

1 *JVGR review paper*

2
3 **Exsolved Volatiles in Magma Reservoirs**

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35 **Abstract**

36 We review our understanding of the exsolved volatile phase co-existing with magmas during pre-
37 eruptive storage at the pressures and temperatures corresponding to crustal magma reservoirs. We
38 explore the consequences and implications of such a volatile phase for magma and ore body
39 petrogenesis and the fluid dynamics of magma reservoirs. We outline the geochemical constraints
40 on the size and composition of the exsolved volatile phase that may co-exist with magmas in the
41 crust. We distinguish between decompression-driven and crystallization-driven exsolution, and
42 describe the implications of the volatiles for the dynamics of the magma reservoir, using key
43 natural examples and case studies. We discuss eruptions triggered by second boiling, and the
44 various regimes of magma mixing and magma overturn that may be induced by second boiling in
45 a layered reservoir. We also explore the control of the volatile content of the magma on the mass
46 erupted during an eruption episode, and compare our models to eruption datasets. We then turn to
47 the mechanisms for magma-volatile separation, noting that in crystal-poor melts convective
48 separation of exsolved volatiles may dominate while in crystal-rich melts, volatiles may generate
49 channels or permeable-flow pathways through the crystal mush, thereby separating from the
50 parent magma. We discuss the implications of the accumulation of the exsolved volatile phase at
51 the roof zones of crystal-rich reservoirs for the large gas emissions observed during explosive
52 eruptions, and for the development of metal-rich porphyry deposits.

53

54 **1. Introduction**

55 Magmas close to the surface of Earth, stored within crustal reservoirs, contain exsolved volatiles.
56 This exsolved volatile phase is made up of water and carbon dioxide primarily, but also
57 significant amounts of sulfur and halogen gases, and trace constituents such as volatile metals
58 (Gerlach, 1980; Symonds et al., 1994). These volatiles are dominantly sourced from the mantle,
59 but may also be assimilated from crustal rocks and fluids (Mason et al., 2017) and are
60 enriched in silicate melts as they cool and fractionate. Typically, basaltic melts leave the mantle
61 carrying 0.1-4 wt% H₂O and <2 wt% CO₂, depending on tectonic setting (Blundy et al., 2010;
62 Plank et al., 2013; Wallace, 2005), which increases due to fractionation prior to volatile
63 saturation, reaching several wt% H₂O for magmas in arc settings (Plank et al., 2013; Sisson and
64 Layne, 1993). These volatile species have a propensity to exsolve or partition into a low density
65 exsolved volatile phase at crustal pressures, which may be a supercritical fluid at high pressures,
66 and a vapour phase at low pressures.

67

68 Despite only being a minor constituent of magmas, exsolved volatiles have enormous importance
69 for a myriad of magma reservoir processes. An exsolved volatile phase lowers the bulk density
70 and increases the buoyancy of magmas (Anderson, 1995), influences phase equilibria (Ghiorso
71 and Gualda, 2015; Moore and Carmichael, 1998), as well driving convection (Cardoso and
72 Woods, 1999; Longo et al., 2006) and promoting magma mingling and mixing through overturn
73 (Huppert et al., 1982; Ruprecht et al., 2008). Recent work has shown that the exsolved volatile
74 phase may be mobile in crystal-rich magma (Barth et al., in review; Oppenheimer et al., 2015;
75 Parmigiani et al., 2016), leading to accumulation in the roof zones of magma reservoirs, which
76 may play a role in triggering eruptions, activating hydrothermal systems through volatile
77 injection (Chiodini et al., 2012) and in the generation of porphyries hosting economic
78 concentrations of metals (Guo and Audétat, 2017; Huber et al., 2012).

79
80 Important new advances in observing and modeling magma reservoirs and their volatile
81 systematics have prompted this review. Our understanding of the form and organisation of crustal
82 magma reservoirs has evolved in recent years, towards a picture of trans-crustal, mush-
83 dominated, heterogeneous magma reservoirs which may be long-lived relative to inter-eruption
84 timescales (Cashman et al., 2017; Cooper and Kent, 2014). Petrological evidence, from melt
85 inclusion and pyroxene barometry (Edmonds et al., 2016; Hartley et al., 2014; Longpré et al.,
86 2017; Métrich and Wallace, 2008; Neave and Putirka, 2017) suggests that magmas may be stored
87 over a wide depth interval and that the magmas typically carry crystal cargos that record complex
88 mixing and mingling histories (Cashman and Blundy, 2013; Cooper and Kent, 2014; Costa et al.,
89 2013; Neave et al., 2013), suggesting repeated recharge and mixing of localised melts and
90 mushes. Geophysical surveys of the crust show that melt-dominated magma bodies do not
91 generally exist; instead it is envisaged that melts are disseminated in mushes or in closely-spaced
92 sill-like bodies (Cordell et al., 2018; Kiser et al., 2016; Pritchard and Gregg, 2016).

93
94 Large fluxes of deep-derived sulfur-rich gas from long-lived eruptions such as Soufrière Hills
95 Volcano (Montserrat) (Christopher et al., 2015; Edmonds et al., 2010; Edmonds et al., 2001) and
96 Bagana Volcano (Papua New Guinea) (Kilbride et al., 2018), which continue during prolonged
97 eruptive pauses indicate that permeable, outgassing pathways through crystal-rich magma may be
98 maintained from depth, perhaps tapping zones where an exsolved volatile phase accumulated
99 near the topmost regions of the magma reservoir system over multiple eruption timescales
100 (Christopher et al., 2015), with the system being recharged from time to time with heat and
101 volatiles by underplating mafic magmas (Bachmann and Bergantz, 2006; Murphy et al., 2000;

102 Plail et al., 2014). The accumulation of an exsolved volatile phase in the roof zone of such a
103 protracted reservoir may play a critical role in triggering and sustaining eruption. Observations of
104 “excess sulfur” using space-borne satellites (Carn et al., 2016; Wallace, 2001) may be direct
105 evidence of such deep accumulations of exsolved volatiles. Large volcanic “outgassing events”
106 which may be observed using strain measurements (Hautmann et al., 2014), may relieve reservoir
107 pressure and prevent magmatic eruptions. Accumulation of an exsolved volatile phase may also
108 have important implications for the interpretation of ground displacements at the surface through
109 the formation of a highly compressible region of the magma reservoir (McCormick Kilbride et
110 al., 2016; Rivalta and Segall, 2008), which may buffer volume changes and suppress ground
111 deformation.

112

113 This review will be concerned with both the geochemistry and physical dynamics of the exsolved
114 volatile phase in the crust prior to eruption. We bring together geochemical and physical
115 perspectives to review the state of the art in our understanding of the role and behaviour of
116 volatiles in magma reservoirs and during magma ascent and eruption. We provide a synopsis of
117 recent progress in this area in the key areas of observation, modeling and experiment and
118 highlight outstanding, critical problems that remain to be solved.

119

120 **2. Achieving volatile saturation in magmas**

121 Magmas exsolve water and carbon dioxide as they rise through the crust, cool and crystallise.
122 Owing to the relationship between volatile solubility and pressure (Dixon et al., 1995; Moore et
123 al., 1998a; Newman and Lowenstern, 2002; Pan et al., 1991; Stolper and Holloway, 1988), melts
124 can reach saturation with respect to an exsolved volatile phase during decompression (termed
125 “first boiling”) or during isobaric cooling and crystallisation (of volatile-poor crystals) in a
126 magma storage area (“second boiling”) (**figure 1A and B**), leading to the formation of an exsolved
127 magmatic volatile phase (MVP) (Candela, 1997). A magma is said to be volatile-saturated when
128 an exsolved volatile phase (which may be a supercritical fluid at high pressures, or a vapour at
129 low pressures) is present and in equilibrium with the melt. Volatile exsolution occurs when the
130 sum of the vapour pressures of the components of a liquid, $\sum(P_i^{vapour})$, is equal to the load
131 pressure (Candela, 1997). Volatile exsolution into bubbles will occur when either the load
132 pressure is reduced, for example during decompression or removal of lithostatic load, “first
133 boiling”, or $\sum(P_i^{vapour})$ is increased, for example during crystallisation, “second boiling”.

134

135 Exsolution of water from silicate melt during decompression-induced degassing, or first boiling,
136 results in a lowering of the solidus, owing to a decrease in the entropy of mixing caused by the
137 loss of water from the melt. Decompression of magma is therefore usually accompanied by
138 extensive degassing-induced crystallisation (Cashman and Blundy, 2000; Cashman, 2004), which
139 has the effect of promoting further volatile exsolution and increasing the bulk viscosity of the
140 magma (Dingwell et al., 1996). These processes are first order controls on eruption style and the
141 onset of fragmentation (Blundy and Cashman, 2005; Dingwell, 1996; Melnik and Sparks, 2002)
142 (see later). Second boiling takes place in magma storage areas in the crust, where magmas cool
143 beneath their solidus temperatures and crystallise. If the fractionating assemblage is dominantly
144 anhydrous then as the liquid fraction decreases, the vapour pressure of the volatile components
145 increases, driving exsolution of volatiles (**figure 1B**). As magmas rise through the crust, the
146 balance between first and second boiling changes. As magma ascent slows, volatile exsolution
147 may be driven dominantly by cooling and crystallisation; whereas rapid rise of magma through
148 dykes to the surface will experience dominantly first boiling vesiculation and expansion. For the
149 case of large, complex magma reservoirs which evolve over long timescales (10^4 - 10^5 yr) (Cooper
150 and Kent, 2014; Hawkesworth et al., 2000) the production of an exsolved volatile phase may
151 occur sporadically, with magma recharge events, superimposed on a background of slower
152 exsolved volatile production driven by cooling and crystallisation.

153
154 The composition of the evolving exsolved magmatic volatile phase will depend on the initial
155 volatile content of the magma, the pressure, temperature, melt composition and oxidation state.
156 There have been many recent formulations of empirical (Moore et al., 1998b) and
157 thermodynamic (Burgisser et al., 2015; Dixon and Stolper, 1995; Moretti et al., 2003; Papale,
158 1999; Witham et al., 2012) models to describe the chemical evolution of melt and exsolved
159 volatile phase in equilibrium during transit through the crust (**figure 2**). The thermodynamic
160 models are calibrated by experimental data on H₂O and CO₂ solubility and the partitioning of
161 sulfur between the aqueous exsolved volatile phase and silicate melt at a range of conditions.
162 **Figure 2** illustrates the effect of CO₂ on the composition and pressure at which volatile-saturation
163 occurs (Candela, 1997). One melt with 400 ppm CO₂ and around 2 wt% H₂O crystallises at a
164 constant pressure of 200 MPa. Initially it is ‘vapor’-undersaturated (here ‘vapor’ refers to the
165 exsolved volatile phase), but reaches vapor-saturation after around 50% crystallization (red dot in
166 **figure 2**). At this stage the concentrations of H₂O and CO₂ have approximately doubled and
167 $\sum(P_i^{vapour})$ is 200 MPa, and the partial pressures of H₂O and CO₂ are both ~ 100 MPa and
168 therefore the molar proportions of CO₂ and H₂O in the co-existing exsolved volatile phase are

169 approximately equal. Further vapor-saturated crystallization will cause the melt to evolve along
170 the 200 MPa isobar, towards more H₂O-rich melt compositions. If no CO₂ were present (**figure 2**,
171 bottom orange dot), then 64% crystallization is required before vapor-saturation occurs. **Figure 3**
172 shows the relationship between the melt H₂O content, the mass of the exsolved volatile phase and
173 the degree of crystallization at 200 MPa for a range of initial water contents. After 50%
174 crystallization, by which time basalt has become basaltic andesite, 1-5 wt% exsolved volatiles is
175 likely to co-exist with the melt. This is a minimum estimate; if CO₂ is present this fraction will be
176 higher. Many authors have suggested that magmas may experience ‘CO₂-flushing’ which will
177 tend to drive the melt towards higher CO₂ concentrations (from right to left along an isobar in
178 **figure 2**) (Caricchi et al., 2018).

179

180 *2.1. Volatiles in natural magmatic systems*

181 Perhaps our best source of information regarding the pre-eruptive volatile concentrations of
182 silicate melts are given by the volatile contents of melt inclusions (Kent, 2008; Lowenstern,
183 1995; Métrich and Wallace, 2008; Sides et al., 2014b). We now understand, however, that many
184 processes modify the concentrations of volatiles in melt inclusions post-entrapment, such as
185 sequestration of CO₂ into a shrinkage bubble (Esposito et al., 2014; Moore et al., 2015; Wallace
186 et al., 2015), perhaps driven by cooling and post-entrapment crystallization (Sides et al., 2014a;
187 Steele-Macinnis et al., 2011), diffusive loss (Bucholz et al., 2013; Gaetani et al., 2012; Lloyd et
188 al., 2013) or gain (Hartley et al., 2015) of hydrogen. A dataset of melt inclusion CO₂ and H₂O
189 data, for A) a range of basalts in hotspot and arc settings and B) andesites, dacites and rhyolites in
190 arc and rift settings, is shown in **Figure 4**. The data for both the mafic magmas and the more
191 evolved magmas show an array in volatile concentrations, caused by a combination of factors
192 such as decompressional degassing concurrent with melt inclusion entrapment and CO₂ fluxing
193 (Métrich and Wallace, 2008), mixing (Dixon et al., 1991) and volatile loss from the inclusions
194 (Gaetani et al., 2012; Maclennan, 2017). A degassing path is one whereby CO₂ is lost at high
195 pressure, followed by H₂O at low pressure (Dixon and Stolper, 1995). The dissolved volatile data
196 for both basalts and rhyolites, in all tectonic settings, is consistent with volatile-saturation at
197 pressures of up to 300 Mpa or more in the crust, equivalent to 11-13 km depth. For ocean island
198 magmas, the exsolved volatile phase at this depth is highly CO₂-rich, owing to the much lower
199 bulk H₂O concentrations; in arcs, the deep exsolved volatile phase is more hydrous. If, as seems
200 likely, the melt inclusion record is overprinted by shallow storage prior to eruption (due to H⁺
201 loss by diffusion, see above), the deeper record may be routinely erased.

202

203

204

205 Phase equilibria studies have been of value in reconstructing the pre-eruptive water contents of
206 arc basalts (Barclay et al., 1998; Sisson and Grove, 1993). In Mexico, phase assemblages in
207 erupted rocks are consistent with experimental assemblages under volatile-saturated conditions at
208 170-200 MPa (Moore and Carmichael, 1998), indicating that an aqueous fluid phase with a high
209 activity of H₂O co-existed with magma prior to eruption. Plagioclase hygrometers (Lange et al.,
210 2009; Putirka, 2005; Waters and Lange, 2015) allow estimates of the water content of basaltic
211 melts to be made: it has been suggested that An contents of plagioclase in arc basalts in Japan
212 indicate pre-eruptive basaltic water contents of 3-5 wt% (Ushioda et al., 2014), which is
213 consistent with melt inclusion studies (Kuritani et al., 2014; Rose-Koga et al., 2014). These
214 basalts have potential to generate large amounts of an exsolved volatile phase upon evolution to
215 higher-silica magmas. Other mineral proxies may be useful to reconstruct melt volatile
216 concentrations. A recent study of hydrogen in the cores of orthopyroxenes erupted during
217 vulcanian activity at Soufrière Hills Volcano were used to suggest that melt H₂O contents were 4-
218 9 wt% prior to eruption, implying volatile-saturated magma storage at depths of up to ~12 km
219 prior to eruption (Edmonds et al., 2016). Anorthite-rich plagioclase in xenocrysts (derived from
220 disaggregated mushes), Al-rich melts and experimental phase equilibria were used to infer that
221 parental basaltic andesites beneath St Kitts (Lesser Antilles) are water-rich (>9 wt%) and they
222 crystallise over a wide pressure range (1.5 to 6.0 kbar) (Melekhova et al., 2017).

223 Geochemical proxies might allow reconstruction of magmatic water budgets. It has been
224 recognised recently that high Sr/Y ratios in magmas are linked to fertility in copper (Richards,
225 2011). High Sr/Y ratios, for example in Tibetan post-collisional magmas, have been attributed to
226 high magmatic water contents of >10 wt%, acting to suppress plagioclase fractionation (and
227 hence Sr removal) during fractional crystallisation of hydrous basalt parent magma at depth
228 (Chiaradia et al., 2012; Lu et al., 2015). These magmas have been compared to others in
229 subduction zone settings: silicic high Sr/Y andesites and dacites at Mount Shasta, a stratovolcano
230 in the southern Cascades, contain melt inclusions with >10 wt% H₂O which crystallised at 0.8
231 GPa (Grove et al., 2002). Similar experimental phase equilibria were shown for the Pinatubo
232 dacite melt: in order to produce high Sr/Y with a starting melt composition of 16.5 wt% Al₂O₃
233 requires a melt water content of >10 wt% (Prouteau and Scaillet, 2003). The existence of
234 hydrous magmas deep in the crust lends support to the notion that magma reservoirs from the
235 mid-crust upward contain substantial fractions of exsolved water as supercritical fluid or low
236 density vapour at lower pressures.

237 Plutonic rocks exposed at the surface, however, present only scant evidence of an exsolved
238 magmatic volatile phase, despite abundant petrological evidence of hydrous components in the
239 melt (the presence of amphibole or biotite, for example). Comb-layered quartz, with its high
240 concentrations of fluid-mobile trace elements, is found in shallow felsic intrusions associated
241 with ore bodies, and is proposed to precipitate at the wallrock-magma contact from exsolved
242 hydrous fluids at the apical regions of magma bodies (Lowenstern and Sinclair, 1996). Some
243 granite bodies and pegmatites contain miarolitic cavities made up of super-solidus minerals,
244 externally nucleated, projecting into a quasi-spherical void (Candela, 1997). These features are
245 accepted as good evidence for magmatic exsolved volatile phase saturation in plutonic igneous
246 rocks (Candela, 1997). The absence of miarolitic cavities in granites and other igneous rocks does
247 not preclude that the magma body was saturated with respect to an exsolved volatile phase; the
248 preservation of such features depends on the depth and water content of the magma, as well as
249 crystal size, and the permeability of the mush during solidification. Some of these factors will be
250 discussed further in section 5.

251 Overall the data from natural systems appear to show that magmas may be volatile-saturated
252 from depths of 9 to 12 km up to the surface in most magmatic systems, including hotspots, arcs
253 and continental rifts. There may be exceptions, however. A recent study using apatites to
254 reconstruct the volatile budget of Campi Flegrei suggested that phonolitic magma at 7 km depth
255 was volatile-undersaturated; while that stored at 3 km was volatile-saturated (Stock et al., 2016).
256 In mid-ocean ridge and hotspot settings, basaltic melts typically contain little H₂O (<1.0 wt% and
257 usually <0.5 wt%; **figure 4**); for these magmas, saturation in an exsolved fluid phase occurs in
258 the mid-crust but the fluid is initially very CO₂-rich; water does not exsolve from the melt in
259 significant quantities until the magma is within a few hundred metres of the surface (Newman
260 and Lowenstern, 2002); or until after significant amounts of isobaric crystallization has taken
261 place.

262

263 *2.2. Sulfur partitioning into an exsolved volatile phase*

264 As well as the primary volatile species, H₂O and CO₂, sulfur is also present at bulk
265 concentrations of up to 0.5 wt% in arc magmas (Carroll and Rutherford, 1985; Jugo, 2009;
266 Metrich and Clocchiatti, 1996; Wallace and Edmonds, 2011). Sulfur has significance owing to its
267 ability to complex with metals in the magmatic exsolved volatile phase and transport them to
268 sites of ore deposits (Botcharnikov et al., 2010; Zajacz and Halter, 2009); but also because
269 “excess” sulfur emissions during large explosive volcanic eruptions may lead to tropospheric
270 cooling owing to the influence of sulfate aerosols on the radiation budget of Earth’s atmosphere

271 (Robock, 2000). It has long been observed that the mass of sulfur released, as observed using
272 satellite sensors, from intermediate, i.e. andesite, dacite, eruptions in arcs is far larger than can be
273 accounted for by the syn-eruptive degassing of melt, as deduced from the sulfur content of melt
274 inclusions (Wallace, 2001; Wallace and Gerlach, 1994). A solution to this conundrum emerged
275 after the 1984 El Chichon eruption, which released 7 Tg sulfur into the atmosphere (Bluth et al.,
276 1997). It was proposed that, prior to eruption, a large fraction of the sulfur existed in a pre-
277 eruptive exsolved volatile phase, rich in H₂S (Luhr et al., 1984). This hypothesis gained traction
278 with the observation of other large explosive eruptions (Wallace, 2001; Wallace and Gerlach,
279 1994; Westrich and Gerlach, 1992), as well as the experimental observations, which followed,
280 that sulfur partitions strongly into aqueous exsolved volatile phase at crustal pressures for water-
281 rich, relatively evolved magmas (Clémente et al., 2004; Keppler, 1999; Scaillet et al., 1998;
282 Scaillet and Pichavant, 2005; Zajacz et al., 2012).

283

284 The sulfur content of an exsolved volatile phase in a magma reservoir may be estimated using a
285 thermodynamic approach using homogeneous equilibria in the C-O-H-S system (Oppenheimer et
286 al., 2011; Scaillet and Pichavant, 2003). An exsolved volatile phase that evolves in a magma
287 reservoir prior to eruption is highly variable in sulfur content, dependent on fO_2 , magma
288 composition and temperature. Furthermore, in order to make the computed composition of the
289 exsolved volatile phase match with that observed at the surface, using satellite-based methods,
290 which has been overprinted by decompression-induced degassing, at least 1 wt% and in some
291 cases up to 5 wt% of an exsolved volatile phase must be present in the magma reservoir. A pre-
292 eruptive volatile phase is not required, or much less is required, to explain volcanic gas
293 composition and output from more mafic systems (Oppenheimer et al., 2011; Scaillet and
294 Pichavant, 2003). Large amounts of exsolved volatiles co-existing with magmas at depth will
295 buffer the C/S ratio of volcanic gases observed at the surface.

296

297 The presence of a few wt% exsolved volatiles from thermodynamic calculations is consistent
298 with studies of melt inclusion geochemistry and models of fractional crystallisation, which
299 suggested that the magma reservoir that fed the Bishop Tuff was zoned with respect to exsolved
300 volatiles, containing up to 6 wt% near the roof zone (Wallace et al., 1999). A recent study of the
301 volcanic products of the Samalas eruption, Indonesia, which involved eruption of at least 40 km³
302 and has been unmasked as the source of the mysterious 1257 sulfate spike in the ice core records
303 (Lavigne et al., 2013) suggests exsolved magmatic volatile contents of >3 wt%, based on melt
304 inclusion systematics and fractionation modeling (Vidal et al., 2016). Sulfur partitioning into this

305 water-rich exsolved volatile phase resulted in a syn-eruptive sulfur emission into the atmosphere
306 of 79 +/- 6 Tg (Vidal et al., 2016). Studies of long-lived eruptions with well-constrained records
307 of magma and gas fluxes, combined with melt inclusion studies, have allowed independent
308 estimates of the proportion of exsolved volatiles present in the magma reservoir. At Soufrière
309 Hills (Montserrat) much of the sulfur exists in a pre-eruptive exsolved volatile phase prior to
310 eruption, which may comprise to to 2-3 wt% of the magma at 150-200 MPa (Edmonds et al.,
311 2014b).

312

313 **3. Consequences of exsolved volatiles in magma reservoirs for rheology and dynamics**

314 The presence and production of an exsolved volatile phase in crustal magma reservoirs can lead
315 to a number of different effects on rheology, compressibility and on eruption duration and style,
316 depending on a range of factors, including whether the magma reservoir is sealed or open to the
317 atmosphere, the crystal content and viscosity of the melt, and on whether the magma reservoir is
318 composed of a single magma body or multiple bodies of melt with different composition and gas
319 content. We first describe the changes in magma rheology that might occur when second boiling
320 occurs (**figure 1**) in a crystal-rich magma, generating an exsolved volatile phase, manifest as
321 disseminated fluid bubbles. We then describe the change in the pressure and density of magma in
322 a chamber resulting from the exsolution of volatiles, associated with second boiling, assuming the
323 mixture of melt, bubbles and crystals remains well-mixed. We then consider the effects of the
324 relative motion of the gas phase, melt and crystals, and consider the implications of this for the
325 dynamics of the system, including the effects of magma mixing, and observations of large
326 volumes of erupted gas (“excess sulfur”; see section 2.2).

327

328 *3.1 Magma Compressibility*

329 In a magma that contains dissolved volatiles, cooling and crystallisation will induce saturation in
330 an exsolved volatile phase (second boiling; as shown in **figure 1**). The exsolved volatile phase is
331 typically of lower density than the melt phase and this process therefore leads to an increase in
332 volume and hence pressure of the magma body as it deforms the surrounding crust. The effect of
333 this increase in pressure may be to trigger an eruption (Blake, 1984; Tait et al., 1989) or the
334 injection of magma to a higher level in the crust. The relation between the change in volume and
335 the change in pressure during both the inflation and the deflation phases of the evolution of the
336 system depends on the compressibility of the melt-volatile-crystal mixture (Huppert and Woods,
337 2002; Rivalta and Segall, 2008; Segall, 2013; Voight et al., 2010). With a volatile-saturated melt,
338 the magma may be of order 10 times more compressible than a magma with no exsolved

339 volatiles. As a result, there may be a much greater volume of magma erupted prior to the
 340 relaxation of the reservoir pressure and closure of the conduit (Huppert and Woods, 2002; Woods
 341 and Huppert, 2003). Although the detailed calculation of the compressibility depends on the
 342 volatile solubility as a function of pressure and crystal content, it is useful to explore the
 343 predictions of a simplified model using (1a) Henry's law, (1b) a relation between the crystal
 344 content and temperature; (1c) a model of an elastic chamber with effective bulk rock
 345 compressibility (the inverse of the bulk modulus) β ; and (1d) an idealized model for the density
 346 of the gas as given by the ideal gas law, which is approximately valid for shallower (upper
 347 crustal) systems.

$$348 \quad n_e = n_o - sP \text{ if } n_o > sP \text{ and } n_e = 0 \text{ if } n_o < sP \quad (1a)$$

$$349 \quad x = \frac{(1200 - T)}{250} \quad (1b)$$

$$350 \quad dV = -V\beta dP \quad (1c)$$

$$351 \quad \frac{1}{\rho} = \left(\frac{n_e RT}{P} + \frac{(1 - n_e)}{\rho_m} \right) \quad (1d)$$

352 where n denotes volatile mass fraction, with subscript e denoting exsolved volatiles and o
 353 denoting the total mass of volatiles per unit mass of original melt. P is pressure and s is a
 354 solubility constant. x denotes crystal fraction (excluding the volatile phase), T temperature (in
 355 Kelvin) and V the chamber volume, while R is the ideal gas constant. The bulk density of the
 356 magma-crystal-volatile mixture is denoted by ρ while the bulk density of the melt-crystal
 357 mixture is ρ_m . Such a simplified model provides insight into the behaviour of the system and
 358 allows us to distinguish the different controlling effects on the evolution of the melt.

359
 360 Combining these relations, we can calculate the compressibility (or the inverse of the bulk
 361 modulus of the mixture) defined as

$$362 \quad \beta_m = -\left(\frac{1}{V}\right) \frac{dV}{dP} \quad (2)$$

363 as a function of pressure. In **figure 5a**, we illustrate the compressibility for the cases of magma
 364 with three different volatile contents at different depths in the crust, which shows that the
 365 compressibility becomes much larger once the melt reaches shallow depths where there are
 366 exsolved volatiles. The compressibility of the melt evolves as the melt cools and crystallises

367 during second boiling. In **figure 5b**, we illustrate the compressibility of magma with bulk H₂O
368 contents of 3, 4 and 6 wt % at a depth of 5 km, with the compressibility shown as a function of
369 the crystal content of the melt. Once the magma crystallises sufficiently such that the melt
370 reaches saturation in an exsolved volatile phase, the magma compressibility increases by over an
371 order of magnitude.

372

373 Magma compressibility, caused by the presence of an exsolved magmatic volatile phase in the
374 reservoir, has implications for the interpretation of volcano monitoring data and impacts the
375 magnitude of ground displacements as well as the composition and flux of volcanic gases (Biggs
376 and Pritchard, 2017; McCormick Kilbride et al., 2016). At Soufrière Hills Volcano, Montserrat
377 for example, a gas-rich compressible magma (the ‘magma sponge’) was invoked to explain why
378 surface displacements, measured by Global Positioning Satellite (GPS) receivers on the volcano’s
379 flanks, yielded a much smaller volume displacement than the actual volume erupted, for
380 particular time periods within the eruption (1995-2011) (Voight et al., 2010). It has been
381 observed that the volume change inferred from magma chambers feeding shallower dyke
382 intrusions is often a factor of 4-5 lower than the volume increase observed in the dyke, which has
383 been explained by a combination of decompression-driven degassing and consequent increase in
384 magma compressibility, combined with the more compliant dyke geometry (Rivalta and Segall,
385 2008). A database of eruptions for which the change in volume of the chamber and the volume
386 erupted were well-constrained by the complementary data sets obtained from satellite
387 measurements of the volcano deformation and ground-based measurements of the mass erupted is
388 summarized by (McCormick Kilbride et al., 2016). They find that the volumes are generally in
389 the ratio 1-20, which indicates the presence of exsolved volatiles in the magma reservoir;
390 however, it is worth noting that it is somewhat difficult to constrain the actual change in volume
391 of the magma chamber without detailed information of the shape and depth of the chamber,
392 which can lead to changes in the effective bulk compressibility β by up to an order of magnitude
393 (Anderson and Segall, 2011; Rivalta and Segall, 2008; Segall, 2013).

394

395 *3.2. Pressure increase during second boiling*

396 Second boiling during cooling and crystallization, generating a magmatic exsolved volatile phase,
397 can produce enough pressure in the magma reservoir to trigger eruption (Blake, 1984; Tait et al.,
398 1989; Woods and Cardoso, 1997). As a simplified calculation, in a more viscous melt, the
399 pressure increase through cooling and the associated crystallization and volatile exsolution can be
400 expressed by the relation

$$401 \quad (\beta + \beta_m) dP = \left(\frac{1}{\rho} \right) \left(\frac{d\rho}{dT} \right) dT = -\rho \left(\frac{nR}{T} + \left(\frac{sRT}{P^{0.5}} \right) \left(\frac{dX}{dT} \right) \right) dT \quad (3)$$

402

403 where the change in volume associated with the cooling and volatile expansion and exsolution is
 404 accommodated by compression of the remaining magma-volatile mixture, with compressibility
 405 β_m , and the surrounding country rock with effective compressibility β .

406

407 The predictions of this model are shown in **figure 5c** in which we see that the change in pressure
 408 per degree of cooling, relative to the critical overpressure to trigger an eruption, as a function of
 409 the crystal content of the melt. In this model, the chamber is assumed to be closed, and cooling
 410 leads to crystallisation, exsolution and deformation of the surrounding country rock. Here the
 411 critical overpressure to drive an eruption, P_c , has been set to the value 10^7 Pa (Blake, 1984;
 412 Woods and Cardoso, 1997) for illustration. It is seen that once the magma is volatile-saturated,
 413 the continued exsolution of volatiles with cooling leads to a significant expansion of the melt-
 414 crystal-volatile mixture and a large associated increase in the pressure. In the calculations shown,
 415 for magma with 3, 4 or 6 wt% H_2O , with cooling of the melt by an amount of order $10^\circ C$, the
 416 pressure will increase up to about 10^7 Pa, leading to eruption. In contrast, if the melt is not
 417 volatile-saturated, then the increase in pressure with cooling is only associated with either the
 418 density change as melt precipitates crystals, or the bulk cooling of the melt, and these are much
 419 smaller effects.

420

421 *3.3 Effect of compressibility on the mass of magma erupted*

422 If the critical pressure to trigger an eruption from a magma reservoir is ΔP_c , where $\Delta P_c \ll P_1$,
 423 (where P_1 is the lithostatic pressure), then the volume increase in the magma reservoir to
 424 accommodate this overpressure, relative to the lithostatic pressure, is given by $\Delta V = V \beta \Delta P_c$,
 425 based on the response of the crust. On eruption, this overpressure is relieved. The mass of melt in
 426 the chamber associated with the overpressure, ΔM , is given by

$$427 \quad \Delta M = V \Delta \rho + \rho \Delta V = M (\beta + \beta_m) \Delta P_c \quad (4)$$

428 and this also corresponds to the mass erupted to relieve the overpressure. The volume erupted,

429 $\Delta V_e = \frac{\Delta M}{\rho}$, depends on the volume change of the chamber, ΔV , according to the relation

$$430 \quad \Delta V_e = \left(\frac{\beta_m}{\beta} + 1 \right) \Delta V \quad (5)$$

431 For a given fracture strength of the crust and hence a given change in volume of the chamber ΔV
 432 the volume erupted, ΔV_e , increases with the volatile content of the magma, since the very large
 433 compressibility of the exsolved volatile phase can accommodate large changes in volume for
 434 relatively small changes in pressure. This leads to much larger erupted volumes for volatile-rich
 435 magmas (Bower and Woods, 1998; Huppert and Woods, 2002).

436
 437 If an eruption is modelled with a simple frictional law associated with the frictional resistance in
 438 the conduit, fL , and the overpressure in the chamber, $\Delta P(t)$, which implicitly requires that the
 439 volatiles separate from the melt (i.e. perfect open system degassing with no gas remaining in the
 440 melt) as the magma ascends to the surface (Jaupart and Allègre, 1991; Menand and Phillips,
 441 2007; Woods and Koyaguchi, 1994), then the mass flow rate is given by

$$442 \quad \frac{dM}{dt} = \frac{\Delta P(t)}{fL} \quad (6)$$

443 Combining this relation with equation (3), it follows that the eruption rate will decay
 444 exponentially with time according to the relation (Huppert and Woods, 2002)

$$445 \quad \frac{dM}{dt} = \Delta P_c \exp\left(-\frac{fLt}{M(\beta + \beta_m)}\right) \quad (7)$$

446 assuming constant coefficients for the frictional resistance. Since typical fracture pressures are of
 447 order $10^6 - 10^7$ Pa (Touloukian et al., 1989), while the bulk compressibility is typically of order
 448 $10^{-8} - 10^{-10}$ Pa⁻¹ for volatile-saturated and unsaturated magma, the volume of melt which erupts in
 449 such conduit or dyke-controlled eruptions may range from a few percent of the total volume of
 450 the chamber to a fraction of a percent in volatile-poor magmas. Equation 7 illustrates that the
 451 eruption duration increases with the compressibility of the magma, with the eruption duration t
 452 scaling as

$$453 \quad t = \frac{M(\beta + \beta_m)}{fL} \quad (8)$$

454 and this is consistent with the increased mass erupted for a volatile-saturated magma. The
 455 eruption rate depends primarily on $\frac{\Delta P(t)}{fL}$ as indicated by equation 6.

456 This model of the overall long-term control on eruption evolution, although simplified, can be
 457 compared with a number of historical eruption episodes for which the erupted volume has been

458 measured as a function of time, including the eruptions of Soufrière, St Vincent (Huppert et al.,
459 1982) and Paracutin, Mexico (Fries Jr, 1953), where the eruption rate seems to follow an
460 exponential decay law with time (**figure 6a**; after (Berkowitz and Woods, 2018 under review)).
461 However, there is growing evidence that in some systems, there are multiple magma
462 reservoirs which can drive an eruption, and in particular, there is evidence that the eruption of
463 Eyjafjallosjokull in 2010 involved mobilization of magma from three different levels in the crust,
464 in sequence, with a shallower magma reservoir being tapped, followed by an intermediate and
465 then a deep magma reservoir (Tarasewicz et al., 2012). This interpretation of multiple reservoirs
466 is also consistent with the eruption of Lonquimay (Moreno and Gardeweg, 1989) in which the
467 eruption rate showed a more complex evolution, with an initially rapid exponential decay during
468 the eruption of the first 50% of the magma, followed by a more gradual exponential decay during
469 the second phase of eruption (**figure 6b**). This eruption rate trend perhaps indicates a two
470 reservoir system, both of comparable size, in which the shallow reservoir initially erupts and
471 decompresses. This is then followed by the onset of eruption from the deeper reservoir, driving
472 new magma into the shallow reservoir, and causing the eruption to persist until the two reservoirs
473 re-equilibrate. In this case, one can envisage a double reservoir model, with a rate constant for
474 flow from the shallow reservoir to the surface, and a second rate constant for the flow between
475 the deeper and shallower reservoir (Berkowitz and Woods, 2018 under review).

476
477 Other historical eruptions also suggest that the gradual decompression of a deep magma reservoir
478 exerts a long term control on the eruption rate. For example, the 1980-1985 eruption of Mt St
479 Helens involved a series of intermittent slow dome-building and short dome-explosion events
480 following the initial major eruption episode (Brantley and Myers, 2000). Analysis of reported
481 data on the eruption volume during this 5-6 year period suggests a slow exponential decay of the
482 time-averaged eruption rate (**figure 6c**). Although the actual eruption history was intermittent,
483 owing to the non-linear controls associated with the formation and failure of the dome (Barmin et
484 al., 2002; Melnik and Sparks, 1999), the long term build up of pressure in the dome, between
485 each event appears to have been controlled by the flux from a gradually waning deeper reservoir
486 (Berkowitz and Woods, 2018 under review). Indeed, by modelling the non-linear valve-like
487 dynamics of the conduit feeding the dome in terms of a small critical overpressure for failure and
488 eruption of a shallow reservoir, the control on the long-term behavior of the system can be
489 rationalised in terms of the decompression of a deep reservoir.

490
491 **4. The role of the exsolved volatile phase in magma mixing and mingling**

492 Exsolved volatiles play an important role in magma mixing and mingling in magma reservoirs
 493 (Bachmann and Bergantz, 2006; Degruyter and Huber, 2014; Edmonds et al., 2010; Huber et al.,
 494 2011; Huppert and Woods, 2002; Pallister et al., 1996; Phillips and Woods, 2002; Ruprecht and
 495 Bachmann, 2010; Woods and Cowan, 2009). Magma intrusion has often been cited as a trigger
 496 for eruption (Eichelberger, 1980; Martin et al., 2008; Pallister et al., 1992; Sparks et al., 1977),
 497 and there is frequently petrological and geochemical evidence of magma mixing and /or heating
 498 shortly before eruptions (Metrich et al., 1993; Murphy et al., 2000; Rae et al., 2016; Ruprecht et
 499 al., 2012). Magma mingling may manifest as mafic inclusions and banded and streaked eruption
 500 products (Coombs et al., 2000; Plail et al., 2018; Sparks et al., 1977; Steiner et al., 2012; Tepley
 501 et al., 1999; Watts et al., 1999); and magma mixing as hybrid magmas, formed by the efficient
 502 mixing of magmas prior to eruption (which may encompass the disaggregation and scavenging of
 503 crystal phases), commonly displaying disequilibrium textures and petrological features (Cassidy et
 504 al., 2015; Sparks and Marshall, 1986; Wright and Fiske, 1971). In this section we explore some
 505 general mechanisms, explored through analogue experiments and simple analytical models, by
 506 which the exsolved volatile phase may play a role in such processes, whereby intruding magma
 507 may interact with overlying magma, triggering mixing and eruption.

508

509 *4.1. Magma mixing and overturn, driven by vesiculation of underplating basalt*

510 If an intrusion of hot, relatively dense magma occurs below an existing body of cooled, evolved
 511 melt, the new lower layer of magma may inflate through cooling, crystallisation and exsolution
 512 of volatiles, but may remain dense relative to the overlying layer. It is possible, however, for this
 513 underplating magma to drive the eruption of the overlying magma (Burgisser and Bergantz,
 514 2011; Huppert et al., 1982; Martin et al., 2008; Murphy et al., 2000; Sparks et al., 1984). The
 515 control on the erupted volume will be associated with the expansion of the volatiles in the
 516 underplating melt body, even though it is the overlying magma which erupts. The mass of magma
 517 erupted depends on the relative volume of the upper and lower layers of magma, since both will
 518 expand as the chamber pressure falls. With the upper layer erupting, the volume erupted as the
 519 pressure falls by an amount ΔP is now given by

$$520 \quad \Delta V = (V_L \beta_L + V_u \beta_u + V \beta) \Delta P \quad (9)$$

521 where $V_L + V_u = V$, the total magma reservoir volume. It follows that as the fractional volume
 522 change of the new volatile-rich magma (V_L) increases, the mass erupted also increases, even if

523 this is the less dense, overlying, degassed magma (V_u). This is because the mass erupted is
524 creating space for the expansion of the volatile-rich magma which is much more compressible
525 than the overlying magma.

526

527 The reduction in the density of the underplating body of magma, caused by cooling and
528 crystallization and associated exsolution of volatiles, may eventually cause this magma to
529 become less dense than the original magma in the reservoir. As a result, there may be an overturn
530 or mixing event, driven by degassing (Huppert et al., 1982; Ruprecht et al., 2008; Sparks et al.,
531 1984; Woods and Cowan, 2009). If the new, initially dense intrusion is relatively thin and of low
532 viscosity compared to the original melt, then the plumes of this low viscosity melt will be thin
533 and may become stirred into the overlying melt, especially if that melt is convecting. This can
534 lead to formation of mafic inclusions or an intermingled melt (Phillips and Woods, 2002). In
535 contrast, with a deeper or more viscous underplating magma, there may be a more complete
536 overturn, with plumes of the underplating magma being larger and accumulating at the top of the
537 overlying magma. In this case, on eruption, the crystalline mafic magma may be the dominant
538 phase which issues from the system. The intermingling, or mixing, produced by pre-eruption
539 cooling and crystallization has been explored in a series of experiments in which an increasing
540 volume of bubbles were created by electrolysis in a layer of viscous saline solution overlain by a
541 less dense, viscous bubble-free solution (**figure 7**) (Woods and Cowan, 2009). The experiments
542 illustrate the important control of the viscosity contrast between the melts as well as the relative
543 depths of the magma layers on whether there is stirring and mixing or overturn.

544

545 It is also possible that, during an eruption, the reduction in the pressure in the magma chamber
546 can lead to a reversal in the relative density of the two magmas (Ruprecht et al., 2008; Woods
547 and Cowan, 2009). This may lead to a change in the composition of the erupting magma during
548 the eruption provided that the eruption is sufficiently long-lived for overturn to occur. This
549 mechanism may provide an explanation for the heterogeneous chemical and textural
550 characteristics of crystal cargoes erupted in hybrid magmas, which suggest complex magmatic
551 mixing histories (Davidson and Tepley, 1997; Ruprecht et al., 2008; Singer et al., 1995). With a
552 typical density contrast of order 10-100 kg/m³, magma of viscosity 100-1000 Pas and of depth
553 10-100 m, the time scale for overturn would be of order 10³-10⁴ s and this requires the eruption to
554 persist for a time of order hours, as is the case in larger explosive eruptions.

555

556 Woods and Cowan (2009) carried out a series of analogue experiments in which a volatile-rich
557 layer of saline and viscous natrasol solution underlay a volatile poor layer of relatively fresh but
558 viscous natrasol solution. The volatile phase was CO₂ dissolved in tap water. As the pressure is
559 reduced to pressures of order 0.01-0.1 of atmospheric pressure, the volatiles exsolve, and, in the
560 viscous solution, they form a bubble-rich layer. In the experiment, the upper solution was
561 generated from previously decompressed water in order to remove the volatiles prior the
562 experiment, and the salt added to the lower layer produced an initially density-stratified system.
563 As the system decompresses and the lower layer becomes less dense, the layers overturned. In a
564 further, unpublished experiment, an enclosed two layer system was connected to an upper tank
565 through two conduits providing a pathway for each of the layers to vent from the chamber, one
566 from the lower and one from the upper layer of system. The whole system was then enclosed in
567 sealed tank connected to the vacuum pump, Initially, as the system reached the pressure at which
568 some volatiles were exsolved from the lower layer, this layer expanded and an eruption began,
569 with the dense volatile-rich lower layer erupting through the lower conduit (right hand conduit,
570 figure 7) and the less dense volatile-poor solution erupting from the upper conduit (left hand
571 conduit, figure 7). However, as the experiment persisted and more volatiles were produced in the
572 lower layer, the density of the lower layer fell below that of the upper layer, and there was a
573 short-lived overturn event in the experimental tank. At this stage, the solution erupting from the
574 two conduits switched, as shown in the last panel in **figure 8**. The withdrawal of a lower viscosity
575 lower magma through an upper more viscous layer during eruption was also considered by
576 (Blake and Ivey, 1986).

577

578 4.2. *Vesiculation of underplating magmas and formation of mafic enclaves*

579 In the above discussion we have assumed that there is no separation of the bubbles from the melt-
580 crystal-bubble mixture, and that the melt-crystal mixture behaves as if it is a single fluid phase.
581 This leads to the idealised bulk flow behaviour described above. Although this is a powerful end-
582 member picture of the possible controls on overturn and eruption, the possible separation of the
583 volatile phase from the melt may influence such mixing processes and also the evolution of an
584 eruption and the eruption products and we now explore such effects, considering the impact of
585 bubble separation from a crystal-poor melt (Bottinga and Javoy, 1991; Jaupart and Vergnolle,
586 1989; Menand and Phillips, 2007; Woods and Cardoso, 1997).

587

588 In a low crystallinity melt, gas bubbles may rise. There are two modes of separation. First, the
589 bubbles can rise individually, following Stokes law, or a modified Stokes law including effects of

590 hindered ascent. Typically, for the bubbles that exist in magma reservoirs in the crust, this leads
 591 to rather slow rise speeds of order $10^{-10} - 10^{-6}$ m/s, depending on the bubble size and the melt
 592 viscosity. In addition to the ascent of individual bubbles, the presence of bubbles can lead to a
 593 convective flow, in which variations in the concentration of the bubbles through the melt lead to
 594 density differences and convective mixing of the melt (Cardoso and Woods, 1999). Cardoso and
 595 Woods (1997) showed, through experiment that this latter process stirs the melt and bubbles, but
 596 also leads to separation of the bubbles from the melt in the upper boundary layer of the flow
 597 through bubble-liquid slip.

598
 599 In comparing the two processes, the ratio of the idealized Stokes speed of a bubble of size b and a
 600 parcel of fluid of size B with excess bubble concentration Δc relative to the surrounding melt is
 601 given by

$$602 \quad \frac{V_b}{V_B} = \frac{b^2}{B^2 \Delta c} \quad (10)$$

603 where the Stokes speed is given by $V_b = \frac{2\Delta\rho g b^2}{9\mu}$. Since the bubbles have typical sizes of
 604 order $b = 0.1-1.0$ mm in the chamber, the bubble rise speed is quite small, of order $10^{-10} - 10^{-6}$
 605 m/s in magma with viscosity of order $10^2 - 10^4$ Pa s, and so if there are small fluctuations in the
 606 concentration of bubbles through the melt layer, for example of magnitude $\Delta c = 0.0001-0.01$, the
 607 convective rise speed of parcels of bubble-rich melt will exceed the individual bubble rise speed
 608 provided the length scale of the parcel, B is greater than $(10-100) b \sim 0.1-10$ cm. If there are
 609 concentration fluctuations over greater length scales, as is very likely in a magma reservoir of
 610 scale 0.1 to 1.0 km, then convective mixing may dominate the individual rise of bubbles through
 611 the melt. Although convective mixing will tend to stir the bubbles throughout the melt, leading to
 612 a reasonably well-mixed suspension, near the top of the melt layer bubbles may separate from the
 613 melt and become trapped in a viscous boundary layer. The time scale for loss of bubbles from the
 614 main body of melt into this boundary layer scales with $\frac{H}{V_b}$, where H is the depth of the magma
 615 layer (Cardoso and Woods, 1999; Thomas et al., 1993; Woods and Cardoso, 1997). As a result, in
 616 either the case of individual bubble ascent or convective separation, the bubble-melt separation
 617 occurs over times which scale as $\frac{H}{V_b}$. With bubbles of size 0.1-1.0 mm in a magma of viscosity
 618 10^2-10^4 Pas, the time to separate from a 100 m thick layer of melt is therefore in the range $10^8 -$
 619 10^{12} s. We infer that, with small bubbles in the more viscous melt, the time for separation may be
 620 relatively long (30,000 years) compared to the time scale of cooling and crystallisation of the

621 melt. In this case, cooling and crystallization will lead to further exsolution of volatiles and a
622 build up of the chamber pressure which will tend to trigger an eruption of the bubble-laden melt.
623 In addition, the accumulation of bubbles in the lower layer of melt will lower the density of this
624 layer and tend to promote mixing or overturn of the magmas.

625
626 In contrast, for larger bubbles or a less viscous melt, bubble separation may occur within years,
627 and so may be a dominant process inhibiting the tendency for large-scale overturn of the magma
628 reservoir. Indeed with more primitive magmas, which have a low viscosity, bubble-melt
629 separation can lead to bubble accumulation at the top of the magma body and the formation of a
630 foam or bubble-rich layer. This is a mechanism proposed to be important for volatile exsolution
631 in basaltic sills in the upper crust (Menand and Phillips, 2007), and in shallow reservoirs e.g., at
632 Kilauea Volcano (Edmonds and Gerlach, 2007). If the system is closed, then the pressure will
633 increase by a similar amount to that shown in the calculation above (*section 3.2*), in which the
634 volatiles in the reservoir are assumed to remain well-mixed. However, in a reservoir of
635 significant vertical extent there may be a significant additional pressurisation associated with the
636 migration of the bubbles to the top of the chamber and the associated decrease in magmatic
637 pressure experienced by the bubbles. For example, in a chamber of order 1 km in vertical extent,
638 bubble separation may lead to an additional overpressure of up to 1-10 MPa, but the effect is
639 smaller in chambers of smaller vertical extent (Woods and Cardoso, 1997). If the magma has 0.5-
640 1.0 wt% CO₂ and <0.5 wt% H₂O, and the reservoir is deeper than 2 km, the exsolved volatile
641 phase will be enriched in CO₂ relative to H₂O. Leakage of this exsolved volatile phase from the
642 roof zone of the magma reservoir may cause broad regions of diffuse CO₂ degassing inside
643 calderas, as is observed at Kīlauea Volcano, Hawai‘i (Gerlach et al., 2002) and at many other
644 active volcanoes exhibiting diffuse degassing between and during eruptive episodes (Salazar et
645 al., 2001; Viveiros et al., 2010).

646
647 In some cases, the separation of the volatiles from the melt may then lead to a more continuous
648 ‘open’ style of degassing of the volcanic system, either through Strombolian-style gas bursts.
649 This can serve to regulate the pressure in the chamber and may tend to suppress larger, explosive
650 eruptions. It has been proposed that if a foam layer develops at the top of a chamber or layer of
651 basaltic melt, liquid drainage through the foam can lead to merger of the bubbles in the foam, and
652 formation of larger gas slugs which may be responsible for larger Strombolian gas burst type
653 eruptions (Houghton and Gonnermann, 2008; Houghton et al., 2016; Jaupart and Vergnolle,
654 1988; Vergnolle and Jaupart, 1986).

655

656 In *section 4.1* we discussed the possibility of a large scale overturn of the system if bubbles are
657 exsolved in the lower layer of melt and this layer of melt is sufficiently viscous to retain the
658 majority of the bubbles until its density falls below that of the upper layer. However, in some
659 systems, there will be partial separation of the gas phase from the lower layer of melt, as
660 described above, and this can lead to accumulation of a foam at the interface between the two
661 layers of melt, especially if the upper layer is more viscous. If a sufficient fraction of the bubbles
662 separate from the lower layer of melt, it may remain denser than the upper layer, suppressing any
663 large scale overturn, while the foam forming at the interface between the layers will typically be
664 less dense than the overlying layer of melt, leading to convective instability and ascent of parcels
665 of foam into the overlying layer of melt. The percolation of a hot exsolved volatile phase from a
666 mafic underplating layer upward into cooler, crystal-rich magma body has been proposed as a
667 mechanism by which cool near-solidus magmas may ‘defrost’, triggering eruption (Bachmann
668 and Bergantz, 2006; Huber et al., 2011; Huber et al., 2010). The exsolved volatile phase may also
669 carry some of the mafic melt into the upper layer, leading to the formation of mafic inclusions in
670 the upper layer (Andrews and Manga, 2014; Bacon, 1986; Eichelberger, 1980; Plail et al., 2014).
671 These may eventually erupt if the overall pressure of the system becomes sufficient to trigger
672 eruption of the upper layer (Phillips and Woods, 2002; Thomas et al., 1993). Good evidence for
673 the accumulation of a foam layer at the interface between underplating basalts and overlying
674 crystal-rich andesite comes from the observation, from enclaves erupted in Soufrière Hills
675 Volcano andesite, that the enclaves are enriched in magnetite, interpreted to be due to the
676 nucleation of aqueous fluid bubbles onto magnetite, which then “floated” up to the interface
677 (Edmonds et al., 2014a).

678

679 Recently, it has been found that some mafic (basalt to basaltic andesite) inclusions (hosted in
680 crystal-rich andesites at Soufrière Hills Volcano, Montserrat) are themselves composed of a
681 mixture of the upper and lower melt (Plail et al., 2018), and this suggests that there is some
682 mixing or intermingling of the magmas in either the foam layer or the lower layer prior to the
683 formation of the mafic inclusions in the upper layer. This observation is common to many
684 andesites containing mafic inclusions, such as the erupted products of Quizapu (Ruprecht et al.,
685 2012) and Unzen, Japan (Browne et al., 2005). This intermingling of the magmas prior to
686 formation of a mafic inclusion may arise if crystals in the upper layer become stirred into the
687 lower layer. These are observed in the enclaves as plagioclase crystals with sieve textures and
688 resorbed rims that have undergone heating after being mixed into more mafic melt (Humphreys

689 et al., 2009), which is a common feature of many mafic inclusions hosted by andesites and
690 dacites (Leonard et al., 2002; Tepley et al., 1999). As the lower layer becomes progressively
691 more crystalline and volatile-rich, a foamy layer may develop at the interface, carrying some of
692 the hybrid interface magma up with the bubbles into the upper layer. Alternatively, when the
693 mafic magma ponds below the existing layer of cooler and less dense melt, the heat transfer
694 across the interface may lead to formation of a thin layer of relatively hot and mobile original
695 magma and a corresponding thin layer of cooled, more crystalline and volatile-rich new magma
696 which supplies the heat to the upper thin layer. Eventually, the lower cooled layer exsolves some
697 volatiles, becomes less dense and intermingles with the heated boundary layer of original magma.
698 This local overturn can produce intermingling and a hybrid layer of melt. As the heat transfer
699 continues, the boundary layer becomes progressively deeper, enabling buoyant plumes from the
700 intermingled boundary layer to rise into the upper, cooler layer of more viscous melt. As the
701 boundary layer becomes deeper, mafic enclaves may become larger. In this multistage fashion,
702 we can envisage that mafic inclusions of intermediate composition may be generated in the upper
703 layer (Plail et al., 2018).

704
705 Whether wholesale overturn of magmas occurs, or whether gas-rich buoyant plumes (or even
706 fractures, see next section) are generated from the interface between magmas, it seems clear that
707 underplating basaltic magmas may supply significant quantities of exsolved volatiles to overlying
708 magma reservoirs. Owing to the high sulfur content at sulfide saturation of basaltic melts (Jugo,
709 2009; Ripley and Li, 2013) it has been suggested that mafic underplating magmas may be an
710 important source of the sulfur observed outgassing during volcanic eruptions via addition of
711 sulfur-rich magmatic exsolved volatile phase to the overlying more evolved magma (Edmonds et
712 al., 2010; Pallister et al., 1992; Roberge et al., 2009). Underplating mafic magmas, under some
713 circumstances, may also play a role in generating more CO₂-rich magmatic vapor which may flux
714 through shallower crystal-rich magmas (Blundy et al., 2010). The supply of volatiles from
715 underplating mafic magmas has also been linked to the formation of copper porphyries. While
716 crystal-rich, evolved magmas have capacity for producing the mass of copper required for a
717 viable economic deposit, it has been suggested that additional sources of sulfur may be necessary,
718 which may be supplied by mafic magmas intruding the evolved magma reservoir (Blundy et al.,
719 2015; Hattori and Keith, 2001).

720

721 **5. Exsolved volatile phase generation and transport through crystal-rich magma bodies**

722 Magma bodies may spend much of their time in the crust near their solidus temperature, in a
723 partially or entirely crystalline state (Cashman et al., 2017; Cooper and Kent, 2014; Huber et al.,
724 2011; Huber et al., 2009; Huber et al., 2010; Koyaguchi and Kaneko, 1999; Marsh, 1981).
725 Volatile-rich basalts rising up into the crust will tend to underplate and interact with such
726 crystalline magma bodies (Bachmann and Bergantz, 2006; Edmonds et al., 2014b; Huber et al.,
727 2011), as discussed above. In this section we review and explore the range of behaviours possible
728 for storage and transport of the exsolved volatile phase in a crystal-rich mush and discuss the
729 implications for gas storage in and/or segregation from a mush, large syn-eruptive sulfur
730 emissions, ore deposits and the role of basaltic underplating in supplying volatiles to large, long-
731 lived crystalline magma bodies.

732

733 *5.1. Transport of an exsolved volatile phase in crystal-rich magma*

734 An underplating basalt will tend to pond and spread out beneath a cooler, less dense, more
735 evolved magma. The basalt will cool and crystallise against the cooler magma, generating a
736 buoyant volatile phase that may accumulate at the interface or rise upward, as described in
737 section 4. The rate of volatile injection into the overlying crystal-rich magma depends on the
738 volatile content of the melt, the cooling rate and the convective motion of the underplating
739 magma (Cardoso and Woods, 1999; Huppert et al., 1982). The accumulation of an exsolved
740 volatile phase at the interface may cause a rheological instability, eventually leading to the foam
741 layer becoming buoyant and rising upward into the crystal-rich magma. It has been proposed that
742 the migration of this exsolved volatile phase into the overlying magma may remobilise the
743 crystal-rich magma due to the transfer of heat and partial melting (Bachmann and Bergantz,
744 2006; Huber et al., 2011; Huber et al., 2010). Reactivation and eruption of crystal-rich magmas
745 may be triggered in this way (Couch et al., 2001; Murphy et al., 2000; Pallister et al., 1992), with
746 the underplating basalt supplying volatiles which may be outgassed during the eruption
747 (Christopher et al., 2010; Edmonds et al., 2010).

748

749 As magma evolves and becomes progressively more crystalline, it is able to support a yield stress
750 and form a crystal framework (Caricchi et al., 2007; Lejeune and Richet, 1995; Pistone et al.,
751 2012) which tends to suppress the convective mixing or the ascent of bubbles. Such effects may
752 begin with crystal volume fractions of 0.2-0.3, depending on the shape and morphology of the
753 crystals, and as the crystal content increases, the yield stress increases (Pistone et al., 2012). In
754 addition, the viscosity of the crystalline mixture tends to be very much larger than the pure melt,
755 suppressing shear motions in the suspension. Recent experimental work has shown, however, that

756 the presence of hydrous fluid bubbles in a mush may have significant effects on its rheology. For
757 mushes with crystal fractions of 0.55 to 0.7, for example, addition of volatiles such that bubble
758 volume fractions of up to 0.1 are generated, leads to a dramatic reduction in viscosity by 3-4
759 orders of magnitude (Pistone et al., 2013). Furthermore, the viscosity of such three phase systems
760 may be strain rate-dependent, with both shear thinning and shear thickening behaviour observed
761 under different strain rates and crystal contents (Pistone et al., 2013).

762

763 As a result of this complex rheology, the process of gas separation from the crystal-rich magma
764 takes on a range of forms. In a crystalline matrix, the migration process may be associated with
765 either (a) gas flow through the porous matrix, which is likely to be a relatively slow process,
766 owing to the low permeability of the formation (Parmigiani et al., 2016; Parmigiani et al., 2014)
767 or (b) the generation of small effective fractures or cracks in the crystal-framework as the
768 pressure exceeds the yield stress or effective fracture stress (Barth et al., in review; Huber et al.,
769 2011). Such cracks then provide much higher permeability pathways through which an exsolved
770 volatile phase may flow (Oppenheimer et al., 2015).

771

772 In more crystalline melts, these transport processes, which may be relatively rapid, may be
773 responsible for the separation of the exsolved volatile phase from the melt and its accumulation at
774 the roof zone, or in liquid-rich lenses in the magma reservoir system, where they may be held up
775 (Parmigiani et al., 2016). These accumulations of exsolved volatiles may lead to large emissions
776 of volcanic gases during eruptions. If the exsolved volatile phase at this pressure has a significant
777 concentration of sulfur (Oppenheimer et al., 2011; Scaillet and Pichavant, 2003) then this
778 mechanism of gas extraction may rationalise the high sulphur dioxide emissions from, for
779 example, large explosive eruptions such as El Chichon (Mexico, 1982) (Bluth et al., 1997) and
780 Pinatubo (Philippines, 1991) (Wallace and Gerlach, 1994). It is also likely that volcanoes which
781 exhibit long-lived and persistent degassing, both during and between eruptions, may be tapping
782 regions of crystal-rich magma reservoirs that have accumulated exsolved volatiles (Christopher et
783 al., 2015; Edmonds et al., 2003).

784

785 In a series of experiments to model the *in situ* production of an exsolved volatile phase in a
786 cooling and crystallising melt, a series of glass balls were placed in a tank filled with polymer-
787 rich liquid and the mixture was gradually decompressed (Ptacek et al., 2018, sub judice). As the
788 mixture reached the vapour-saturation pressure, volatiles gradually came out of solution and as
789 the pressure was slowly decreased, the pack of beads gradually inflated. As this dilation occurred,

790 the effective permeability of the particle pack increased substantially and the gas was able to
791 migrate through the interstices between the glass beads, forming small gas channels in the matrix.
792 Once the gas escape rate exceeded the bubble generation rate, the expansion of the system was
793 arrested and indeed the gas-derived dilation reduced (panel 3, **figure 9**). In this way the gas was
794 able to separate from the melt-crystal mixture, and the pack relaxed back to a depth at which the
795 remaining gas phase was trapped as individual bubbles in the interstices between crystals, as may
796 be seen in panel 4 of **figure 9** (Ptacek et al., 2018, sub judice).

797

798 The processes controlling the rate of migration of gas through such a deformable permeable
799 matrix may involve both a regime of slow capillary gas flow through the permeable matrix
800 (Parmigiani et al., 2016) and a regime of more rapid gas flow following deformation of the pack
801 in which high permeability channels are created (Oppenheimer et al 2016). In the analogue
802 system of a clay-water mixture, the continued supply of gas to a clay-filled elastic reservoir may
803 lead to episodic pressurisation of the reservoir, followed by fracture of the clay seal, gas release
804 and build up of a further pressurisation cycle (Rocco et al., 2017). This process may be
805 analogous to the intermittent gas-escape cycles which develop in a magmatic system as gas
806 pressure builds up and eventually exceeds the yield strength of the viscous-crystalline melt,
807 driving a pure gas channel through the particle-pack and allowing the system to decompress. A
808 similar phenomenon arises when gas is supplied from below to a close-packed mixture of
809 particles suspended in a high-viscosity liquid (Barth et al., in review). The gas tends to
810 accumulate in localised pockets within the deformable liquid-crystal pack. As the pressure builds
811 up, it is eventually able to generate a small fracture which provides a pathway through the
812 system, allowing the gas to continue rising through the formation, until the overpressure is
813 dissipated and the gas builds up a new pocket higher in the system.

814

815 These experiments of the migration of gas through a highly crystalline permeable matrix,
816 partially filled with melt, leads to a different mechanism of gas separation from underplating
817 basalts as the melt cools and crystallises. For a typical crystalline matrix, which is poorly
818 consolidated, the fracture strength or yielding strength may be of order 10^5 - 10^6 Pa (Saar et al.,
819 2001), which is smaller than the strength of a solid rock. As exsolved gases form and pressurize
820 the system they will therefore be able to open up fractures within the basalt along which they may
821 escape. We deduce that if a new intrusion of basalt cools and crystallises to the extent that it
822 becomes immobile or develops a yield stress while remaining denser than the overlying layer of

823 melt, then as volatiles are exsolved from this layer of melt, these volatiles can be transported via
824 fractures into the overlying original magma, as discussed by Huber et al., (2012).

825

826 A further phenomenon associated with gas migration through crystalline magmas is that, once the
827 main pulse of gas has passed through the magma, there will be a small fraction which is retained
828 in the pore spaces by capillary effects, and this will lead to the magma remaining in a more
829 compressible state than the pure volatile-free melt phase. Typically the residual saturation of gas
830 in a porous layer depends on the grain size and the interfacial tension between the solid minerals
831 and the melt phase. More experimental data is required to constrain these gas fractions, which
832 depend on the wetting angle of the gas and crystals relative to that of the melt and crystals.

833

834 The exsolved volatile phase may also play a role in magma differentiation and crystal-liquid
835 segregation in undercooled magmatic intrusions and in thin sheets of primitive magma
836 underplating cooler magma reservoirs: as the magma cools and crystallises, the buildup of gas
837 pressure due to second boiling may be relieved by expulsion of melt out of the region of
838 crystallization (Anderson Jr et al., 1984; Sisson and Bacon, 1999).

839

840 *5.2. Implications of our new understanding of the transport of the exsolved volatile phase*
841 *through crystalline magmas for ore deposits*

842 The exsolved magmatic volatile phase is understood to play a critical role in the formation of
843 porphyries which contain economic grades of metal deposits (Hedenquist and Lowenstern, 1994;
844 Lowenstern et al., 1991; Williams-Jones and Heinrich, 2005). Recent advances, discussed above,
845 in understanding how the exsolved volatile phase may be retained, and/or transported through
846 crystal-rich magma bodies have important implications for models of ore deposit formation and
847 for understanding why some magma reservoirs yield metal accumulations of economic
848 importance and others do not. As discussed in section 2, magma may saturate in an exsolved
849 volatile phase during decompression, or during cooling and crystallization. Depending on the
850 initial H₂O and chlorine contents of the melt, and the pressure and temperature, a brine phase
851 may also form (Webster, 1997, Metrich and Rutherford, 1992, Webster, 1992, Shinohara, 1994).
852 The chlorinity of the exsolved volatile phase is critical for its metal-carrying capacity (Halter and
853 Webster, 2004). The exsolved volatile phase may migrate to the upper reaches of the magma
854 reservoir system through channelised flow through a crystal-rich magma, perhaps accumulating
855 in pockets before rising further up through the mush, as discussed in section 5. Over time, the
856 lower density exsolved volatile phase will accumulate in the upper zones of the reservoir, or in

857 liquid lenses, where fracturing cannot occur (Parmigiani et al., 2016). This process of fluid
858 migration through quasi-brittle fractures may be much faster than by bubble rise through
859 permeable flow or through magma convection (Lowenstern, 1994; Shinohara et al., 1995). This
860 therefore provides an efficient mechanism for segregation and accumulation of metals carried by
861 saline, low density, exsolved fluids at the roof zones of mushy magma reservoirs (**figure 10**).

862
863 The “tonnage” of metals that may be accumulated in this way might then be expected to be
864 proportional to the water content of the magma, which will determine the mass fraction of
865 exsolved volatile phase, its chlorinity, which will depend on the chlorine content of the melt,
866 temperature and pressure, the overall size of the magma reservoir and perhaps just as importantly,
867 the crystallinity and structure of the reservoir. Extensive mush zones will allow rapid and
868 efficient transport of metal-rich magmatic vapor to the roof zones of magma reservoirs, where
869 excess pressure there will promote the intrusion of stocks and/or the triggering of volcanic
870 eruptions.

871
872 For example, porphyries hosting molybdenum (Mo) deposits are formed from highly evolved,
873 crystal-rich magmas, whereby Mo partitions from the silicate melt into a co-existing single fluid,
874 at pressures above the critical point, with a fluid-melt partition coefficient of 5-20 (Audetat and
875 Li, 2017; Zajacz et al., 2008). At lower pressures or temperatures, as the solidus is approached,
876 this fluid may condense a brine phase. There is abundant evidence for extensive fluid transport at
877 magmatic conditions: Mo mineralization commonly occurs only in the uppermost parts of
878 vertically elongate magma bodies. The available data from a recent review of a number of Mo
879 porphyries suggest that the parent magmas of such bodies were not particularly Mo-rich and the
880 fluids from both mineralising and barren magma bodies are similar in terms of their geochemistry
881 (Audétat and Li, 2017; Lowenstern, 1994), suggesting that instead, magma volume and depth, as
882 well as the mechanism of volatile extraction from the mush, may be the most important factor.
883 Previous workers have suggested magma convection and open system degassing as a mechanism
884 for extracting the fluid phase from a large body of magma (Lowenstern, 1994; Shinohara et al.,
885 1995). However, following recent work, as discussed above, on the mechanisms of porous flow
886 and fracturing of crystal-rich magma, we now understand that fluid extraction from large-scale
887 mush systems may take place efficiently on rapid timescales, thereby building accumulations of
888 metals at the roof zones of large crystal-rich magma bodies (Huber et al., 2012).

889
890 **6. Summary and problems for the future**

891 In this review we have described some of the complex processes associated with the dynamical
892 interactions of melt, crystals and volatiles. We show that most magmas, particularly those in
893 subduction zone settings, are in equilibrium with an exsolved vapor phase through much of the
894 evolution from the mid-crust to the surface and this exsolved volatile phase plays a fundamental
895 role in magma differentiation, eruption triggering and the formation of hydrothermal ore deposits.
896 We have distinguished between low crystal content, fluid-like systems in which the gas may
897 drive convection in the melt, and higher crystal content, mushy systems, in which the gas may
898 migrate by permeable flow or along fractures through the mush. We have also emphasized the
899 role of exsolved volatiles in regulating the density and pressure of the magma and the chamber,
900 with the exsolved volatile phase constraining the compressibility of the magma reservoir and the
901 volume of melt erupted, given the change in volume of the magma chamber. Density changes in a
902 stratified magma reservoir can lead to overturn or mixing and the formation of mafic inclusions
903 especially in the case of fluid-like melt; whereas with a more crystalline system, density reversals
904 may be less effective in that the convective overturn requires a buoyancy-derived stress to exceed
905 the yield stress of the crystal-pack. However, if a series of fractures or channels open up to allow
906 the exsolved volatiles to leak from the melt-crystal mixture, this can stabilise the vertical density
907 profile of the magma reservoir over time. This mechanism of exsolved volatile phase migration
908 to the apical zones of a magma reservoir over time can reconcile observations of large and/or
909 persistent sulfur dioxide fluxes from volcanoes, as well as rapid timescales for the formation of
910 hydrothermal ore deposits (e.g Cu porphyry-type deposits). Future work in this area might
911 involve the use of volatile radionuclides at actively degassing volcanoes to establish the likely
912 timescales of exsolved volatile accumulation in magma reservoirs.

913
914 There have been a series of key experiments measuring the shear stress in crystalline and crystal-
915 bubble mixtures subjected to shear (Pistone et al., 2013), and some more recent experiments
916 exploring the migration of gas through crystal-melt suspensions (Barth et al., in review;
917 Oppenheimer et al., 2015; Ptacek et al., 2018, sub judice), but there is much more to learn about
918 the macro-scale dynamics of the bulk fluid and how this interacts with the local dynamics of gas-
919 melt separation, which can be facilitated through local deformation and fracture formation. Field
920 observations of deformed bubbles and apparent shear bands in which melt is localized provide
921 critical clues about the local processes of deformation and their relation to the large scale flow.
922 Such observations suggest that there may also be critical effects associated with the varying
923 compressibility of the magma as it ascends through the crust, and decompresses, perhaps leading
924 to waves of volatile-rich and volatile-poor magma, connected by local shear bands or fractures.

925 The physical processes controlling crystal-rich magmas offers an area ripe for novel laboratory
926 experiments, and these, combined with new field observations and careful geochemical analysis
927 may be key in future efforts to unravel the history of co-erupted gas, melt and crystals.

928

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931

932

933 **References**

934

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- 1508

1509

1510 **Figure captions**

1511

1512 **Figure 1:** Schematic diagram to illustrate how magmas may achieve volatile-saturation. A:
 1513 decompressing silicate melt achieves saturation in an exsolved volatile phase, which promotes
 1514 degassing-induced crystallisation. B: Isobaric crystallisation in a crustal magma reservoir causes
 1515 second boiling and development of an exsolved volatile phase. MVP: (exsolved) magmatic
 1516 volatile phase.

1517

1518 **Figure 2:** Solubility model for rhyolite (Newman and Lowenstern, 2002), showing melt CO₂
 1519 concentration versus melt water concentration. Isobars are in black solid lines, and represent the

1520 locus of values for dissolved H₂O and CO₂ in rhyolitic melt in equilibrium with H₂O–CO₂ vapor
1521 at 800C and selected pressures (labelled, in MPa). Isoleths are dashed lines, and represent the
1522 locus of rhyolitic melt compositions in equilibrium with the given exsolved volatile phase
1523 compositions: 20 mol% H₂O (80 mol% CO₂), 50 mol% H₂O and CO₂; and 80 mol% H₂O (20
1524 mol% CO₂) at 800 C. The orange and red dots show the behaviour of vapour-undersaturated and
1525 vapor-saturated melts (respectively) with (top orange dot) and without (bottom orange dot) CO₂.
1526 These examples are discussed in the text.

1527

1528 **Figure 3:** Evolution of melt H₂O content (red, in wt%) and total fraction of exsolved water (blue,
1529 in wt%) during crystallization from basalt to rhyolite, at 200 MPa, illustrating how substantial
1530 fractions of exsolved volatiles may be generated during isobaric crystallization. Curves are
1531 labelled with the initial basalt melt H₂O content, in wt%. If the evolved liquids and exsolved
1532 volatiles are segregated from their crystalline products, the fraction of exsolved volatiles in the
1533 magma (melt) is much higher than shown. For magma crystallization at lower and higher
1534 pressures, fractions of exsolved volatiles reach higher and lower values respectively (due to the
1535 lower and higher solubilities of water, respectively). Calculations are made using RhyoliteMelts
1536 (Ghiorso and Gualda, 2015).

1537

1538 **Figure 4:** Melt inclusion CO₂ and H₂O concentrations; and isobars to show volatile
1539 concentrations in equilibrium with an exsolved volatile phase for different pressures (marked). A:
1540 Volatile data for basaltic melt inclusions, hosted by olivine. Isobars are appropriate for Canary
1541 Island basalt (Burgisser et al., 2015). Data are shown for Fernandina and Santiago from (Koleszar
1542 et al., 2009); for Kīlauea from (Sides et al., 2014b), for El Hierro from (Longpré et al., 2017) for
1543 Laki (Hartley et al., 2014) , for Holuhraun (Bali et al., 2018), for Stromboli (Métrich et al., 2009),
1544 for Etna (Spilliaert et al., 2006) for Llaima (De Maisonneuve et al., 2012; Ruth et al., 2016) and
1545 for Fuego (Berlo et al., 2012; Lloyd et al., 2016; Roggensack, 2001; Sisson and Layne, 1993). B:
1546 Data for dacitic and rhyolitic melt inclusions, hosted by plagioclase and quartz. Isobars
1547 appropriate for rhyolite (Newman and Lowenstern, 2002). Data are shown for Pine Grove
1548 (Lowenstern et al., 1994), Soufrière Hills (Edmonds et al., 2014b), Bishop Tuff (Wallace et al.,
1549 1999), Oruanui, Taupo (Liu et al., 2006), Rabaul (Roggensack et al., 1996), Popocatepetl (Atlas
1550 et al., 2006) and Satsuma-Iwojima (Saito et al., 2001).

1551

1552 **Figure 5.** The effects of an exsolved volatile phase on magma compressibility and how it may
1553 evolve during magma fractionation in a magma reservoir. A) Effective compressibility of a melt-

1554 volatile mixture (with no crystals) in the case that there is 2, 4 and 6 wt% H₂O in the melt at 1200
1555 K. B) The dependence of the compressibility of a melt-crystal-volatile mixture on the crystal
1556 content of the melt. As the magma cools and crystallises, the melt becomes volatile-saturated and
1557 the compressibility rapidly increases beyond that for the solid rock. Curves are shown for melt
1558 with a H₂O content of 3, 4 and 6 wt%, in a reservoir of depth 5 km, with magma temperature
1559 varying with the crystal content of the melt. C) Rate of change of the reservoir pressure during
1560 second boiling as a function of the change in temperature, normalised to the critical pressure
1561 required to trigger an eruption, here taken to be 10 MPa (Blake, 1984). Curves are shown for H₂O
1562 contents of 3, 4 and 6 wt%.

1563

1564 **Figure 6.** Data on the cumulative mass erupted from (A) Soufriere St Vincent and (B) Paracutin,
1565 as a function of time, compared to the exponential model for the gradual waning of the eruption
1566 rate; (C) Lanquimay, compared to a model for the mass erupted from a two magma chamber
1567 system, with the dashed line illustrating the erupted mass from the deeper chamber into the upper
1568 chamber, and the black line the erupted mass from the upper chamber to the surface. The lower
1569 chamber only starts to supply a significant flux of magma once the upper chamber has
1570 decompressed. (D) May St Helens, after May 18 1980 until 1985. Here the symbols represent the
1571 incremental erupted volume during successive dome explosion events; the black line is a model
1572 with an idealized steadily waning eruption rate and the dashed line is the model in which each
1573 explosive event occurs once a prescribed overpressure has been reached (for details see
1574 (Berkowitz and Woods, 2018 under review)).

1575

1576 **Figure 7:** Illustration of the process of overturn associated with volatile exsolution from a melt
1577 prior to an eruption. A: the lower layer of melt is deep and viscous, leading to a dominant plume
1578 of bubble-rich buoyant melt rising up through the overlying volatile poor melt, leading to large
1579 scale overturn. B: the lower layer is less viscous and shallower, leading to smaller plumes of
1580 bubble-laden buoyant melt rising up into the overlying layer of volatile-poor melt. These plumes
1581 become unstable and are mixed into the overlying layer of melt rather than leading to a dominant
1582 overturn event (Woods and Cowan, 2009). The vertical scale of the experiments is ~ 20 cm.

1583

1584 **Figure 8.** Overturn during an eruption, as the lower layer of volatile-rich melt expands and
1585 becomes less dense than the original upper layer. This leads to a change in the erupting melt
1586 issuing from the summit ‘vent’ and the flank ‘vent’ following the overturn (Experiment courtesy
1587 of Alex Cowan). The vertical scale of the experimental tank is ~ 20 cm.

1588

1589 **Figure 9:** Results of experiments to show the exsolution of gas in a particle-laden viscous
1590 polymer solution as the solution is decompressed. Top: sequence of images showing the solution
1591 during and after decompression from 1 Bar to 35 mBar, leading to (i) exsolution of gas, (ii) liquid
1592 displacement within the pack and some dilation of the porous pack, producing an increase in
1593 permeability of the pack, (iii) the development of channelised permeable flow of gas from the
1594 pack; (iv) gradual relaxation of the depth of the porous layer once the connected gas phase has
1595 been driven out of the system. The tank is 18.2 cm wide. Owing to presence of residual gas, and
1596 the original expulsion of some liquid from the pack, a thin layer of melt may be seen above the
1597 porous pack (light layer at top of pack). Bottom: Graph illustrating the height of the particle-laden
1598 solution as a function of time when the pack is subjected to a decompression to different final
1599 pressures, as shown in the legend. With high final pressure, the effect is a monotonic increase in
1600 depth, as there is less gas exsolved. With a low final pressure the effect is a much larger increase
1601 in depth during the initial exsolution, but then the pack relaxes as gas is able to escape from the
1602 system. After Ptacek et al., sub judice (2018).

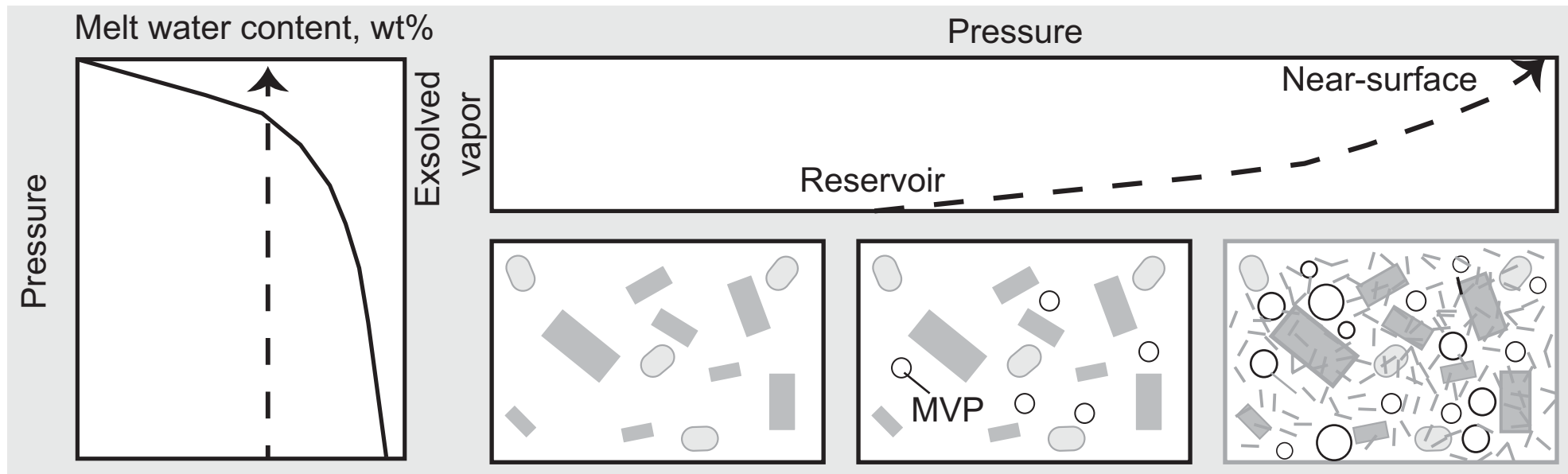
1603

1604 **Figure 10:** Cartoon diagram to summarise the generation, transport and accumulation
1605 mechanisms of the exsolved volatile phase, and the ways in which the exsolved volatile phase
1606 plays a role in driving magma mingling and mixing involving underplating mafic magmas.

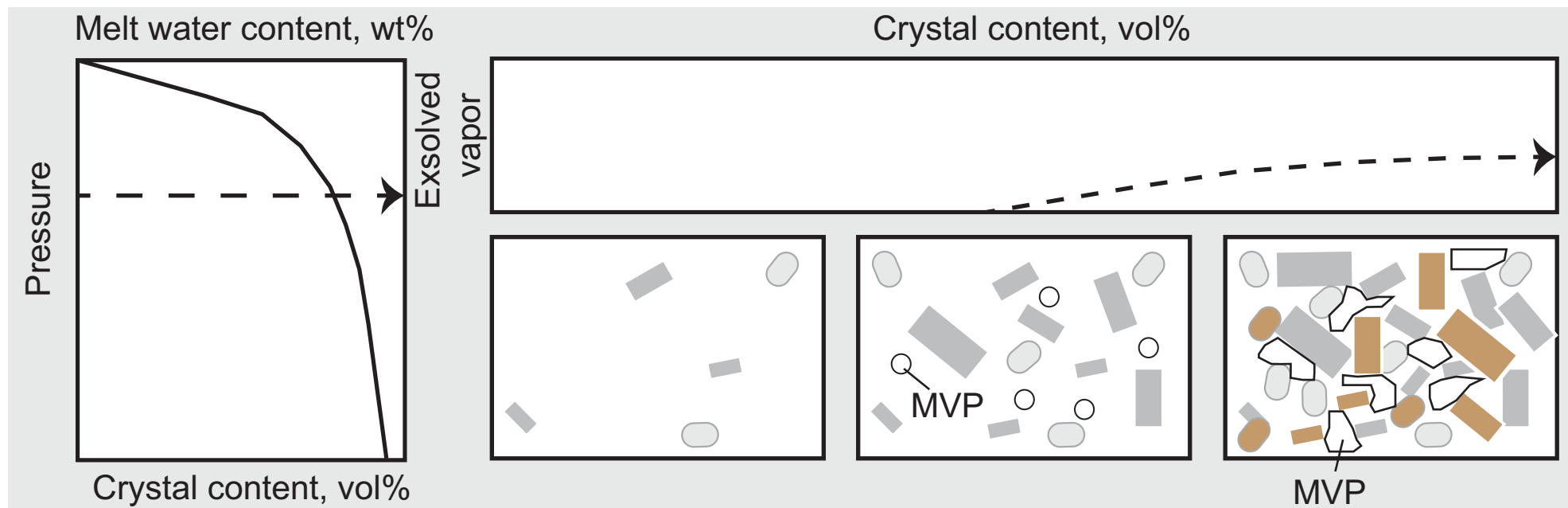
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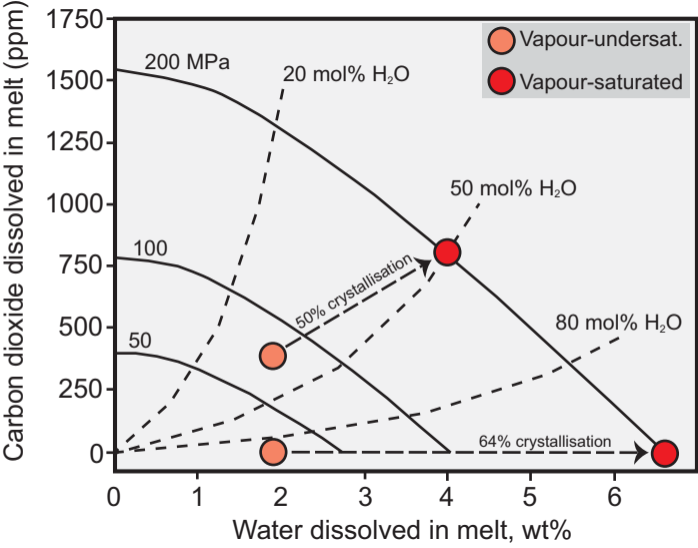
1608

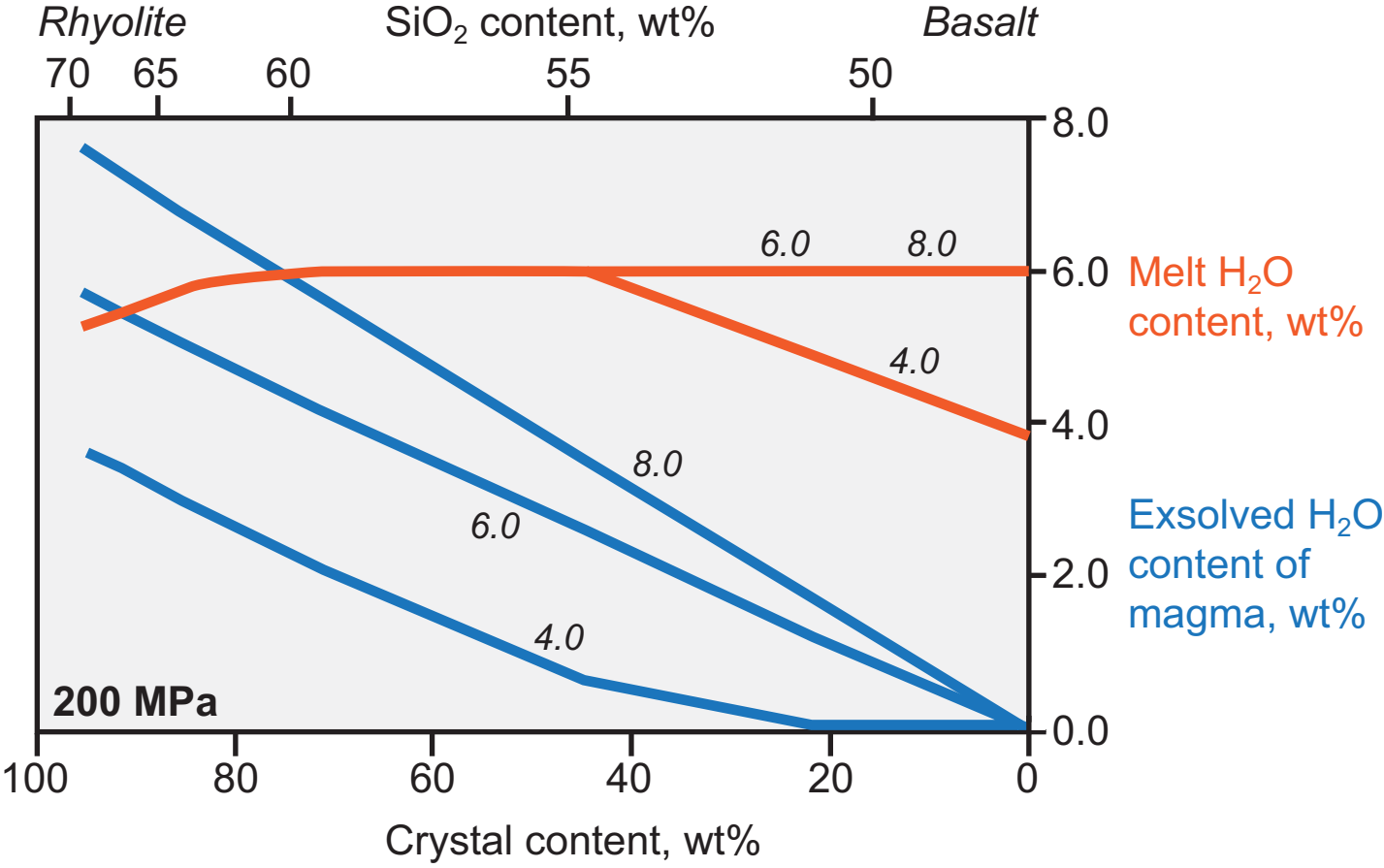
A: Exsolution of volatiles during decompression, "first boiling"

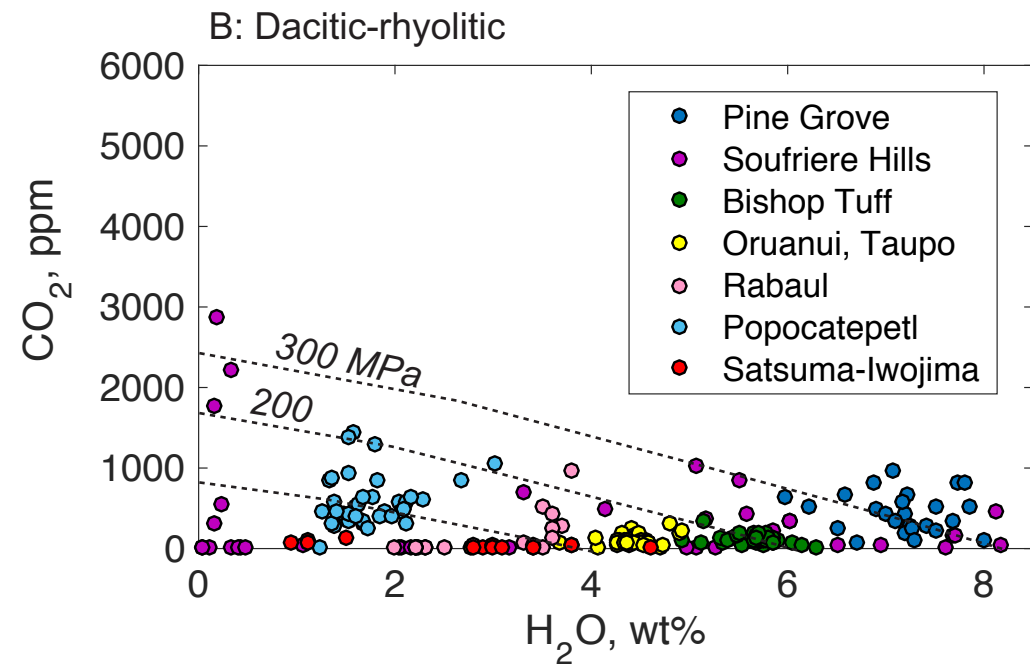
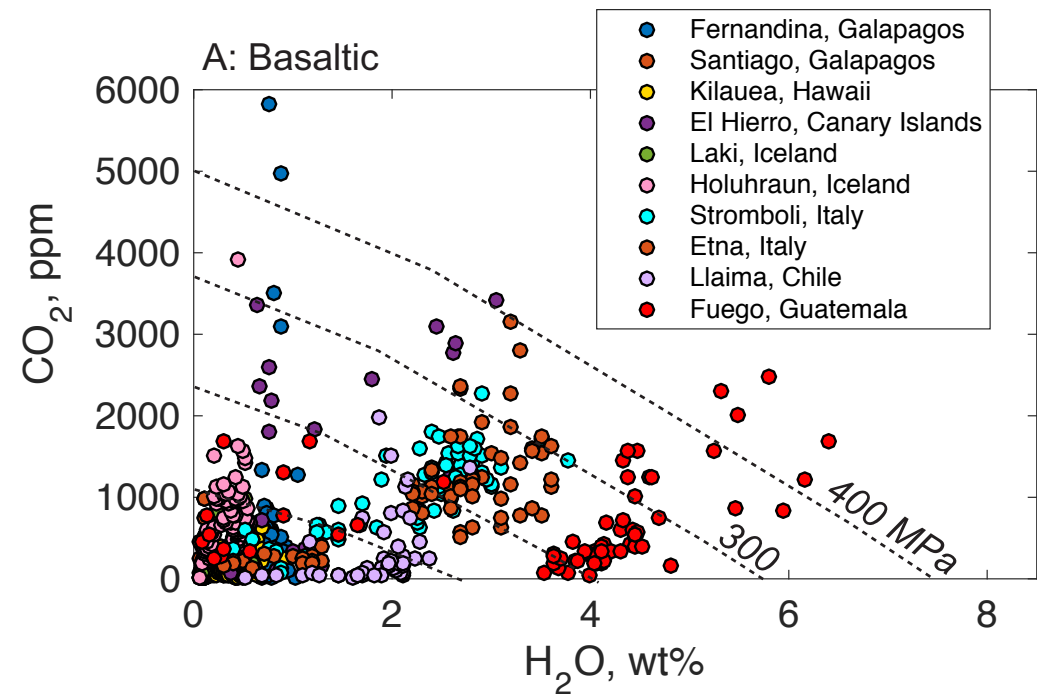


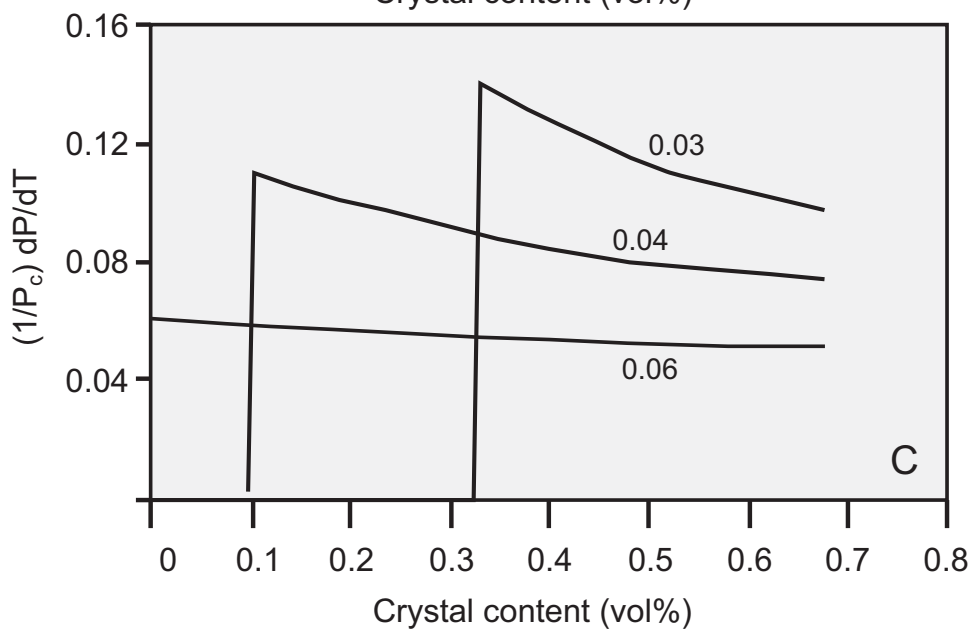
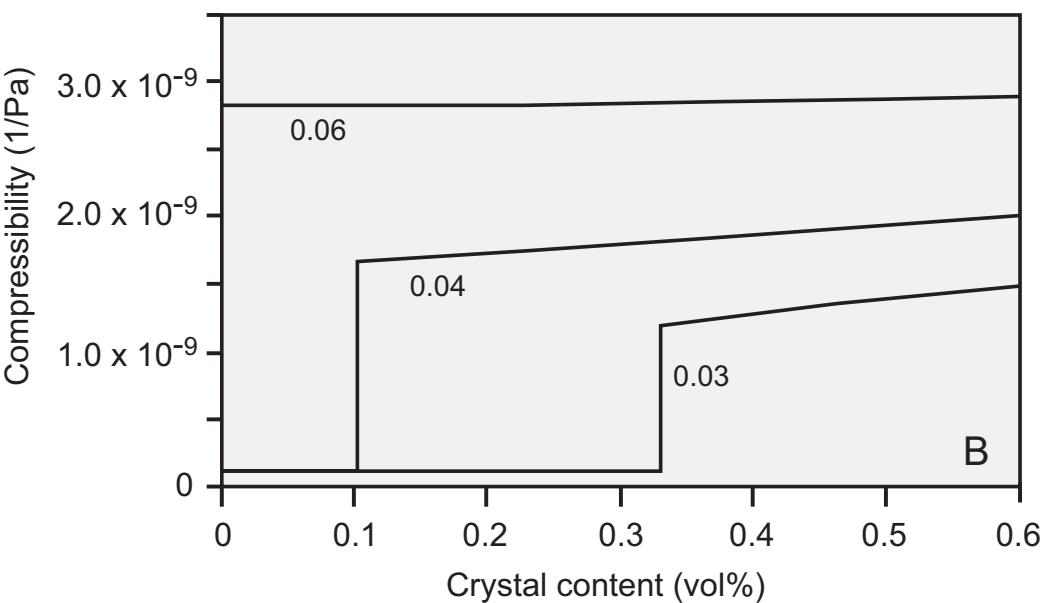
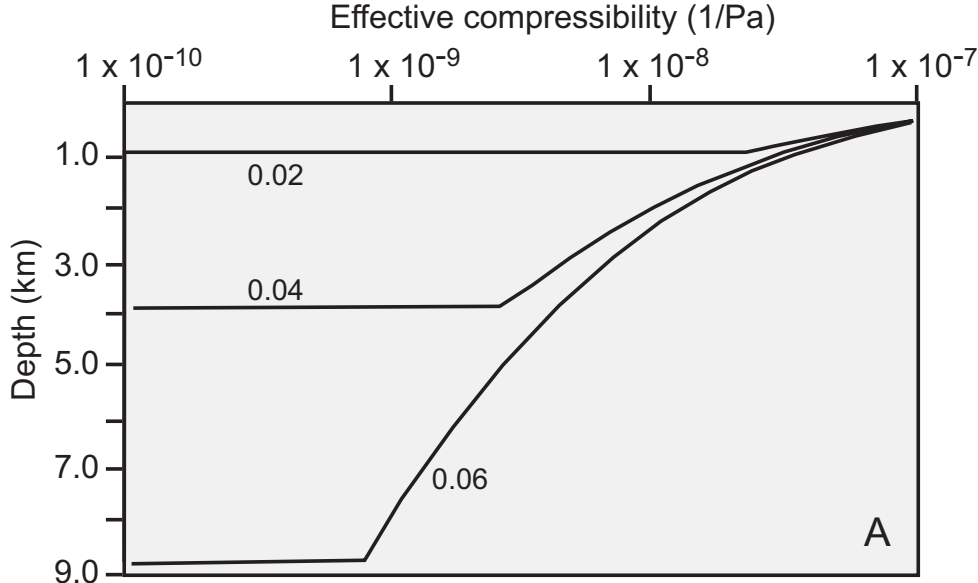
B: Exsolution of volatiles during cooling and crystallisation, "second boiling"

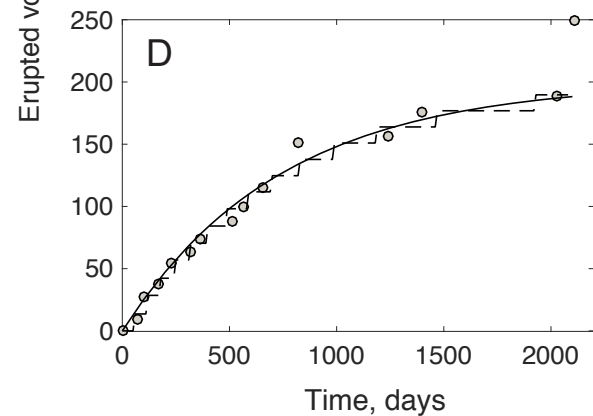
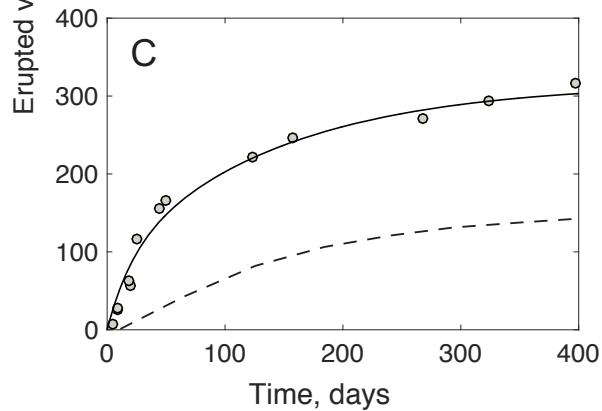
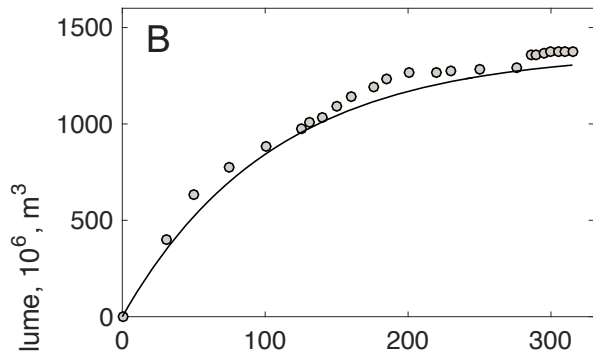
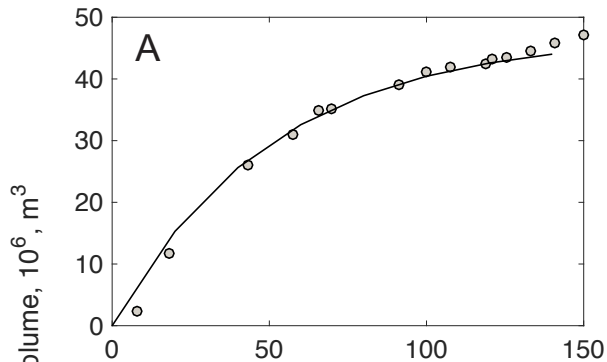




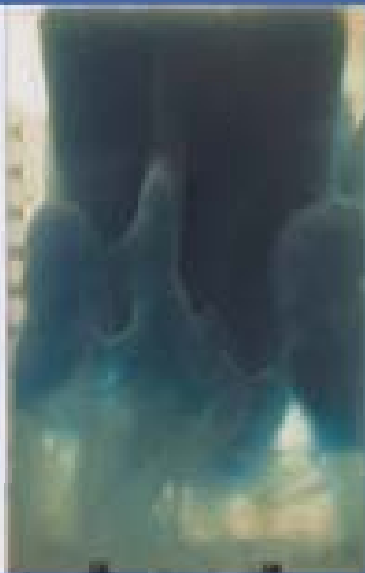








A



B

