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► **To cite this version:**

Etienne Médard, Jean-Luc Le Pennec. Petrologic imaging of the magma reservoirs that feed large silicic eruptions. *Lithos*, 2022, 428-429, pp.106812. 10.1016/j.lithos.2022.106812 . insu-03849589

**HAL Id: insu-03849589**

**<https://insu.hal.science/insu-03849589>**

Submitted on 11 Nov 2022

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1 **Petrologic imaging of the magma reservoirs that feed**

2 **large silicic eruptions**

3

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5

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11

12

13 **ABSTRACT**

14

15 Large silicic explosive eruptions are one of Nature's most hazardous phenomena.  
16 Very few have been witnessed and unravelling the complex conditions that lead to these  
17 eruptions remains a difficult task because the characteristics of the feeding magma  
18 reservoirs are still insufficiently constrained from geophysical imaging. Here we show that  
19 a barometer based on the composition of amphibole in equilibrium with plagioclase and  
20 biotite, common minerals in the eruptive products of large silicic eruptions, records the  
21 thickness and depth of magma reservoirs with uncertainties of 0.8 and 2.7 km, respectively.  
22 Pressures are given by the equation:  $P \text{ (MPa)} = 892 \cdot {}^{\text{VI}}\text{Al} + 101$ , where  ${}^{\text{VI}}\text{Al}$  is the  
23 octahedral aluminum content of the amphibole. With the example of a Miocene Turkish  
24 ignimbrite, we show that reservoirs feeding large silicic eruptions can be pancake-shaped.  
25 Our new barometer, valid between 650 and 950 °C, can be used in combination with  
26 volcanological and geophysical data to infer the size, shape and depth of magma reservoirs  
27 and may serve as a tool for monitoring future activity. This temperature-independent  
28 barometer is also applicable to any volcanic or plutonic rock containing amphibole +  
29 plagioclase + biotite and is in excellent agreement with previously published temperature-  
30 dependent barometers within their calibration range.

31

32 **1. Introduction**

33

34 Large silicic eruptions can bury large areas (thousands of km<sup>2</sup>) beneath thick tephra  
35 covers around their source (e.g., Miller and Wark, 2008) and may have a pronounced

36 impact on global atmospheric circulation and climate (e.g., Self and Blake, 2008). None has  
37 been observed in detail by volcanologists, but geophysical monitoring signals suggest new  
38 large silicic eruptions could occur in the geologically near-future (e.g., Laguna del Maule in  
39 Chile, Le Mével et al., 2016; Campi Flegrei in Italy, Kilburn et al., 2017; Long Valley and  
40 Yellowstone in the USA, Lowenstern et al., 2006; Seccia et al., 2011; Taupo in New  
41 Zealand, Heise et al., 2010), thus prompting scientists to better understand how these  
42 eruptions get prepared at depth. Geophysical signals resulting from magma motion in the  
43 crust are often used to monitor unrest and warn of upcoming eruptions. However, there is  
44 little information on reservoirs' characteristics and precursor signals of large silicic  
45 eruptions. For instance, the largest closely monitored eruption, the 1991 VEI 6 eruption of  
46 Mt Pinatubo, only erupted  $\sim 10 \text{ km}^3$  of magma (Newhall and Punongbayan, 1996). Larger  
47 eruptions with erupted bulk volumes of 100's  $\text{km}^3$  are common in the geological record  
48 (Miller and Wark, 2008), but we lack constraints on the behavior of large silicic feeding  
49 system to uniquely interpret the geophysical information (Lowenstern et al., 2006;  
50 Pritchard and Gregg, 2016). The depth and shape of magmatic reservoirs can be  
51 investigated through mineral-based barometry (Putirka, 2008), with the most accurate  
52 models relying on the composition of amphibole, a common mineral in pyroclastic rocks  
53 from large silicic eruptions (e.g., Johnson and Rutherford, 1989). Here we present a new  
54 amphibole barometer that yields accurate results of reservoir depth and shape, with further  
55 constraints on size when combined with tephra/magma volume estimates.

56 Al-in amphibole barometry relates the aluminum content of amphibole crystals to  
57 their pressure of crystallization that correlates directly to crystallization depth. The  
58 barometric potential of the Al concentration in amphibole was first identified by comparing

59 amphibole compositions in plutonic intrusives to pressure derived from thermobarometric  
60 study of their metamorphic contact aureoles (Hammarstrom and Zen, 1986; Hollister et al.,  
61 1987), before being calibrated experimentally (Johnson and Rutherford, 1989; Schmidt,  
62 1992). Calibrations are, in theory, only applicable to rocks containing the equilibrium  
63 assemblage amphibole + biotite + quartz + plagioclase + sanidine + magnetite + titanite or  
64 ilmenite. However, the absence of some of these phases results only in small additional  
65 uncertainties, and any silicic magmatic system containing equilibrium amphibole + biotite  
66 + plagioclase, and reasonably close to saturation with quartz and sanidine, can be  
67 constrained by amphibole barometry (Johnson and Rutherford, 1989).

68 Amphiboles are silicate minerals with a large range of possible substitutions  
69 (Hawthorne et al., 2012). The simplest formula is that of tremolite,  
70  $\square\text{Ca}_2\text{Mg}_5^{\text{VI}}\text{Si}_8\text{O}_{22}(\text{OH})_2$ . It contains no trivalent cation thus aluminum ( $\text{Al}^{3+}$ ) can only be  
71 incorporated through heterovalent substitutions, either in the octahedrally coordinated ( $^{\text{VI}}$ )  
72 site or the tetrahedrally coordinated ( $^{\text{IV}}$ ) site. Three major substitutions have been described  
73 in calc-alkaline silicic rocks:

74 (1) the tschermak substitution:  $^{\text{VI}}\text{Mg} + ^{\text{IV}}\text{Si} = ^{\text{VI}}\text{Al} + ^{\text{IV}}\text{Al}$

75 (2) the edenite substitution:  $\square + ^{\text{IV}}\text{Si} = \text{Na} + ^{\text{IV}}\text{Al}$

76 (3) the Ti-tschermak substitution:  $^{\text{VI}}\text{Mg} + 2^{\text{IV}}\text{Si} = ^{\text{VI}}\text{Ti} + 2^{\text{IV}}\text{Al}$

77 The tschermak substitution is pressure-dependent (Schmidt, 1992), whereas the edenite and  
78 Ti-tschermak substitutions are temperature-dependent (Bachmann and Dungan, 2002;  
79 Holland and Blundy, 1994), so the variations in total Al are controlled by both pressure and  
80 temperature. Barometers based on total Al in amphibole are thus only valid when  
81 temperature is fixed - either on an isotherm (Johnson and Rutherford, 1989) or along the

82 granite solidus (Schmidt, 1992). However, the pressure-dependent tschermak substitution is  
83 the only one to involve substitution of aluminum in the octahedral sites of the amphibole  
84 structure. By calculating structural formulae, the tschermak substitution can be dissociated  
85 from other substitutions by considering only octahedral aluminum <sup>VI</sup>Al, allowing for the  
86 construction of a temperature-independent barometer. Schmidt (1992) noticed the excellent  
87 correlation between <sup>VI</sup>Al and pressure, and attributed it to the tschermak substitution, but  
88 preferred to use total Al for his calibration. Two attempts have been made at calibrating a  
89 barometer based on <sup>VI</sup>Al versus pressure (Larocque and Canil 2010, Krawczynski et al.  
90 2012) but are restricted to high-Mg rocks and have not gained wide acceptance in the  
91 community.

92 In this paper, we use published analyses of experimental amphiboles to calibrate a  
93 new temperature-independent amphibole-barometer based on the octahedral aluminum  
94 content. Combination of this new barometer with volume estimate for an ignimbritic  
95 eruption allow to constrain the depth and shape of a large silicic magma reservoir. We show  
96 that this method is a priori applicable to other large silicic systems and may be combined  
97 with possible geophysical precursor signals at potentially reactivating silicic volcanoes to  
98 better constrain their pre-eruptive evolution.

99

## 100 **2. Methods**

### 101 *2.1. Barometer calibration database*

102 A total of 47 amphibole compositions from experiments on rhyolitic and dacitic bulk  
103 compositions were compiled from the literature (Johnson and Rutherford, 1989; Mutch et  
104 al., 2016; Rutter et al., 1989; Schmidt, 1992; Thomas and Ernst, 1990; Supplementary Data

105 1). All amphiboles are in equilibrium with biotite, plagioclase, magnetite and a rhyolitic  
106 silicate melt. Most amphiboles are in equilibrium with quartz and sanidine. We also  
107 included a few experiments that did not contain quartz and/or sanidine, but in which the  
108 silicate melt is SiO<sub>2</sub>- (> 75 wt%) and K<sub>2</sub>O- (> 5 wt%) rich, indicating that it is close to  
109 saturation with both minerals. Four amphibole compositions were excluded from the  
110 database because they showed large deviations from compositional trends. The first was  
111 extracted from a set of reversal experiments (Thomas and Ernst, 1990). The excluded  
112 reversal composition differs strongly from the corresponding forward experiment and does  
113 not plot on the linear trend defined by all other experiments. In two piston-cylinder  
114 experiments from the most recent study (Mutch et al., 2016), <sup>VI</sup>Al concentrations in  
115 amphibole are half the concentrations analysed in all other studies at similar pressures  
116 (Johnson and Rutherford, 1989; Rutter et al., 1989; Schmidt, 1992; Thomas and Ernst,  
117 1990). Amphibole crystals in these experiments are zoned (Mutch et al., 2016), a possible  
118 indication that equilibrium has not been reached. For consistency, we discarded all three  
119 piston-cylinder experiments from this study. The final dataset comprises 43 amphibole  
120 compositions and covers a pressure range of 80 to 1,300 MPa, and experimental  
121 temperatures from 650 to 850 °C, with oxygen fugacities mostly constrained near the NNO  
122 buffer (Supplementary Data 1).

123

## 124 2.2. *Samples and sample preparation*

125 Three pumice samples were collected from the Kızılıkaya ignimbrite, in the  
126 Cappadocia area of Central Turkey. Sample KZ-041010-4a (UTM zone 36S; X = 671344;  
127 Y = 4234915; Z = 1380 m) is from the Plinian fall deposit exposed below the main

128 ignimbrite sheet. Samples KZ-021010-1 (UTM zone 36S; X = 589770; Y = 4225687; Z =  
129 1068 m) and KZ-021010-2 (UTM zone 36S; X = 588204; Y = 4220509; Z = 1102 m) are  
130 from different levels of the main flow unit of the ignimbrite. Each sample was crushed and  
131 sieved, and heavy minerals were separated into fractions using a Wilfley table, a magnetic  
132 separator, and heavy liquids. Euhedral amphibole and plagioclase crystals were then hand-  
133 picked, mounted in epoxy and polished. Samples of pumice from the three sites were also  
134 mounted in epoxy resin and polished. A large pumice sample was collected from the  
135 Paipatja pyroclastic density current deposit unit of the most recent ( $218 \pm 14$   $^{14}\text{C}$  years BP)  
136 eruption of Tutupaca volcano in Peru (UTM zone 19K; X = 362215; Y = 8119045; Z =  
137 4680 m; Manrique et al., 2020; Samaniego et al., 2015). A zoned amphibole crystal and a  
138 neighbouring zoned plagioclase crystal were extracted from the pumice and mounted in  
139 Epoxy.

140

### 141 *2.3. Electron microprobe analyses*

142 Amphibole and plagioclase crystals were analysed using a SX-100 electron  
143 microprobe at Laboratoire Magmas et Volcans (Université Clermont Auvergne, Clermont-  
144 Ferrand, France), using a 15 kV acceleration voltage, a 40 nA beam current, and 5  $\mu\text{m}$   
145 diameter beam. The beam current is higher than commonly used for major element  
146 analyses, with the goal of reducing analytical uncertainties and thus improving the  
147 precision of pressure estimates. We analysed a total of 188 amphibole phenocrysts rims  
148 from the three samples of the Kızılkaya ignimbrite, as well as plagioclase rims from the  
149 same samples (Supplementary Data 2). Standards used for calibration include natural  
150 wollastonite (Si, Ca), orthoclase (Al, K), and albite (Na), and synthetic  $\text{TiMnO}_3$  (Ti, Mn),

151 fayalite (Fe), forsterite (Mg), and Cr<sub>2</sub>O<sub>3</sub> (Cr). All investigated phenocrysts show euhedral  
152 shapes and are homogeneous, i.e., with no significant zoning. Amphibole and plagioclase  
153 compositions from the Tutupaca sample were analysed with the same procedure. We also  
154 analysed glasses from pumices of the Kızılkaaya ignimbrite, using an enlarged beam size (20  
155 µm diameter), and decreased beam current (5 nA) to avoid Na-loss. We used the same  
156 standards than for mineral analyses, with the addition of natural fluorapatite for P.  
157 Analytical results, analytical uncertainties, calculated structural formulae, and calculated  
158 pressures are presented in Supplementary Data 2.

159

#### 160 *2.4. Raman spectrometry*

161 Water concentrations were analysed by Raman spectroscopy in a series of melt  
162 inclusions trapped in amphibole crystals from the Kızılkaaya ignimbrite. These  
163 measurements are then used to calculate volatile-saturation pressures and compare them  
164 with mineral barometry. Raman spectra were collected using a Renishaw InVia confocal  
165 Raman micro-spectrometer with a 532 nm wavelength. Detailed analytical conditions are  
166 presented in Schiavi et al. (2018). Reference glasses used for calibration include a series of  
167 in-house rhyolitic glasses. We used an internal calibration based on the correlation between  
168 water content and the area ratio of the water band and the alumino-silicate band (Schiavi et  
169 al., 2018).

170

#### 171 *2.5. Amphibole-plagioclase thermometry*

172 Pre-eruptive temperatures for the Kızılkaaya ignimbrite were estimated using  
173 amphibole-plagioclase thermometry. All three samples contain quartz phenocrysts, so we

174 can use thermometry based on the equilibrium edenite + quartz = tremolite + albite  
175 (Holland and Blundy, 1994). Amphibole and plagioclase rims from the Kızılkaya  
176 ignimbrite equilibrated at  $728 \pm 13$  °C.

177

### 178 **3. Results: temperature-independent amphibole barometer**

#### 179 *3.1. Barometer calibration*

180 We calculated structural formulae from published experimental amphibole  
181 compositions in equilibrium with biotite, plagioclase, magnetite, quartz and sanidine at  
182 650-850 °C (Supplementary Data 1). Correlations between total Al and pressure and  
183 between octahedral Al and pressure are presented in Figure 1. Correlations between total Al  
184 and pressure are usually excellent within a given dataset ( $R^2 = 0.99$ , Johnson and  
185 Rutherford 1989; Schmidt 1992, Figure 1a). However, these datasets were acquired over a  
186 restricted temperature range (750-780 °C for Johnson and Rutherford 1989, 655-700 °C for  
187 Schmidt 1992). When comparing datasets at different temperatures, the offset between  
188 calibrations becomes significant (145 MPa between the calibrations of Johnson and  
189 Rutherford 1989 and Schmidt 1992), confirming that total Al depends not only on pressure  
190 but also on temperature, and that calibrations based on total Al can only be used at  
191 temperatures close to their calibration temperatures. Individual correlations are usually  
192 linear, except for the very-low pressure set of data ( $\leq 200$  MPa) published by Mutch et al.  
193 (2016) which shows some non-linearity. This non-linearity is, however, purely due to  $^{IV}Al$   
194 and is not visible in the  $^{VI}Al$  versus pressure plot (Figure 1b), thus is likely due to  
195 temperature effects.

196  $^{\text{VI}}\text{Al}$  shows a very good linear correlation with pressure, independently of  
197 experimental temperature ( $R^2 = 0.939$ , Figure 1b):

198

$$199 \quad P \text{ (MPa)} = 892 \cdot ^{\text{VI}}\text{Al} + 101 \quad (1)$$

200

201 For the entire calibration range, the standard error of estimate (SEE = square root of  
202 average squared value of the difference between calculated and experimental pressures),  
203 i.e., the accuracy of the barometer, is 86 MPa (Supplementary Data 1).

204

### 205 *3.2. Precision and accuracy*

206 Pressure estimates are affected by calibration and analytical uncertainties. Analytical  
207 uncertainties control the precision of the pressure estimate. For barometers relying solely  
208 on the aluminum content of the amphibole, those uncertainties are negligible (around 8  
209 MPa) and only depend on the precision of aluminum measurements. Since our new  
210 barometer uses octahedral aluminum, other elements have an influence on structural  
211 formula calculations, resulting in a lower precision. We propagated analytical errors using a  
212 Monte-Carlo approach, by randomly generating a thousand synthetic analyses following a  
213 gaussian distribution around the measured value for each analyzed element, computing the  
214 resulting pressure for each of the thousand analyses, and calculating the resulting standard  
215 deviation. Regular analytical conditions used in our lab (15 kV, 15 nA, 10 s counting time)  
216 produce a  $2\sigma$  precision of 36 MPa. This value can be decreased to at least 20 MPa by  
217 increasing beam current and counting time (Figure 2). Most of the analyses presented in  
218 this paper were performed using a 40 nA beam current and a 30 s counting time, resulting  
219 in a 21 MPa precision (at the  $2\sigma$  level). For magmatic systems of rhyolitic to dacitic

220 compositions located in the upper crust, and assuming a granitic crust with a density of  
221  $2700 \text{ kg}\cdot\text{m}^{-3}$ , the depth precision of our barometer is thus 0.8 km.

222 Calibration uncertainties control the accuracy of the barometer. We estimated the  
223 standard error of estimate of our barometer using the 43 experimental amphiboles used for  
224 the calibration. The standard error of estimate (SEE), i.e., the accuracy of the barometer, for  
225 the entire dataset is 86 MPa. The main silicic reservoirs feeding super-volcanoes are,  
226 however, located in the upper crust (depth  $\leq 15$  km, Bachmann and Bergantz, 2008, Huber  
227 et al. 2019). Within the corresponding pressure range ( $\leq 400$  MPa), the experimental  
228 pressures are reproduced with an improved accuracy of 72 MPa. For upper crustal  
229 magmatic systems of rhyolitic to dacitic compositions, assuming a crustal density of  $2700$   
230  $\text{kg}\cdot\text{m}^{-3}$ , the depth accuracy of the barometer is thus 2.7 km. This accuracy is slightly higher  
231 than that reported for previous barometers based on total aluminum (50 MPa, Johnson and  
232 Rutherford 1989; 60 MPa, Schmidt et al. 1992; 50 MPa, Mutch et al. 2016). However,  
233 when comparing to our entire database, and not only each author's calibration database, the  
234 barometers are less accurate i.e. the SEE is higher: 133 MPa for Johnson and Rutherford  
235 (1989), 125 MPa for Schmidt (1992), 96 MPa for Mutch et al. 2016).

236

### 237 *3.3. Influence of temperature*

238 We tested the validity of our new barometer by analyzing a zoned amphibole crystal  
239 from the most recent eruption of Tutupaca volcano in Peru (Samaniego et al., 2015). The  
240 host rock is a dacitic pumice with phenocrysts of plagioclase, amphibole, biotite,  
241 clinopyroxene, titanite, magnetite, apatite and quartz. The investigated amphibole crystal  
242 records a sharp increase in temperature from  $750 \text{ }^\circ\text{C}$  in the core to  $950 \text{ }^\circ\text{C}$  in the rim

243 (Manrique et al., 2020, Figure 3, Supplementary Data 2), indicating that the magma  
244 reservoir was re-heated before crystallization of the mineral rims.

245 Crystallization pressures predicted for the core of the investigated amphibole crystal  
246 by previously published Al-in amphibole barometers (Johnson and Rutherford, 1989;  
247 Mutch et al., 2016; Ridolfi et al., 2010; Ridolfi and Renzulli, 2012; Rutter et al., 1989;  
248 Thomas and Ernst, 1990) varies from 126 to 386 MPa (Figure 3). These pressures are  
249 directly correlated to the temperature at which the barometer was calibrated (Figure 4),  
250 further highlighting the influence of temperature on aluminum concentration in amphibole.  
251 Barometers based on total aluminum concentration in amphiboles can thus only be used if  
252 the temperature is close to their average calibration temperature. All these barometers also  
253 predict a 120-256 MPa increase in pressure from core to rim, equivalent to a 4.5-9.7 km  
254 increase in depth (Figure 3). Such an increase is physically highly unlikely, and previous  
255 studies on similar material have shown that it is an artefact because these barometers do not  
256 differentiate the influence of temperature and pressure on aluminum concentration  
257 (Bachmann and Dungan, 2002; Rutherford and Devine, 2003; Shane and Smith, 2013).  
258 Although only one profile is reported in this paper, core (“high-Al”) + rim (“low-Al”)  
259 analyses have been performed on a large number of amphibole crystals from the same  
260 eruption by Manrique et al. (2020). Data reported in their Table 7 for all crystals confirm  
261 the results presented here, with a 120-270 MPa “pressure” increase between core and rim  
262 calculated by barometers relying on total Al, and only a 23 MPa difference recorder by our  
263 new barometer, well within uncertainties. Although amphibole-barometry is an extremely  
264 powerful tool, total aluminum concentrations should only be used after a thorough

265 investigation of crystal chemistry variations and with a good estimate of crystallization  
266 temperatures.

267 In the investigated crystal, the Al variation can be fully explained by temperature-  
268 dependent variations in <sup>IV</sup>Al through edenite and Ti-tschermak substitutions. Our new  
269 barometer produces no pressure variation throughout the amphibole crystal (Figure 3),  
270 despite the 200 °C temperature increase, evidencing that it is independent of temperature.  
271 Calculated core pressure (crystallization temperature 750 °C) is in excellent agreement with  
272 that calculated using a calibration based on total aluminum performed at an average  
273 temperature of 760 °C (Johnson and Rutherford, 1989). At high temperatures (950 °C for  
274 the rim), our model is consistent with a model calibrated on dacitic to basaltic rocks  
275 (Ridolfi and Renzulli, 2012) at an average temperature of 936 °C, despite using a different  
276 calibration database with no common data points (rhyolites to dacites at 650-850 °C vs  
277 dacites to basalts at 800-1130 °C). These results suggest that our model can be extrapolated  
278 to higher temperatures beyond its calibration range. To demonstrate its applicability at  
279 higher temperature, we tested our model against a recent independent set of experimental  
280 amphibole compositions in equilibrium with plagioclase and biotite at 900-950 °C (8  
281 datapoints from Rondet et al., 2019). Experimental pressures are reproduced with an  
282 excellent SEE of 41 MPa (Figure 5).

283 Since biotite is less common in silicic rocks than amphibole and plagioclase, we  
284 tested the applicability of our barometer to biotite-free rocks using a recent series of  
285 experiments on a biotite-free dacite from Quizapu, Chile (First et al. 2021). We selected  
286 fourteen experiments for which at least five different amphibole analyses were reported.  
287 These experiments were run between 75-175 MPa and 810-890 °C and contain both

288 amphibole and plagioclase, half of them biotite-free, half of them containing biotite. Our  
289 new barometer reproduces experimental pressure with an SEE of 74 MPa for biotite-  
290 bearing experiments (Figure 5, similar to the 72 MPa SEE of the calibration database).  
291 However, for biotite-free experiments, the calculated pressure is on average 146 MPa  
292 higher than experimental pressure (SEE of 144 MPa). Although this is only one example,  
293 and amphibole-barometry in biotite-free rocks requires further investigation, we do not  
294 recommend using our new barometer on biotite-free rocks, since this may result in  
295 overestimated calculated pressures.

296

#### 297 *3.4. Influence of oxygen fugacity*

298 The distribution of aluminum between tetrahedral and octahedral sites is sensitive to  
299 the way structural formulae are calculated, and in particular the proportions of ferric and  
300 ferrous iron. Most amphibole crystals in our experimental database have either been  
301 synthesized under controlled oxygen fugacity buffered by a mix of Ni and NiO (NNO  
302 buffer; Schmidt, 1992, and part of the data from Johnson and Rutherford, 1989) or under  
303 conditions demonstrated to be close to those of the NNO buffer (Mutch et al., 2016; Rutter  
304 et al., 1989; Thomas and Ernst, 1990). According to Clowe et al. (1988), calcic amphiboles  
305 equilibrated near the NNO buffer have  $\text{Fe}^{3+}/\Sigma\text{Fe} \sim 0.2$ . The structural formula of  
306 experimental amphiboles in the calibration database was thus calculated based on 46  
307 charges (23 oxygens) and assuming 20% of iron is ferric (Clowe et al., 1988), unless the  
308 sum of cations excluding  $\text{Na}^+$  and  $\text{K}^+$  exceeds 15. In that latter case, the ratio  $\text{Fe}^{3+}/\Sigma\text{Fe}$  is  
309 increased to satisfy stoichiometry criteria (Supplementary Data 1). This was only required  
310 for one amphibole composition where  $\text{Fe}^{3+}/\Sigma\text{Fe}$  was increased to 0.36. Varying the

311  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratio from 0.1 to 0.3 (Clowe et al., 1988; Schmidt, 1992) does not significantly  
312 change the results: it produces parallel calibrations, with pressure offsets of +30 MPa  
313 ( $\text{Fe}^{3+}/\Sigma\text{Fe} = 0.3$ ) to -25 MPa ( $\text{Fe}^{3+}/\Sigma\text{Fe} = 0.1$ ), well within the barometer uncertainty. Some  
314 of the experiments by Johnson and Rutherford (1989) are more oxidized (up to the MNO  
315 buffer), however, there is no systematic variations in octahedral Al content with  
316 experimental oxygen fugacity. Data from a recent study by Rondet et al. (2019) on  
317 amphiboles in equilibrium with plagioclase and biotite at oxygen fugacities between  
318 NNO+0.5 and NNO+2.1 also do not show any systematic variations in the predicted  
319 pressure as a function of oxygen fugacity. For applications of our new amphibole  
320 barometer, we thus recommend using the same procedure for structural formulae  
321 calculation, with known  $\text{Fe}^{3+}/\Sigma\text{Fe}$  when available. In the absence of independent constraints  
322 on  $\text{Fe}^{3+}/\Sigma\text{Fe}$ , we recommend using a  $\text{Fe}^{3+}/\Sigma\text{Fe}$  ratio of 0.2. If the investigated amphiboles  
323 are more oxidized, the pressure will be slightly underestimated by this approach.

324

#### 325 **4. Characterization of the magma reservoir that fed a large silicic eruption**

##### 326 *4.1. The Kızilkaya ignimbrite in Turkey*

327 The Kızilkaya ignimbrite is the product of a large eruption in the Cappadocia area of  
328 Central Turkey. This area superbly exposes a succession of Neogene dacitic to rhyolitic  
329 ignimbrites and fallout deposits, recording 10 Ma of magmatic activity (Aydar, 2012; Le  
330 Pennec et al., 1994; Viereck-Goette et al., 2010). The succession provides an excellent  
331 example of a long-lived silicic system, with ignimbrite eruptions at mid-to-low frequency  
332 in Mio-Pliocene times, and with a moderate average magma output rate (about  $10^{-3}$  km<sup>3</sup>/a),  
333 but large silicic eruptions (bulk volumes exceeding 300 and 180 km<sup>3</sup> for Cemilköy and

334 Kızılıkaya ignimbrite units, respectively). The Kızılıkaya ignimbrite is U-Pb dated at  $5.43 \pm$   
335  $0.06$  Ma (Paquette and Le Pennec, 2012) and covers an area of more than 10 000 km<sup>2</sup>.  
336 Juvenile pumice is rhyolitic (71-73 wt% SiO<sub>2</sub>), with phenocrysts of plagioclase, biotite,  
337 amphibole, magnetite, ilmenite, quartz, apatite and zircon (Temel et al., 1998). No  
338 significant variation in mineral or glass composition was observed over the three  
339 investigated samples. For example, the SiO<sub>2</sub> content in the glass is  $77.6 \pm 0.4$  (2 $\sigma$ ), a  
340 variation within the analytical uncertainty of the electron microprobe.

341

#### 342 *4.2. Average depth of magma storage*

343 A total of 188 amphibole phenocrysts were analyzed by electron microprobe to  
344 constrain pre-eruptive pressures and temperatures (Supplementary Data 2). All investigated  
345 crystals are euhedral and homogeneous. They are thus all juvenile phenocrysts, and  
346 amphibole xenocrysts and antecrysts, if present (Paquette and Le Pennec, 2012), were  
347 either not sampled, or dissolved prior to the eruption. Pre-eruptive temperature is  $728 \pm 13$   
348 °C, well within the calibration range of our new barometer (650-850 °C). The pressure  
349 distribution of all crystal rims (Figure 6a), calculated from their <sup>VI</sup>Al content, is close to  
350 Gaussian with an average value of 214 MPa and a standard deviation of 18 MPa. Assuming  
351 a crustal density of 2700 kg·m<sup>-3</sup>, the average depth of the magma chamber is  $8.1 \pm 2.7$  km.  
352 This value is within the range of typical eruptible rhyolitic reservoirs, as shown by  
353 petrological and geophysical methods (Huber et al. 2019 and references therein). It is also  
354 in good agreement with thermo-mechanical models that show an optimal depth for  
355 eruptible large silicic reservoirs around  $200 \pm 50$  MPa – at lower pressures, volatile  
356 exsolution hinders chamber growth whereas at higher pressure the low viscosity of the crust

357 inhibits eruption (Huber et al. 2019). We also analyzed water concentrations in primary  
358 rhyolitic melt inclusions trapped in amphibole crystals by Raman spectrometry (Table 1).  
359 The maximum H<sub>2</sub>O concentration is 4.95 wt%, well within the “optimal” 4.0-5.5 wt. %  
360 H<sub>2</sub>O concentration estimated by Poppa et al. (2021) to trigger explosive silicic eruptions.  
361 Assuming that the magma is saturated with a pure H<sub>2</sub>O volatile phase, the storage pressure  
362 is estimated at 170 MPa (Papale et al., 2006), slightly lower but well within uncertainty of  
363 the pressure calculated by amphibole barometry ( $214 \pm 72$  MPa). If the saturating fluid  
364 phase contained small amount of CO<sub>2</sub>, the entrapment pressure would be higher. Also,  
365 since the eruptible part of the magma body is likely slightly below fluid-saturation (e.g.,  
366 Poppa et al. 2021), this saturation pressure is expected to be lower than the storage  
367 pressure. Volatile saturation and Al-in-amphibole barometry are thus in good agreement  
368 with each other.  
369

crystal	OH area		Si area		OH/Si	H <sub>2</sub> O
	raw	normalized	Raw	normalized		
KZ0210-a2	746743	519	1264272	1317	0.39	3.71
KZ0210-a7	903204	627	1242755	1295	0.48	4.52
KZ0210-a8	733016	509	1301905	1356	0.38	3.55
KZ0210-a9	590354	410	1284089	1338	0.31	2.93
KZ0210-a10	643556	447	1193751	1243	0.36	3.41
KZ0210-a11	906644	630	1165255	1214	0.52	4.83
KZ0210-a13	899845	625	1127344	1174	0.53	4.95

370 Table 1: H<sub>2</sub>O concentrations in melt inclusions in amphiboles from sample KZ021010-1

371

### 372 4.3. Volume of erupted material

373 The bulk volume of the Kızılkaya ignimbrite has been estimated in a previous work  
374 to 180 km<sup>3</sup> (Le Pennec et al., 1994), and no refinements have been proposed since. In that

375 work, the authors reconstructed the approximate pre-erosion distribution of the ignimbrite  
376 by drawing the envelope of all known outcrops of the Kızılkaya unit (10 600 km<sup>2</sup>). The  
377 deposit shows a fairly constant thickness of 15-30 m on the Nevsehir Plateau, and local  
378 overthickening (up to 80 m) in valleys. The ignimbrite thins distally to 2-3 m, 120 km from  
379 the presumed source. To account for uneven welding intensity, these authors assigned a  
380 welding intensity at localities where the ignimbrite is exposed, which allowed recalculating  
381 the pre-(welding) compaction thickness to obtain a homogenous density of 1000 kg·m<sup>-3</sup>.  
382 The bulk volume of 180 km<sup>3</sup> obtained with this method is considered a minimum estimate  
383 because large fractions of the loose top of the ignimbrite were likely removed by erosion  
384 and underestimated in the calculation (this may represent 10 to 20 % of the total bulk  
385 volume of the ignimbrite, maybe more).

386 The volume of the tephra fall deposit was not estimated in previous studies, notably  
387 because the overlying ignimbrite incorporated a large amount of material from the  
388 underlying loose Plinian tephra fall layer. Thickness variations in the Kızılkaya fall layer  
389 are thus poorly correlated to distance from source and constructing a reliable isopach map  
390 proved difficult (Le Pennec et al., 1994). To obtain a minimum estimate of the tephra fall  
391 layer volume we selected the 30 cm isopach contour that, in our interpretation, is the most  
392 reliable non-eroded one (e.g. it is exposed nicely 44 km east of the source, along the main  
393 dispersal axis). The area inside the 30 cm isopach is about 691 km<sup>2</sup>. Using a single isopach  
394 approximation approach (Legros, 2000), we thus estimate the minimum bulk volume at 2.6  
395 km<sup>3</sup>, which is negligible when compared to that of the ignimbrite. Removing 15 vol.% of  
396 xenolithic material from the ignimbrite and the Plinian fall deposit yields bulk volumes of  
397 pure juvenile material of 153 km<sup>3</sup> and 2.2 km<sup>3</sup>, respectively. Using a density of 2400 kg·m<sup>-3</sup>

398 <sup>3</sup> for dense rhyolitic material implies a Dense Rock Equivalent (D.R.E.) volume of at least  
399 65 km<sup>3</sup> for the magma body that fed the Kızılkaya eruption (the additional volume of the  
400 intracaldera fill is unknown). This result is a minimal estimate and we surmise that the total  
401 D.R.E. volume is more likely in the range of 70-100 km<sup>3</sup>, the eruption of which typically  
402 produces collapse structures of 15-20 km in diameter (Geshi et al., 2014).

403

#### 404 *4.4. Shape of the erupted reservoir*

405       Given the extremely homogeneous composition and identical mineralogy of the three  
406 samples collected at three different sites, there is no evidence of magma mixing or zoning  
407 in the reservoir. The near-Gaussian pressure distribution (Figure 6a) is unlikely to be solely  
408 the result of a natural phenomenon. Instead, it can be readily explained by analytical  
409 uncertainties of the electron microprobe (which produce a Gaussian distribution) combined  
410 with an initial amphibole depth distribution in the magma chamber. Theoretical pressure  
411 distributions were modelled for different magma chamber thicknesses by (1) considering  
412 amphibole crystals originating at different depths in the eruptible magma body, with a  
413 homogeneous repartition with depth, (2) calculating a Gaussian pressure distribution for  
414 each crystal, using average pressure and the analytical standard deviation from the  
415 microprobe (21 MPa) and (3) summing the resulting pressure distributions for all the  
416 crystals (Figure 6a). We then calculated the residual sum of squares between the modelled  
417 and the observed pressure distributions, as a function of the magma chamber thickness  
418 (Figure 6a). The best fit is obtained for a magma chamber thickness of 1.1 km, equivalent  
419 to 30 MPa. When looking at pressure variations instead of absolute pressures, the precision

420 ( $2\sigma$ ) of the barometer is about 21 MPa (0.8 km). The inferred thickness of the erupted  
421 fraction of the magma chamber that fed the Kızılıkaya eruption is thus  $1.1 \pm 0.8$  km.

422 The minimum estimated volume for the Kızılıkaya ignimbrite and concomitant Plinian  
423 tephra fall deposit is  $65 \text{ km}^3$  D.R.E. of magma. Considering a sill-like magma body (Figure  
424 6b), with a thickness of  $1.1 \pm 0.8$  km, the estimated diameter of the magma reservoir is 7-17  
425 km. A caldera of larger diameter (at least on the order of 15-20 km) should be expected  
426 around the source of the Kızılıkaya eruption (Figure 6b), as collapse of the magma  
427 chamber's roof involves formation of inclined faults above it, which translate into a wider  
428 collapse structure at the surface (Roche et al., 2000). Volcanological (Le Pennec et al.,  
429 1994) and geophysical (Froger et al., 1998) investigations in the Cappadocia area have  
430 identified two caldera complexes formed by nested collapse structures. One northern  
431 collapse structure is about 15 km in diameter, while the southern, more complicated  
432 collapsed area is roughly 35 km long by 23 km wide. The size of the southern nested  
433 collapse system is consistent with the presence of a caldera similar to that formed above the  
434 Kızılıkaya magma reservoir, coalescent with other calderas of previous large silicic  
435 eruptions from the same system. The range of aspect ratios (thickness over diameter) for  
436 the reconstructed Kızılıkaya magma reservoir is between 0.02 and 0.3, supporting a pre-  
437 eruptive pancake-shaped magma body (Figure 6b). These aspect ratios are in excellent  
438 agreement with reservoir shapes inferred from geophysical studies and modelling of other  
439 silicic systems worldwide (Bachmann and Bergantz, 2008; Gregg et al., 2012; Wilson,  
440 2001).

441

## 442 **5. Application to future large silicic eruptions**

443       Based on the approach above, our new temperature-independent barometer is a  
444 promising tool for imaging silicic magma chambers since the silicic products of most large  
445 silicic eruptions contain amphibole crystals. Statistical analysis of large amphibole  
446 populations is much more accurate than random analyses of a limited number of crystals.  
447 This methodology can unfortunately only be used on past eruptions since only full eruption  
448 of the magma lenses can provide an image of their crystal cargo. However, many large  
449 silicic eruptions are preceded by small volume precursor eruptions (e.g., over 20 ka for the  
450 caldera-forming Crater Lake eruption, Bacon and Lanphere, 2006) that can be used to  
451 locate the active magma reservoirs and aid in interpretation of geophysical signals. For  
452 example, the Laguna del Maule volcanic complex in Chile has been proposed as a  
453 developing future large silicic eruption (Feigl et al., 2014; Singer et al., 2014). Small  
454 amphibole-bearing rhyodacitic domes have been emplaced in the area over the past 20 ka  
455 (Andersen et al., 2017). Using the amphibole compositions published by Andersen et al.,  
456 (2017), our barometer predicts that these small volumes of magma were initially stored at  
457 depths close to 5 km (110-140 MPa, Table 2), with one exception being stored deeper at ~8  
458 km (220 MPa). This value coincides with the deformation source estimated from INSAR  
459 data ( $5.2 \pm 0.3$  km, Feigl et al., 2014), and is within error of the  $200 \pm 50$  MPa optimal  
460 reservoir depth calculated by Huber et al. (2019). The pre-eruptive storage depth of  
461 potential small precursor eruptions may thus possibly be indicative of the uppermost part of  
462 the growing large silicic reservoir and serve as an indicator for the interpretation of  
463 seismological and other geophysical data. Note that those quick calculations are only  
464 presented here for illustration purposes, and a full investigation of magma storage depth at  
465 Laguna del Maule is beyond the scope of this study. Also, uncertainties associated with

466 using literature data might be higher than our stated 72 MPa uncertainty since we do not  
 467 know the analytical conditions used by Andersen et al. (2017).

468

dome name	rdcn	Rdcd	Rdne	Rdno	rdac
average pressure (MPa)	127	222	137	126	113
uncertainty, $2\sigma$ (MPa)	72	112	72	80	72
average depth (km)	4.8	8.4	5.2	4.8	4.3
uncertainty, $2\sigma$ (km)	2.7	4.2	2.7	3.0	2.7
age (ka)	3.5	8.0	>19	>19	20.0
uncertainty (ka)	2.3	0.8			1.2

469 Table 2. Estimated storage pressures and depths for five recent rhyodacite domes in the  
 470 Laguna del Maule area, calculated from the analytical data of (Andersen et al., 2017).  
 471 Depth estimations assume a  $2700 \text{ kg}\cdot\text{m}^{-3}$  density for the granitic crust. Reported ages are  
 472  $^{40}\text{Ar}/^{39}\text{Ar}$  ages from Andersen et al. (2017). Uncertainties are reported at the  $2\sigma$  level as  
 473 either data dispersion, indicating analytical errors or multiple crystal provenance, or model  
 474 accuracy ( $72 \text{ MPa} = 2.7 \text{ km}$ ), whichever is higher.

475

476 Combination of petrological and volcanological data thus provides refined constraints  
 477 on the shape of large silicic magma reservoirs and can serve as a guide for geophysical  
 478 monitoring of future eruptions, particularly in areas where signals suggest the Earth is  
 479 getting ready for a new large silicic eruption and pre-climactic amphibole-bearing products  
 480 are emitted (e.g., Heise et al., 2010; Le Mével et al., 2016; Seccia et al., 2011). Further  
 481 improvement of amphibole barometry, relying on crystal chemistry and thermodynamics  
 482 rather than empirical correlations, and grounded in high-quality experimental data is  
 483 desirable in order to better image past and active silicic volcanic systems.

484

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636

637 **Acknowledgements:** This research was supported by the MISTRAL/Termex program of  
638 CNRS/INSU through the “Volhexan” project. The Tutupaca sample was collected during  
639 fieldwork in Peru funded by the Institut de Recherche pour le Développement (IRD) and  
640 with help from Pablo Samaniego (IRD) and Marco Rivera (Instituto Geofísico del Perú).  
641 Claire Fonquernie and Jean-Louis Paquette helped with the preparation of the Kızılkaya  
642 samples. Federica Schiavi helped with the Raman analyses. This manuscript benefited from  
643 discussions with Audrey M. Martin, Justine E. Francomme, Pablo Samaniego, Abidin  
644 Temel, and Nélide Manrique, as well as constructive reviews by Olivier Bachmann and  
645 Jake Lowenstern and editorial handling by Michael Roden. This is Labex Clervolc  
646 contribution xxx.

647

## 648 **Figure Captions**

649

650 **Figure 1.** Total aluminum (a) and octahedral aluminum ( $^{VI}\text{Al}$ , b) per formula unit as a  
651 function of pressure. Pressure uncertainties are smaller than the symbols. Uncertainties for

652 total aluminum are 8 MPa, and for octahedral aluminum close to 36 MPa. Existing  
653 calibrations based on total aluminum are plotted for comparison in (a): Johnson and  
654 Rutherford (1989), red dashed line; Schmidt (1992), green dashed line; Mutch et al. (2016),  
655 black curve. Experimental data are from Johnson and Rutherford (1989), Mutch et al.  
656 (2016), Rutter et al. (1989), Schmidt (1992), Thomas and Ernst (1990). In (b), a least-  
657 squares linear regression through the data gives a correlation coefficient of 0.939 and a  
658 standard error of estimate of 86 MPa (72 MPa for data  $\leq$  400 MPa).

659

660 **Figure 2.** Microprobe analytical precision for our new barometer based on octahedral  
661 aluminum as a function of the product of beam current and counting time. By optimizing  
662 analytical conditions, the precision can be decreased to 10-11 MPa (at the  $1\sigma$  level).

663

664 **Figure 3.** a) SEM image of a zoned amphibole crystal from the Paipatja unit of the  $218 \pm$   
665 14 aBP eruption of Tutupaca volcano in Peru (Manrique et al., 2020). B) Calculated  
666 pressure profiles using existing temperature-dependent amphibole barometers (Johnson and  
667 Rutherford, 1989; Mutch et al., 2016; Ridolfi et al., 2010; Ridolfi and Renzulli, 2012;  
668 Rutter et al., 1989; Thomas and Ernst, 1990) and our new pressure-independent barometer.  
669 Note that the tentative calibrations of amphibole-melt barometers proposed by Putirka  
670 (2016) are not discussed here because melt compositions are mostly not available in our  
671 calibration database, and not often available in natural samples. The pressure profile shows  
672 a significant step at the core-rim boundaries for all previously published barometers. This  
673 step is an artefact linked to the increase in temperature (Bachmann and Dungan, 2002;  
674 Rutherford and Devine, 2003; Shane and Smith, 2013). The grey area represents the

675 average and  $2\sigma$  standard deviation of our data. Temperature on the right indicate the  
676 average experimental temperature in the calibration database for each model.

677

678 **Figure 4.** Calculated crystallization pressure for the core of the investigated amphibole  
679 crystal from the Paipatja unit of the  $218 \pm 14$  aBP eruption of Tutupaca volcano in Peru  
680 (Manrique et al., 2020) using published barometers as a function of the average calibration  
681 temperature of the barometer. There is a very good correlation between calibration  
682 temperature and calculated pressure, except for one barometer (Thomas and Ernst, 1990).  
683 The white circle represents the pressure estimated with our barometer and the estimated  
684 temperature of the crystal core (Manrique et al., 2020).

685

686 **Figure 5.** Calculated crystallization pressure as a function of experimental pressure for a  
687 series of experiments containing amphibole and plagioclase. Experiments from the  
688 calibration database contain amphibole, plagioclase, biotite, sanidine and quartz (or very  
689 close to saturation with sanidine and quartz, Johnson and Rutherford (1989), Mutch et al.  
690 (2016), Rutter et al. (1989), Schmidt (1992), Thomas and Ernst (1990)). Samples from  
691 Rondet et al. (2019) still contain amphibole, plagioclase and biotite, but are far from  
692 saturation with sanidine and quartz (trachy-andesitic composition). Experiments from First  
693 et al. (2021) were run on a dacitic composition and contain amphibole and plagioclase with  
694 (green circles with black rim) and without (green circles with green rim) biotite. Our new  
695 barometer reproduces well experimental pressure for samples containing at least  
696 amphibole, plagioclase and biotite.

697

698

699 **Figure 6.** a) Pressure distribution for the 188 investigated amphibole crystals. The  
700 distribution is close to a Gaussian one, with an average value of 214 MPa and a  $1\sigma$  standard  
701 deviation of 18 MPa. Also plotted are modelled pressure distributions calculated for various  
702 magma chamber equivalent thicknesses: 60 MPa (flat top curve); 30 MPa (best fit to the  
703 data, solid line); and 0 MPa (theoretical infinitely flat chamber). The inset shows the  
704 residual sum of squares between modelled and observed pressure distributions as a function  
705 of magma chamber equivalent thickness. The data are fitted by a fourth-order polynomial  
706 with a minimum value at  $\sim 30$  MPa. b) Reconstructed pre-eruptive shape of the magma  
707 chamber and post-eruptive feature, together with a schematic interpretation of magma  
708 reservoirs (redrawn from Bachmann and Bergantz, 2008).

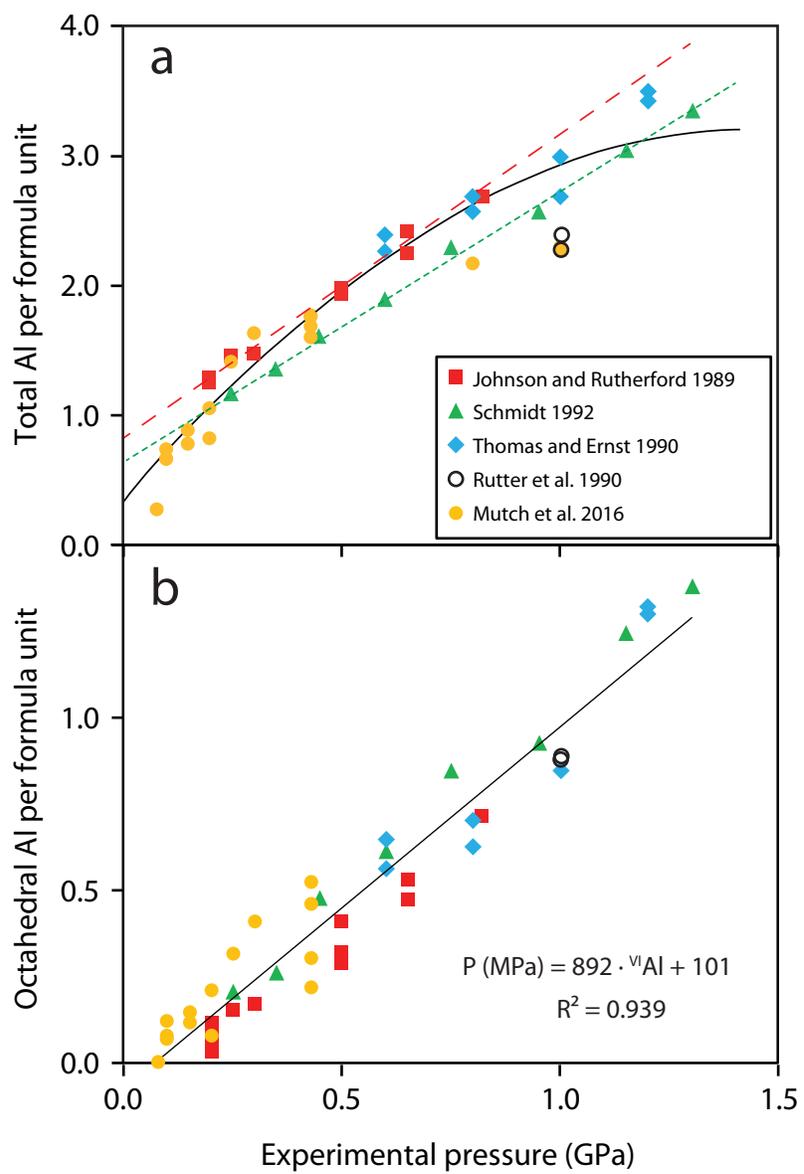


Figure 1

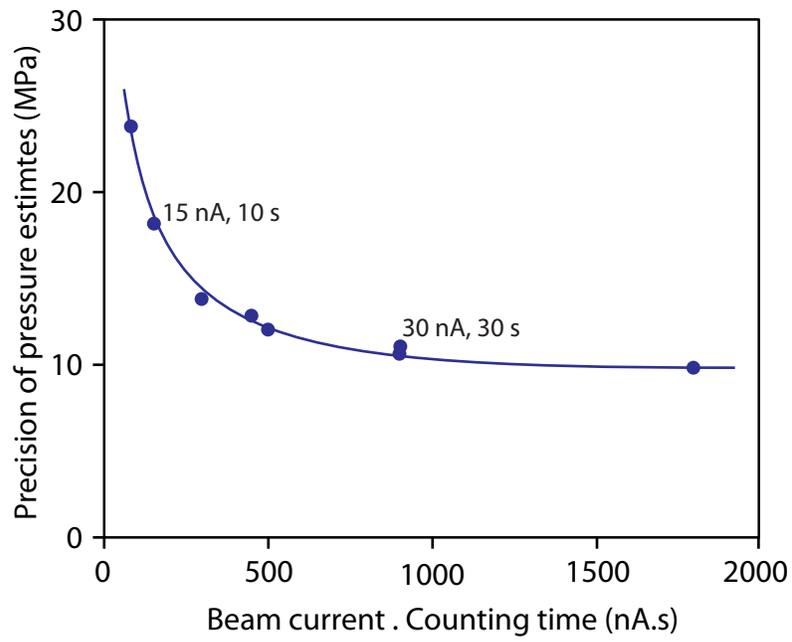


Figure 2

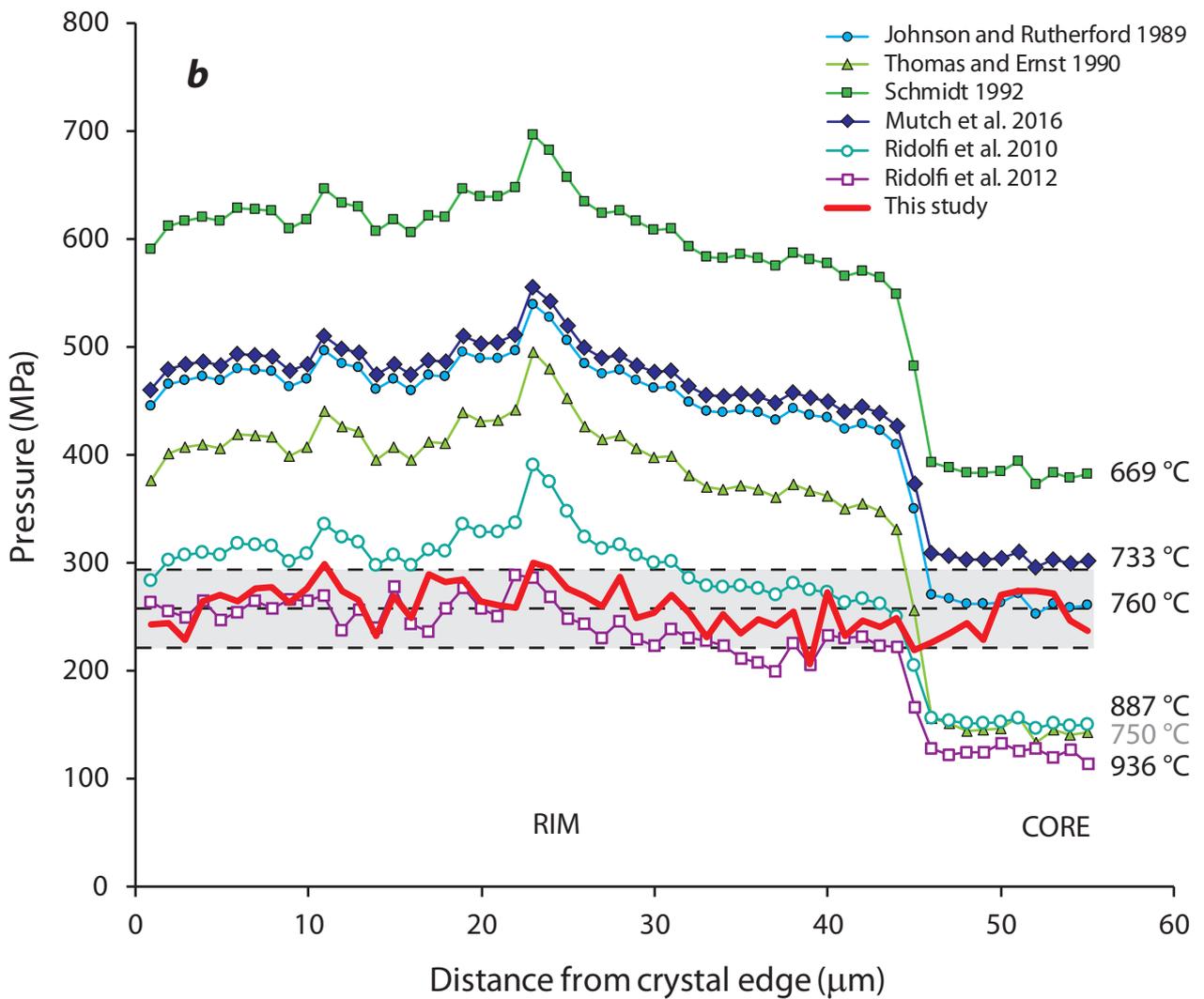
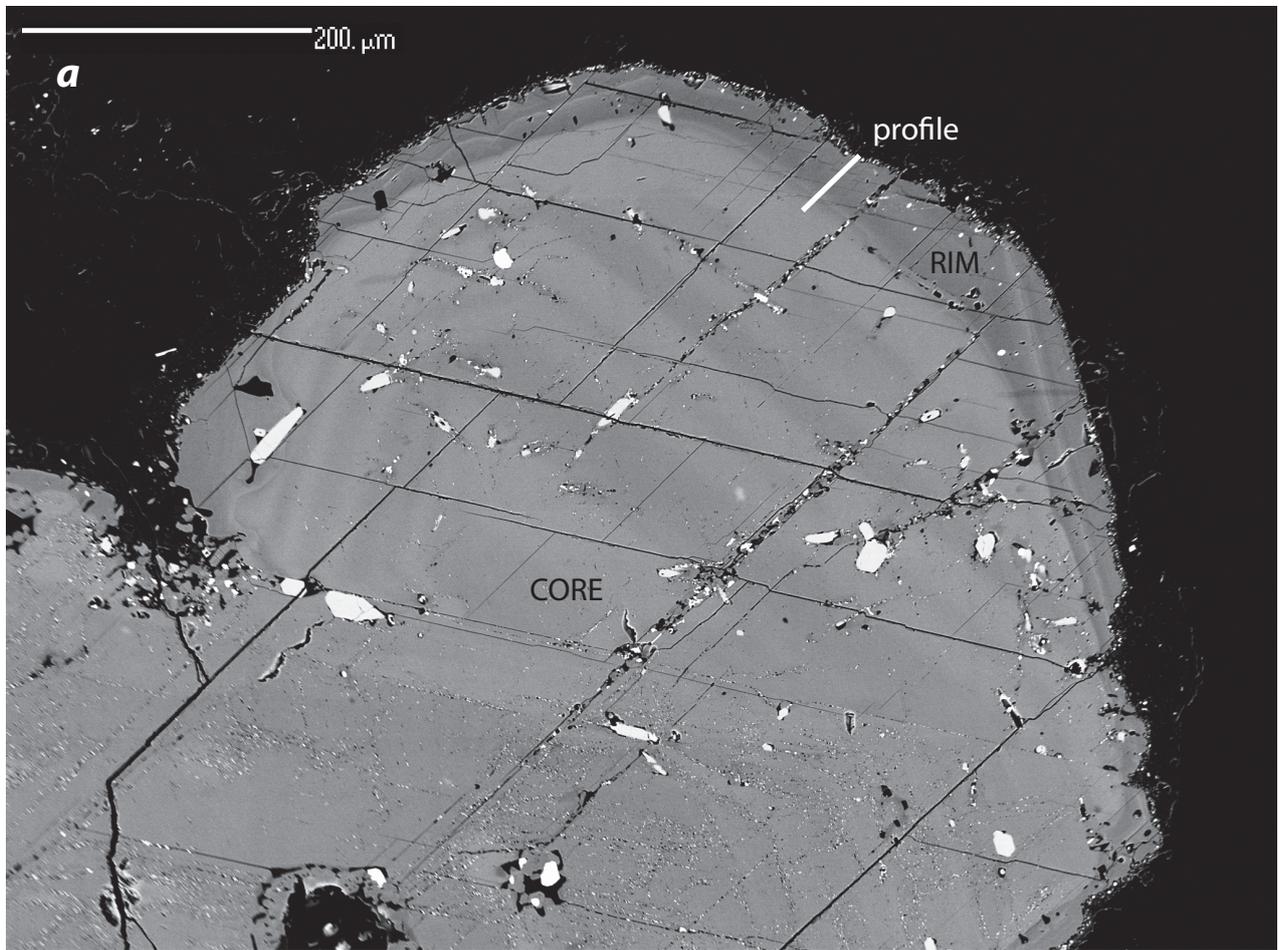


Figure 3

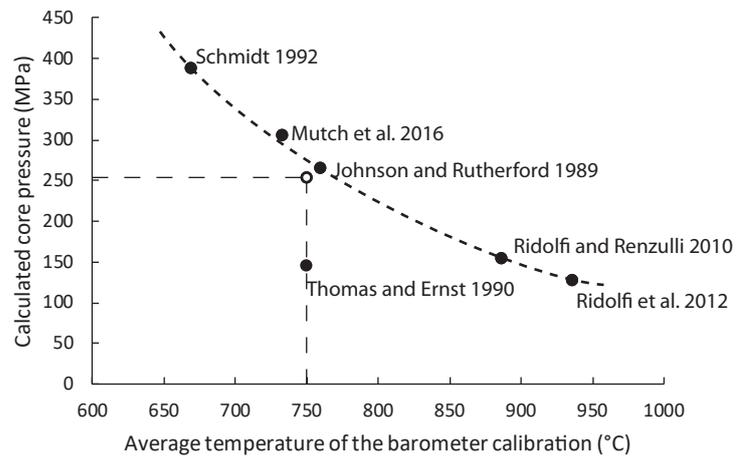


Figure 4

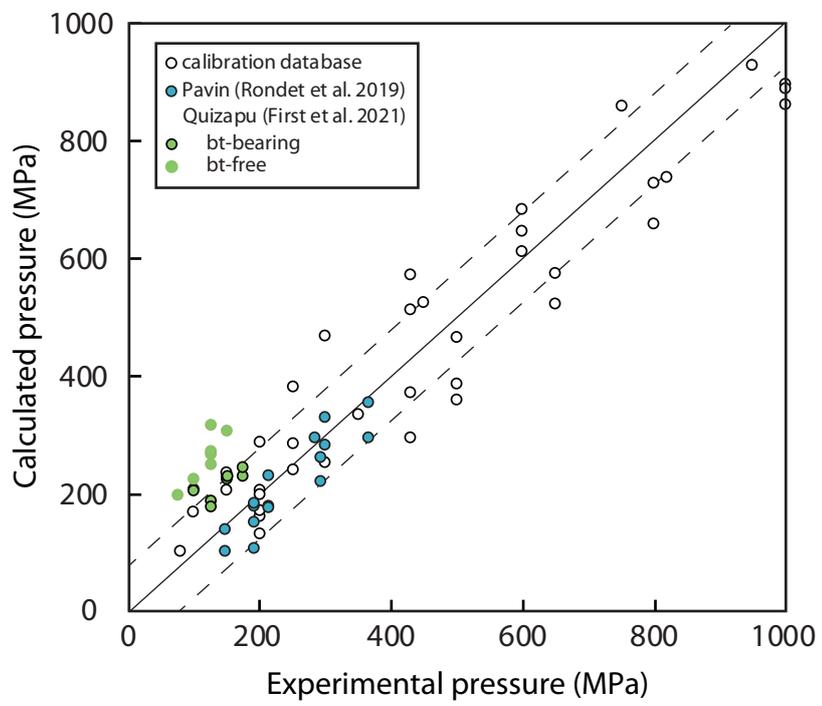


Figure 5

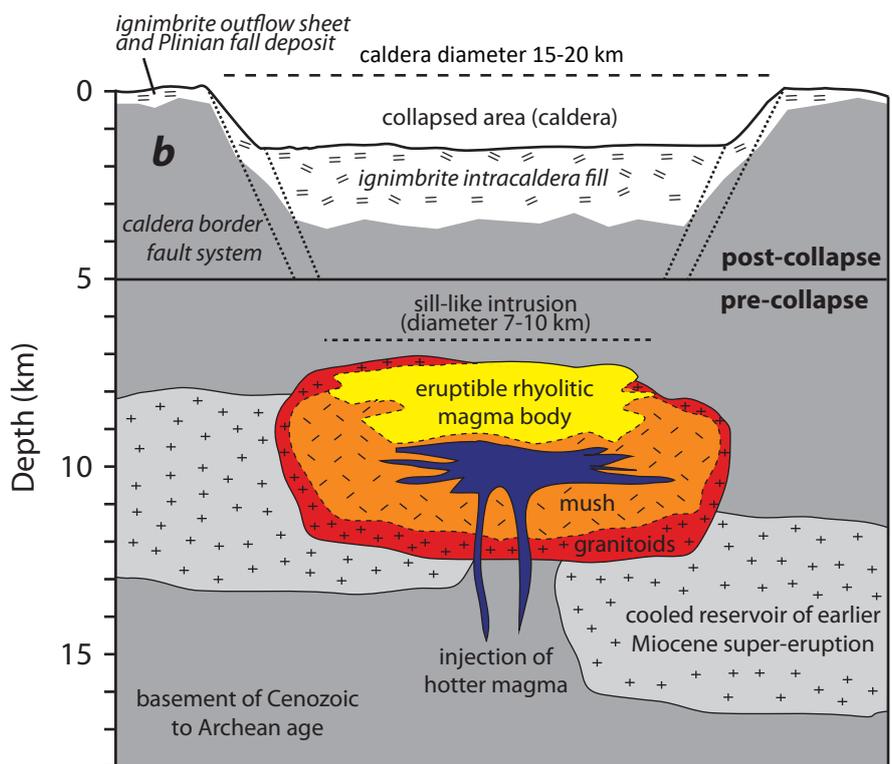
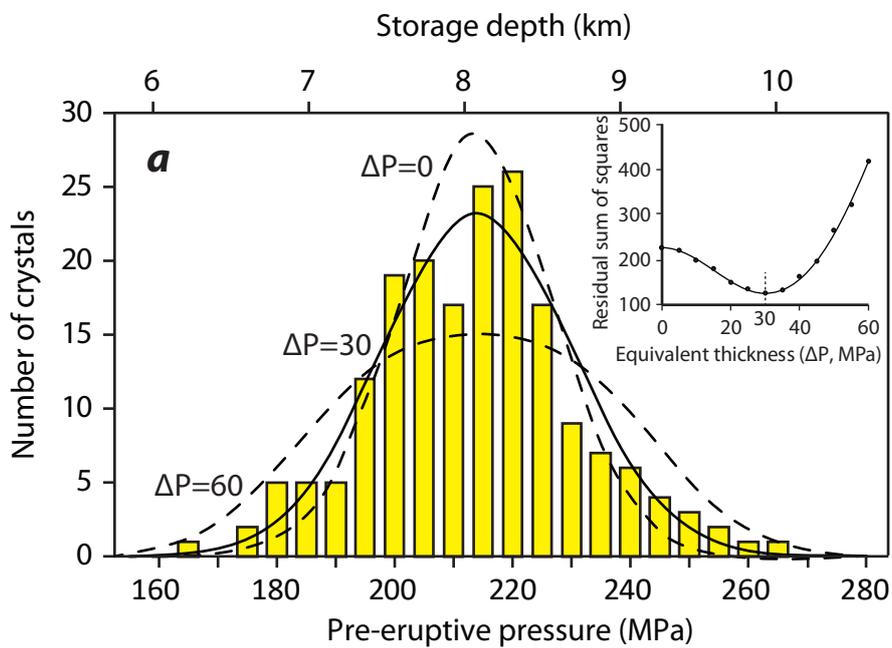


Figure 6