radius $\geq 5 \mu\text{m}$ connect only 35% of the porosity in andesite sample
253 B5 (porosity = 0.076) (Figure 4a). Only 10% of the porosity in samples C8 and B5 is connected by
254 pore throats with radii $\leq 0.5 \mu\text{m}$ (Figure 4a). The average pore throat radius was determined to be
255 2.15 and 1.05 μm for andesites C8 and B5, respectively. The mercury porosimetry data for the basalt
256 show that 65% of the porosity is connected by pore throats with a radius below 0.5 μm (Figure 4b).
257 The average pore throat radius for the basalt was determined to be 0.17 μm .

258

259 3.2 Influence of porosity on gas and water permeabilities

260 Gas and water permeabilities were measured for a suite of variably porous andesites from
261 Volcán de Colima (Figure 5; Table 1). These measurements were all collected under a constant
262 confining pressure of 2 MPa. First, and as observed in previous studies [e.g., *Farquharson et al.*, 2015],
263 permeability is higher at higher porosities. Our data further show that gas permeability is higher than
264 water permeability over the entire porosity range (0.08 to 0.27) (Figure 5; Table 1). The difference
265 between gas and water permeability is between a factor of 1.1 and 5.5 (Figure 5; Table 1).

266

267 3.3 Influence of confining pressure on gas and water permeabilities

268 Gas and water permeabilities were measured on a sample of andesite (B5) and a sample of
269 basalt as a function of confining pressure (from 1 to 50 MPa) (Figure 6; Table 1). For both samples,
270 and both pore fluids, large decreases in permeability are observed between 1 and 10 MPa (Figure 6).
271 At confining pressures of 15 MPa and above, the permeability decrease per increment of confining
272 pressure is reduced (Figure 6). We also note that the absolute decrease in permeability from 1 to 50
273 MPa is much greater in the basalt than in the andesite: an order of magnitude in the case of the former
274 and only a factor of three for the latter (Figure 6; Table 1). The difference between gas and water
275 permeabilities is about a factor of three for the andesite and about a factor of four for the basalt (Table
276 1). For both rock types, this offset does not change significantly as confining pressure is increased
277 (Figure 6; Table 1).

278

279 **4. Discussion**

280 A difference in permeability when using different fluids is usually considered the
281 consequence of a physicochemical reaction between the mineral constituents of the rock and the pore
282 fluid, such as the reaction between liquid water and clay [*Faulkner and Rutter*, 2000, 2003; *Tanikawa*

283 and Shimamoto 2006; Davy et al., 2007; Tanikawa and Shimamoto 2009; Behnsen and Faulkner, 2011]
 284 or the reaction between CO₂-enriched water and calcite [Noiriel et al., 2004; Luquot and Gouze, 2009].
 285 It is for this reason that gas and water permeabilities are rarely measured and compared when the
 286 rock-forming minerals are not expected to react with the pore fluid. One such study [Brace et al.,
 287 1968] showed that gas and water permeabilities are essentially equal in intact Westerly granite
 288 (porosity = 0.008) over a range of confining pressures between 10 and 100 MPa. Our new data show
 289 that gas and water permeabilities can differ in volcanic rocks by up to a factor of five (Figures 5 and
 290 6). Since these volcanic rocks do not contain minerals (such as clay) that are expected to undergo
 291 significant physicochemical reactions in the presence of water, there must be another explanation
 292 for the measured difference in gas and water permeability.

293 To better understand the microstructural path taken by the gas, we can use the Klinkenberg
 294 slip factor, b , (which has the units of pressure) [Klinkenberg, 1941] to provide an estimate of the
 295 average radius of the pores used by the gas molecules. The Klinkenberg slip factor has previously
 296 been used to examine the average pore radius of the flow path in low-porosity rocks such as shales
 297 [e.g., Heller et al., 2014; Firouzi et al., 2014; Letham and Bustin, 2015]. Since the mean free path is
 298 inversely proportional to P_m , Poiseuille's law for gas flow in a cylindrical tube and Darcy's law for
 299 flow in porous media yields the following relation:

300

$$301 \quad k_{gas} = k_{gas_{raw}} \left(1 + \frac{b}{P_m} \right), \quad (4)$$

302

303 where k_{gas} is the true (Klinkenberg-corrected) gas permeability. Therefore, and assuming tube-
 304 shaped pores, the average pore radius r can be estimated using the following relation [Civan, 2010]:

305

$$306 \quad r = \frac{4}{b} \eta \sqrt{\frac{\pi R_g T}{2 M_w}}, \quad (5)$$

307
308 where R_g is the ideal gas constant (taken as $8.31 \text{ J mol}^{-1} \text{ K}^{-1}$), T is the temperature (taken as 293 K),
309 and M_w is the molecular mass of the pore fluid (taken as $0.03995 \text{ kg mol}^{-1}$). The average pore radius
310 that controls the flow of gas molecules (calculated using Equation (5)) for the sample of andesite (B5)
311 and basalt as a function of confining pressure is shown in Figure 7. The calculations show that the
312 average pore radius in the andesite is $\sim 0.5 \text{ }\mu\text{m}$ at low pressure (between 1 and 2 MPa) and evolves
313 to, and stays constant at, $\sim 0.2\text{-}0.25 \text{ }\mu\text{m}$ when the confining pressure is at or above 15 MPa (Figure 7;
314 Table 1). The average pore radius in the basalt is $\sim 0.3 \text{ }\mu\text{m}$ at a confining pressure of 1 MPa (Figure 7;
315 Table 1). The average pore radius in the basalt reduces to, and stays constant at, $\sim 0.13\text{-}0.16 \text{ }\mu\text{m}$ when
316 the confining pressure is at or above 15 MPa (Figure 7; Table 1). The Klinkenberg slip factor can also
317 be used to estimate the width of “slit-shaped” pores (i.e. microcracks) [e.g., *Heller et al., 2014; Letham*
318 *and Bustin, 2015*], which, according to our microstructural analyses (Figures 1 and 2) and mercury
319 injection data (Figure 4), may better suit these rocks, especially the basalt (Figures 2 and 4b).
320 However, the widths predicted using the equation presented in *Heller et al. [2014]* and *Letham and*
321 *Bustin [2015]* are within the range $\sim 3\text{-}11.5$ and $\sim 3\text{-}6.5 \text{ }\mu\text{m}$ for the andesite and basalt, respectively.
322 We consider such widths unrealistically high. The basalt, for example, not only has an average pore
323 throat diameter of $0.34 \text{ }\mu\text{m}$ (determined by mercury porosimetry), but the mercury injection data
324 also show that only $\sim 2\%$ of the void space is connected by pore throats with diameters larger than
325 $6.5 \text{ }\mu\text{m}$ (Figure 4b). Although it is difficult at present to forward a reason as to why the average width
326 of a slit-shaped pore provides an overestimation of the size of the microstructural elements carrying
327 the gas, while the average radius of a tube-shaped pore does not, we highlight that these geometries
328 only represent end-member geometries in rocks characterised by geometrically complex pore
329 networks (Figures 1, 2, and 4). As a result, although the average pore radii provided in Figure 7
330 inform on the size of the microstructural element used by the gas molecules (i.e. submicron), we
331 highlight that these values are estimations that assume a cylindrical pore shape. The average pore

332 radius used by the gas molecules in the basalt predicted using Equation 5 ($\sim 0.13\text{-}0.3\ \mu\text{m}$; Figure 7)
333 is very similar to the average pore throat radius determined by the mercury porosimetry ($0.17\ \mu\text{m}$).
334 This is likely a consequence of the relatively narrow range of pore and pore throat sizes (Figure 4b)
335 within the basalt: 80% of the void space is connected by pore throat radii between ~ 0.7 and $\sim 1.5\ \mu\text{m}$
336 (Figure 4b). However, the average pore throat radius determined by mercury porosimetry is 2.15
337 and $1.05\ \mu\text{m}$ for andesites C8 and B5, respectively, much greater than the radii predicted using the
338 Klinkenberg slip factor ($\sim 0.25\text{-}0.5\ \mu\text{m}$; Figure 7). Although the andesites contain microstructural
339 elements (microcracks and tubes) of varying diameter (Figure 4a), resulting in a high average pore
340 throat radius, pores within this size range are likely not used for flow, which is likely obliged to
341 negotiate through narrow microstructural elements (on the submicron scale).

342 Based on the mercury porosimetry (Figure 4) and the average pore radii (assuming a
343 cylindrical pore shape) predicted using Equation 5 (Figure 7), it is likely that the gas in both samples
344 (andesite and basalt) is travelling through thin and tortuous/rough microcracks. This inference is
345 supported by the evolution of permeability as confining pressure increases (Figure 6). A reduction in
346 the permeability of microcracked volcanic rocks with increasing confining pressure has been
347 previously explained by the narrowing of microcrack apertures or the closing of microcracks [e.g.,
348 *Vinciguerra et al.*, 2005; *Nara et al.*, 2011; *Fortin et al.*, 2011; *Heap et al.*, 2017], supported here by the
349 decrease in our average pore radii estimations with increasing confining pressure (Figure 7).
350 However, the reduction in permeability with confining pressure for the volcanic rocks studied herein
351 (Figure 6) are much less than those typically observed for microcracked granites [e.g., *Le Ravalec et*
352 *al.*, 1996; *David et al.*, 1999; *Darot and Reuschlé*, 2000]. We consider this a consequence of the
353 tortuous/rough nature of the microcracks within the studied andesite and basalt (Figures 1 and 2):
354 tortuous and rough cracks require higher pressures to close than straight and smooth cracks [e.g.,
355 *Pérez-Flores et al.*, 2017]. Further, the more significant decrease in the permeability of the basalt with
356 increasing confining pressure—one order of magnitude compared to a factor of three in the andesite

357 (Figure 6)—must require that the microcracks supporting fluid flow in the andesite are more difficult
358 to close than those in the basalt. Indeed, qualitative microstructural observations suggest that the
359 microcracks in the andesite are shorter and more tortuous/kinked than those in the basalt (Figures
360 1 and 2), adding confidence to this hypothesis.

361 The estimations of average pore radii shown in Figure 7, and the average pore throat radii
362 determined from the mercury porosimetry data, highlight the microstructural complexity of volcanic
363 rocks. To emphasise, although ~90% of the void volume in the andesite is connected by pore throats
364 greater than 1 μm (Figure 4a), our pore radii predictions (Figure 7) suggest the gas flow is controlled
365 by microstructural elements that have an average radius of ~0.2-0.25 μm . To the authors' knowledge,
366 this represents the first time the Klinkenberg slip factor has been used to estimate the average pore
367 radius of gas flow paths in volcanic materials; we recommend that this method is utilised in future
368 studies.

369 We must now consider why these thin microstructural elements could be inaccessible to
370 water. *Faulkner and Rutter* [2000] inferred that the lower permeability to water than to gas in clay-
371 bearing fault gouge was a result of layers of structured water adsorbed onto the phyllosilicate
372 mineral surfaces, thereby reducing the effective pore throat aperture and reducing permeability.
373 However, there are no clay minerals in our studied materials. The permeability experiments of
374 *Klinkenberg* [1941], although designed to test the hypothesis that adsorbed molecules on the walls
375 of small capillaries in porous materials could inhibit fluid flow, found that the permeabilities to
376 different liquids were within experimental error. However, as discussed in *Faulkner and Rutter*
377 [2000], any water adsorption effect in the high-permeability ($\sim 10^{-14} \text{ m}^2$) glass filters of *Klinkenberg*
378 [1941] was likely masked by their large pore throat apertures. In other words, perhaps the pore radii
379 of the volcanic rocks tested herein are sufficiently narrow to observe a difference due to water
380 adsorption, while the apertures of glass filters tested by *Klinkenberg* [1941] are not. Although we
381 cannot advance a definitive reason for the difference between gas and water permeability in the

382 volcanic rocks measured herein, we speculate that water molecules adsorbed onto the surface of the
383 thin ($\sim 0.1\text{-}0.5\ \mu\text{m}$) microstructural elements are capable of narrowing their aperture or rendering
384 them inaccessible to the flow of water. It is the complex nature and poor connectivity of the void
385 space in these volcanic rocks (Figures 1, 2, and 4) that obliges fluid to negotiate these thin
386 microstructural elements, which are also characterised by complex geometries (i.e. tortuous, kinked,
387 and rough; see discussion above). We further speculate that it is the tortuous, kinked, and rough
388 nature of the microcracks that allows the adsorption of water to reduce the water permeability.
389 Adsorbed water molecules need not obstruct the entire length of a microcrack, but only a small
390 section – perhaps a particularly rough-walled section or at a tight bend or kink. Although this complex
391 microstructure is typical of lavas [e.g., *Colombier et al.*, 2017], it is unclear at present whether volcanic
392 rocks that contain a permeable backbone of large, well-connected tubes—such as some pumices—
393 will also display differences between gas and water permeabilities.

394 The implication of these data is that the permeability of the rocks comprising a volcanic
395 edifice will be higher to gas (e.g., CO_2 and SO_2 ; *Edmonds et al.* [2003]) than to liquid (e.g., the
396 infiltration of meteoric and seawater and the circulation of groundwater/hydrothermal fluids;
397 *Hurwitz et al.*, [2003]). Further, permeability may vary in zones that experience fluctuations in pore
398 fluid state as a result of fluctuating temperature or pressure. For example, a reduction in temperature
399 that results in a change in fluid state from gas to liquid may promote pore pressure build-up as a
400 result of a decrease in permeability. Alternatively, unloading of the edifice (due to mass movement
401 events, for example) could trigger decompression of the interstitial pore fluid, prompting a phase
402 change in the opposite direction (i.e. liquid to vapour). Importantly, the saturation curve for water
403 (which delineates the pressure-temperature boundary between liquid and vapour) occurs within
404 conditions relevant for a shallow edifice. Indeed, electrical resistivity tomography of the shallow
405 hydrothermal system of Campi Flegrei (Italy) shows a complex configuration of gaseous and liquid
406 zones, and zones characterised by mixtures of gases and liquids [*Byrdina et al.*, 2014]. The data

407 presented herein therefore have important ramifications for the distribution and build-up of pore
408 pressure in a volcanic system. Further, the choice of pore fluid used in laboratory investigations and
409 the choice of permeability values to be used in fluid flow modelling should be carefully considered
410 [e.g., *Collombet, 2009; Collinson and Neuberg, 2012; Fournier and Chardot, 2012; Chevalier et al.,*
411 *2017*]. Finally, we also highlight that care should be taken when collating porosity-permeability data
412 from published studies.

413

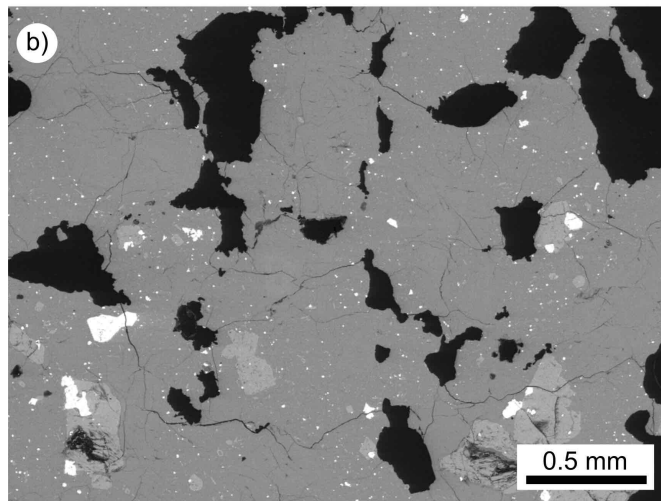
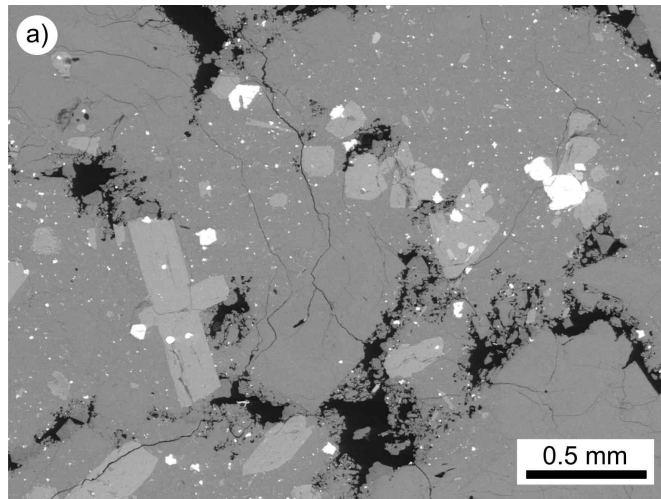
414 **5. Conclusions**

415 Our laboratory measurements highlight that gas permeability can be a factor of two to five
416 higher than water permeability in volcanic rocks. Using the Klinkenberg slip factor (assuming tube-
417 shaped pores), we estimate the average radius of the microstructural elements used by the gas flow
418 to be $\sim 0.1\text{-}0.5\ \mu\text{m}$. Although we cannot definitively advance a reason for the difference in gas and
419 water permeability, we speculate that water adsorption on the surfaces of these thin microstructural
420 elements—assumed here to be microcracks—may reduce their effective radius and/or prevent
421 access, thus reducing the water permeability of these volcanic rocks. We further speculate that it is
422 the tortuous, kinked, and rough nature of the microcracks that allows the adsorption of water to
423 reduce the water permeability. Absorbed water molecules need not obstruct the entire length of a
424 microcrack, but only a small section – perhaps a particularly rough-walled section or at a tight bend
425 or kink. Our data highlight the need for further studies that explore differences between gas and
426 water permeabilities in a range of volcanic materials. A difference between gas and water
427 permeabilities in volcanic materials has important ramifications for the distribution and build-up of
428 pore pressure in hydrothermal and geothermal systems in volcanically active regions, as well as
429 volcanic systems themselves.

430

431 **Acknowledgements**

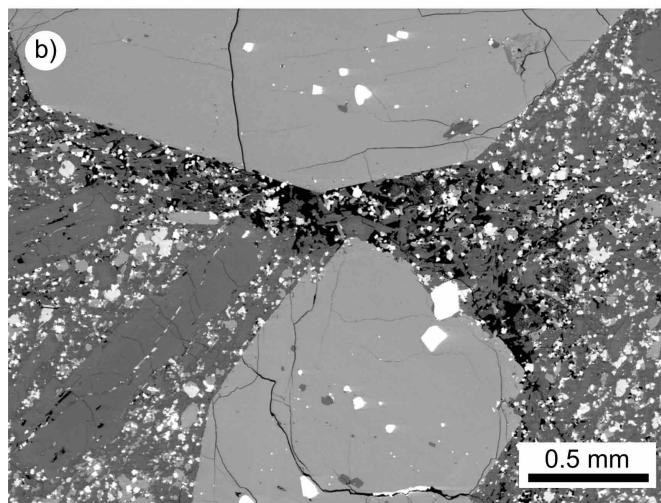
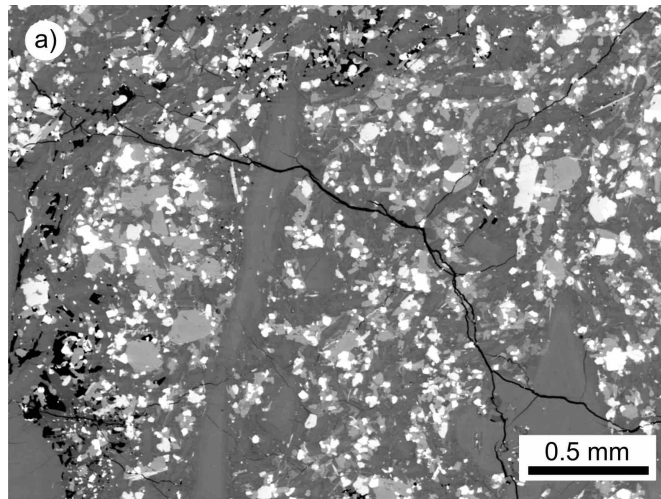
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439 of basalt from Mt Etna. The mercury porosimetry was performed at the University of Aberdeen;
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441 anonymous reviewers and the editor (Kelly Russell) for comments that helped improve this
442 manuscript.
443



444

445 **Figure 1.** (a) Backscattered scanning electron microscope image of andesite sample B5 (from Volcán
446 de Colima, Mexico). (b) Backscattered scanning electron microscope image of andesite sample C8
447 (from Volcán de Colima). Both samples are characterised by a complex microstructure containing
448 both pores and microcracks (see text for details).

449



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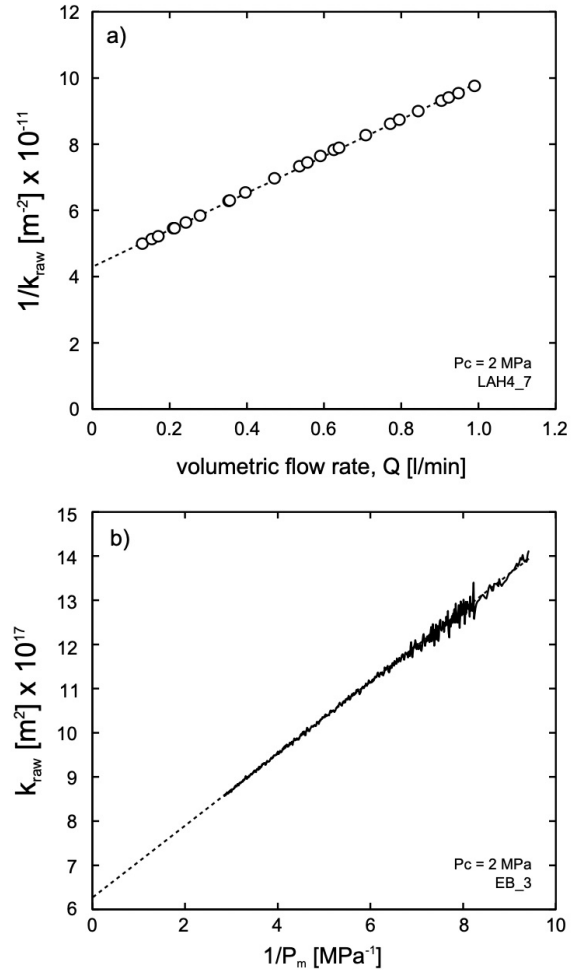
451 **Figure 2.** (a) Backscattered scanning electron microscope image of the basalt from Mt Etna (Italy)

452 showing a long microcrack that traverses the crystallised groundmass. (b) Backscattered scanning

453 electron microscope image of the basalt from Mt Etna showing a pocket of micropores sandwiched

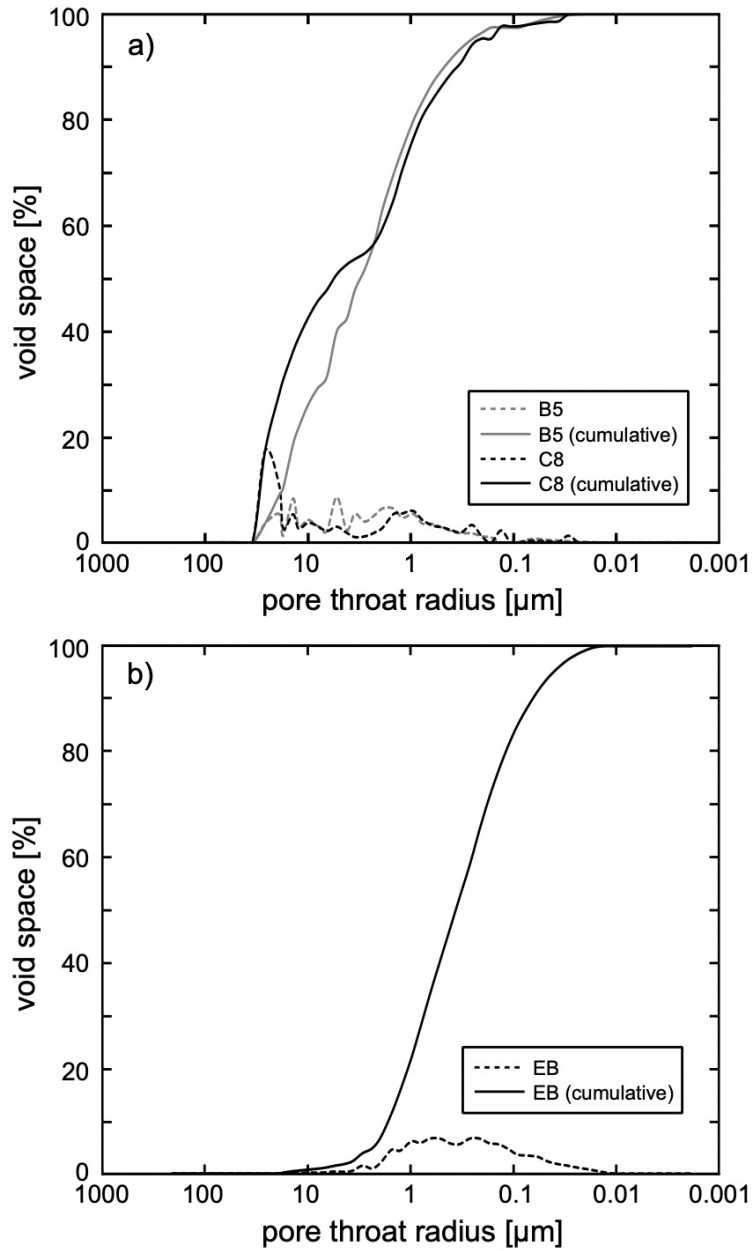
454 between two phenocrysts (see text for details).

455



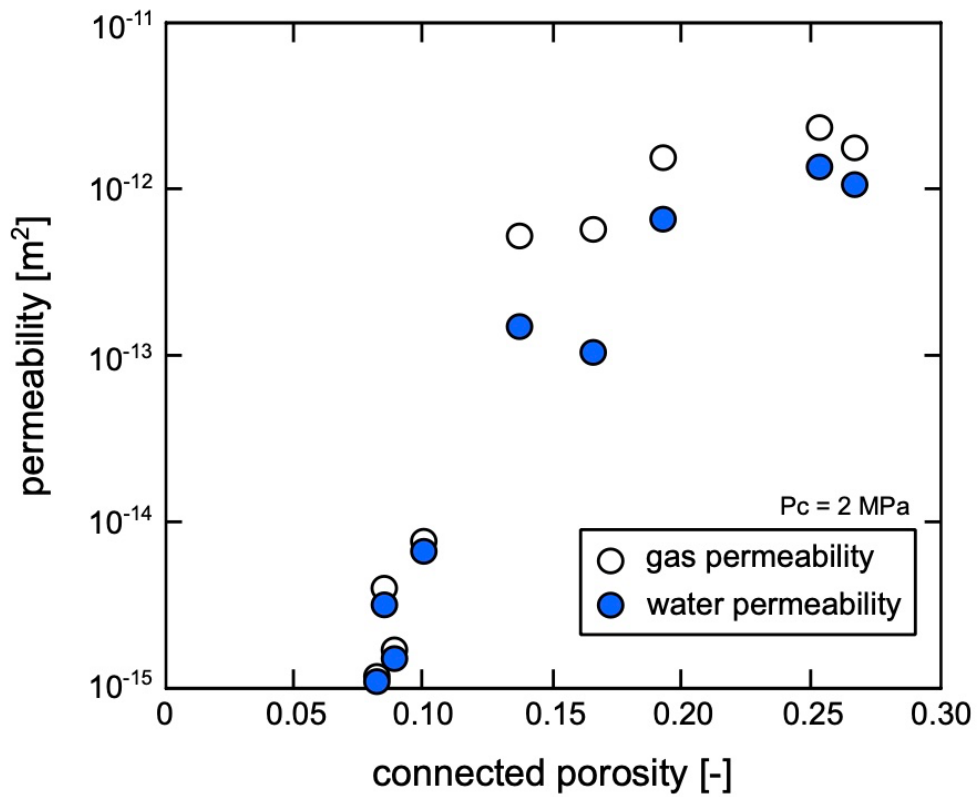
456

457 **Figure 3.** (a) Data that require a Forchheimer correction. The graph is a plot of $1/k_{gas_raw}$ as a
 458 function of volumetric flow rate, Q . The Forchheimer correction is necessary if the data can be well
 459 described by a positive linear slope. The Forchheimer-corrected permeability is taken as the inverse
 460 of the y -intercept of the best-fit linear regression. In the example shown here, the permeability is 2.33
 461 $\times 10^{-12} \text{ m}^2$ (Table 1). (b) Data that require a Klinkenberg correction. The graph is a plot of k_{gas_raw} as
 462 a function of the reciprocal mean pressure $1/P_m$, where P_m is the mean pore fluid pressure (i.e. $(P_{up} +$
 463 $P_{down})/2$). The Klinkenberg correction is required if the data can be well described by a positive
 464 linear slope and, if true, the Klinkenberg-corrected permeability is taken as the y -intercept of the
 465 best-fit linear regression. In the example shown here, the permeability is $6.29 \times 10^{-17} \text{ m}^2$ (Table 1).



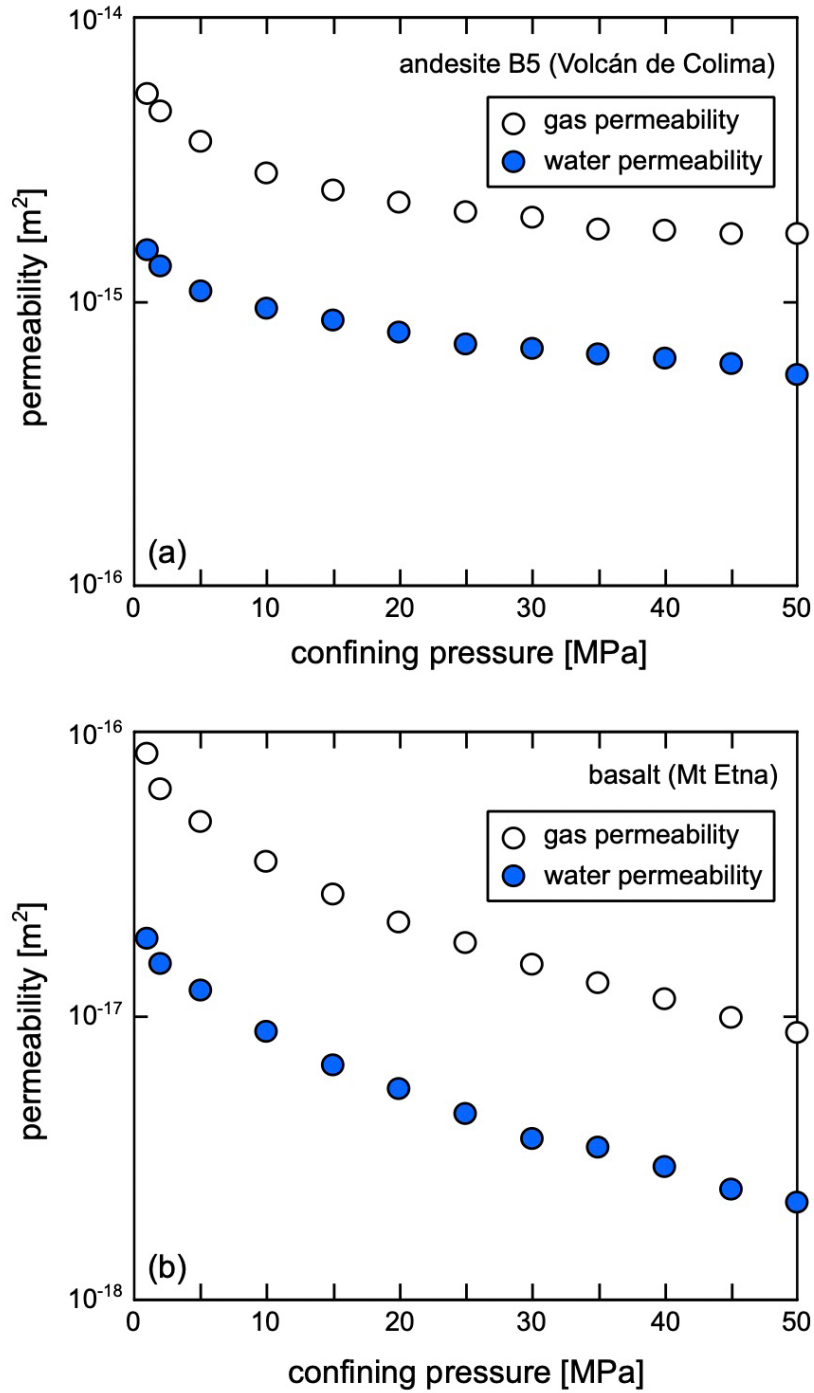
466

467 **Figure 4.** (a) Pore throat radius distributions, determined by mercury injection, for andesite samples
 468 B5 and C8 (from Volcán de Colima, Mexico). (b) Pore throat radius distribution, determined by
 469 mercury injection, for the basalt from Mt Etna (Italy). Dashed curves show the void space and the
 470 solid curves show the cumulative void space. The mercury injection data was corrected for the “low
 471 pressure correction” recommended by ASTM International [ASTM D4404-10].



472

473 **Figure 5.** Gas (white circles) and water (blue circles) permeability as a function of connected porosity
 474 for a suite of andesites from Volcán de Colima (Mexico). All measurements were performed under a
 475 confining pressure of 2 MPa. Error due to transducer precision is smaller than the symbol size. Data
 476 available in Table 1.



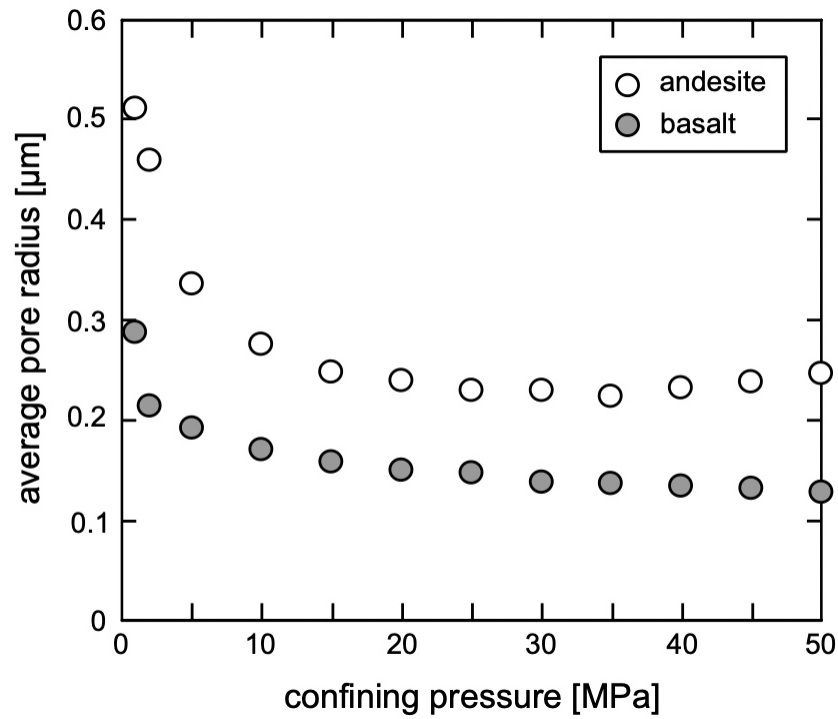
477

478 **Figure 6.** Gas (white circles) and water (blue circles) permeability as a function of confining pressure

479 (from 1 to 50 MPa) for (a) andesite sample B5 (from Volcán de Colima, Mexico) and (b) the basalt

480 from Mt Etna (Italy). Error due to transducer precision is smaller than the symbol size. Data available

481 in Table 1.



482

483 **Figure 7.** Average pore radius for andesite sample B5 (from Volcán de Colima, Mexico) (white circles)
 484 and the basalt from Mt Etna (Italy) (grey circles) as a function of confining pressure (from 1 to 50
 485 MPa), as calculated using the Klinkenberg slip factor (see Equation 5 and text for details). Data
 486 available in Table 1.

487

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698 **Table 1.** Summary of the experimental data collected for this study. Steady-state permeabilities were collected under a pressure gradient
699 of 0.5 MPa. The pressure gradient at the start of the pulse decay measurements was 0.5 MPa. *Specific surface area of this sample was not
700 measured and is assumed here to be equal to that of sample B5_5. **Specific surface areas of these samples were not measured and are
701 assumed here to be equal to that of another measured sample (EB_7).

702

Sample	Connected porosity	Specific surface area (m ² kg ⁻¹)	Method	Confining pressure (MPa)	Permeability to water (m ²)	Permeability to gas (m ²)	Permeability to gas / permeability to water	Klinkenberg slip factor, <i>b</i> (MPa)	Average pore diameter determined using <i>b</i> (μm)
B4_2	0.082	26	steady-state	2	1.09 × 10 ⁻¹⁵	1.17 × 10 ⁻¹⁵	1.07	0.503	0.054
B5_5	0.085	100	steady-state	2	3.13 × 10 ⁻¹⁵	3.97 × 10 ⁻¹⁵	1.27	0.069	0.396
A5_11	0.089	15	steady-state	2	1.49 × 10 ⁻¹⁵	1.69 × 10 ⁻¹⁵	1.13	0.204	0.134
A5_1	0.100	16	steady-state	2	6.55 × 10 ⁻¹⁵	7.61 × 10 ⁻¹⁵	1.16	0.134	0.204
A5_7	0.137	20	steady-state	2	1.48 × 10 ⁻¹³	5.14 × 10 ⁻¹³	3.47	Forchheimer	-
C8_9	0.166	35	steady-state	2	1.04 × 10 ⁻¹³	5.68 × 10 ⁻¹³	5.46	Forchheimer	-
C8_8	0.193	28	steady-state	2	6.51 × 10 ⁻¹³	1.52 × 10 ⁻¹²	2.33	Forchheimer	-
LAH4_7	0.253	51	steady-state	2	1.34 × 10 ⁻¹²	2.33 × 10 ⁻¹²	1.74	Forchheimer	-
LAH4_9	0.267	57	steady-state	2	1.05 × 10 ⁻¹²	1.76 × 10 ⁻¹²	1.68	Forchheimer	-
B5_5	0.081	100	steady-state	1	-	5.36 × 10 ⁻¹⁵	-	0.054	0.507
B5_5	0.081	100	steady-state	2	-	4.66 × 10 ⁻¹⁵	-	0.060	0.456
B5_5	0.081	100	steady-state	5	-	3.65 × 10 ⁻¹⁵	-	0.082	0.334
B5_5	0.081	100	steady-state	10	-	2.82 × 10 ⁻¹⁵	-	0.100	0.274
B5_5	0.081	100	steady-state	15	-	2.45 × 10 ⁻¹⁵	-	0.111	0.246
B5_5	0.081	100	steady-state	20	-	2.23 × 10 ⁻¹⁵	-	0.115	0.238
B5_5	0.081	100	steady-state	25	-	2.06 × 10 ⁻¹⁵	-	0.120	0.228
B5_5	0.081	100	steady-state	30	-	1.97 × 10 ⁻¹⁵	-	0.120	0.228
B5_5	0.081	100	steady-state	35	-	1.79 × 10 ⁻¹⁵	-	0.123	0.222
B5_5	0.081	100	steady-state	40	-	1.77 × 10 ⁻¹⁵	-	0.119	0.230
B5_5	0.081	100	steady-state	45	-	1.73 × 10 ⁻¹⁵	-	0.116	0.236
B5_5	0.081	100	steady-state	50	-	1.73 × 10 ⁻¹⁵	-	0.112	0.244
B5_21	0.085	100*	steady-state	1	1.51 × 10 ⁻¹⁵	-	-	-	-
B5_21	0.085	100*	steady-state	2	1.33 × 10 ⁻¹⁵	-	-	-	-
B5_21	0.085	100*	steady-state	5	1.09 × 10 ⁻¹⁵	-	-	-	-
B5_21	0.085	100*	steady-state	10	9.47 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	15	8.55 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	20	7.82 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	25	7.07 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	30	6.82 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	35	6.53 × 10 ⁻¹⁶	-	-	-	-
B5_21	0.085	100*	steady-state	40	6.30 × 10 ⁻¹⁶	-	-	-	-

B5_21	0.085	100*	steady-state	45	6.02×10^{-16}	-	-	-	-
B5_21	0.085	100*	steady-state	50	5.52×10^{-16}	-	-	-	-
EB_3	0.041	126**	pulse-decay	1	-	8.40×10^{-17}	-	0.096	0.285
EB_3	0.041	126**	pulse-decay	2	-	6.29×10^{-17}	-	0.129	0.212
EB_3	0.041	126**	pulse-decay	5	-	4.83×10^{-17}	-	0.143	0.191
EB_3	0.041	126**	pulse-decay	10	-	3.49×10^{-17}	-	0.162	0.169
EB_3	0.041	126**	pulse-decay	15	-	2.69×10^{-17}	-	0.174	0.157
EB_3	0.041	126**	pulse-decay	20	-	2.14×10^{-17}	-	0.184	0.149
EB_3	0.041	126**	pulse-decay	25	-	1.81×10^{-17}	-	0.187	0.146
EB_3	0.041	126**	pulse-decay	30	-	1.51×10^{-17}	-	0.199	0.137
EB_3	0.041	126**	pulse-decay	35	-	1.31×10^{-17}	-	0.201	0.136
EB_3	0.041	126**	pulse-decay	40	-	1.15×10^{-17}	-	0.206	0.133
EB_3	0.041	126**	pulse-decay	45	-	9.89×10^{-18}	-	0.208	0.131
EB_3	0.041	126**	pulse-decay	50	-	8.73×10^{-18}	-	0.214	0.128
EB_5	0.041	126**	steady-state	1	1.87×10^{-17}	-	-	-	-
EB_5	0.041	126**	steady-state	2	1.53×10^{-17}	-	-	-	-
EB_5	0.041	126**	steady-state	5	1.23×10^{-17}	-	-	-	-
EB_5	0.041	126**	steady-state	10	8.82×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	15	6.70×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	20	5.55×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	25	4.51×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	30	3.70×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	35	3.44×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	40	2.94×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	45	2.46×10^{-18}	-	-	-	-
EB_5	0.041	126**	steady-state	50	2.20×10^{-18}	-	-	-	-

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