# The role of H<sub>2</sub>O on the extraction of melt from crystallising magmas

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14 ABSTRACT

15 The segregation and accumulation of felsic melts, from crystallising crustal magma reservoirs, is 16 essential for the chemical evolution of the crust and is a phenomenon preceding some of the 17 largest eruptions on Earth. The physical properties of residual melt and magma and the time over 18 which the conditions remain appropriate for melt extraction are important factors controlling the 19 efficiency of melt extraction and the distribution of melt in magma reservoirs. Here we focus on 20 the initial water content  $(H_2O_i)$  of magma as it affects both the physical properties of the residual 21 melt and the timescales at which conditions remain appropriate for melt extraction during 22 progressive magma crystallisation. We use rhyolite-MELTS simulations to evaluate the physical 23 evolution of crystallising granodioritic (or dacitic) hydrous magma (i.e.  $\geq 1$  wt.%  $H_2O_i$ ) at 24 shallow depth at 200 MPa. To constrain the solidification timescales of reservoirs containing

25 magmas with initially different water content, we perform 2.5D thermal modelling. We combine 26 these results with calculations of melt extraction velocity by compaction and hindered settling to 27 identify the optimal conditions at which melt segregation occurs. These calculations suggest that 28 hydrous felsic magmas that attain water saturation after 40 wt.% crystallisation (rheological 29 locking point) are best suited for melt extraction. Once water-saturation is achieved, the rate of 30 release of latent heat of crystallisation and with it the time magma spends within a given 31 temperature interval increases while the viscosity of the residual liquid and crystal-liquid density 32 contrast remain favourable for melt segregation. We first test our findings on the Takidani pluton 33 (Japan) because it shows evidence of residual melt segregation from crystallising magma, and is 34 associated with caldera-forming eruptions. We finally generalise our results to crustal magma 35 reservoirs containing hydrous felsic magmas. Our results suggest that if segregation starts at 36 rheological locking (i.e. crystallinity of 40–50 wt.%) upper crustal reservoirs of  $\geq 100 \text{ km}^3$ 37 granodioritic (i.e. dacitic) magma with >2 wt.%  $H_2O_i$  can produce large melt-rich caps at the top 38 of partially crystallised magma reservoirs in few hundreds to few thousands of years. The 39 formation of separate melt lenses becomes more likely when segregation of melt starts at 40 crystallinities >0.6. Our results suggest that  $H_2O_i$  plays an important role in modulating the 41 distribution of eruptible melt in upper crustal reservoirs. Reservoirs of felsic and water-poor 42 magma (<2 wt.%  $H_2O_i$ ) tend to be associated with the formation of isolated pockets of crystal-43 poor and eruptible magma, which could account for the often-observed geochemical 44 heterogeneity of the products of large caldera-forming eruptions in the Snake River Plane. The 45 limited dimensions of these eruptible magma pockets make their detection by geophysical 46 methods challenging.

## 47 **1 Introduction**

48 The accumulation and storage of viscous high-silica melt in shallow magmatic reservoirs is a 49 process preceding some of the largest explosive eruptions on Earth. The distribution of crystal-50 poor and eruptible lenses of magmas within highly crystallised magma reservoirs cannot be 51 resolved by geophysical methods for intrinsic limits of spatial resolution (Bedrosian et al., 2018; 52 Huang et al., 2015). The timescales of extraction of viscous felsic melt from highly crystallised 53 magma are also only broadly constrained varying from months to thousands of years (Bachmann 54 and Huber, 2018; Druitt et al., 2012; Gualda et al., 2018; Huber et al., 2012; Wilson and 55 Charlier, 2016). Here we use a field example of extraction of felsic melt from crystallising 56 magma in combination with thermal modelling to investigate the role of magma water content on 57 the distribution and timescales of extraction of felsic melts from reservoirs of crystallising felsic 58 magma. 59 Segregation of interstitial melt from a rheologically-locked partially-crystallised magma body is 60 a potential mechanism for the accumulation of crystal-poor and eruptible rhyolite (Bachmann 61 and Bergantz, 2004; Dufek and Bachmann, 2010; Hildreth, 2004, 1981; Hildreth and Wilson, 62 2007; Marsh, 1981). Thermo-mechanical simulations suggest that the efficiency of melt 63 extraction for common hydrous silicic magma compositions is highest at crystal contents 64 between 50% and 70% (Dufek and Bachmann, 2010). Moreover, these studies emphasise that the 65 probability of extraction and the amount of interstitial melt segregated is not only controlled by 66 the physical properties of residual melt and magma, but also by the time spent by magma at 67 conditions best suited for melt extraction (Dufek and Bachmann, 2010; Huber et al., 2009). This, 68 in turn, is a function of the ratio between the rate of heat loss and the rate of release of latent heat 69 of crystallisation during progressive cooling and solidification of a magma reservoir, and

70 evolution of the physical properties of the residual melt and magma with increasing crystallinity 71 (Caricchi and Blundy, 2015; Huber et al., 2009; Lee et al., 2015; Melekhova et al., 2013). The 72 results of these studies permit to draw some general conclusions about extraction of residual melt 73 in felsic systems: i) Independently of the process leading to the extraction of residual melt in 74 crystallising felsic magmas, the separation between residual melt and crystals occurs when 75 magma is rheologically locked (i.e. crystal fraction >0.4; Dufek and Bachmann, 2010; Huber et 76 al., 2010; Marsh, 1981); ii) The velocity of residual melt extraction is directly proportional to the 77 ratio between the density difference of crystals and residual melt and the viscosity of the residual 78 melt (Bachmann and Bergantz, 2004; Dufek and Bachmann, 2010); iii) The longer magma 79 spends at conditions suitable for residual melt extraction, the larger is the amount of extracted 80 residual melt (Dufek and Bachmann, 2010; Huber et al., 2009). 81 Large-scale segregation of rhyolitic melts from highly crystallised magma is commonly inferred 82 to precede the eruption of rhyolitic magma (Bachmann and Bergantz, 2004; Deering et al., 2011; 83 Hildreth and Wilson, 2007), however, evidence for such segregation processes remains scarce or 84 obscure in the intrusive record (Coleman et al., 2004; Gelman et al., 2014; Lee et al., 2015; 85 Vigneresse, 2014). In recent years, various studies have targeted melt extraction processes in the 86 geological record using radiogenic isotopes (Andersen et al., 2017), whole-rock and mineral 87 chemistry (Barnes et al., 2017; Hartung et al., 2017), and rock fabrics (Holness et al., 2017). The 88 Takidani pluton in Central Japan shows evidence of melt segregation processes and is used here 89 as a case study to investigate the formation of bodies of crystal-poor and eruptible melt in the 90 upper crust (Hartung et al., 2017). This pluton has been shown to present the source of dacitic 91 and rhyolitic volcanic eruptions (Harayama, 1992; Kimura and Nagahashi, 2007; Nagahashi et 92 al., 2000). The pluton is texturally zoned, with a gradual transition (over about 50 m) from

equigranular granodiorite to porphyritic granite (Fig. 1). Whole rock and mineral chemistry
suggest that the porphyritic unit (pGT) was extracted from the underlying granodiorite (GDT)
once the residual melt fraction dropped to 40–50 wt.% (Figs. 1, 2a; Hartung et al., 2017). The
initial water content of the granodiorite associated with melt segregation is estimated between 3
and 4 wt.% H<sub>2</sub>O based on mineral chemistry, crystallisation sequence and relative abundance of
mineral phases (Hartung et al., 2017).

99 In this study, we focus on the effects of the initial water content  $(H_2O_i)$  on the timescales magma 100 spends at crystallinities larger than about 40 wt.%, which are considered favourable for the 101 extraction of residual melt (Bachmann and Bergantz, 2004; Dufek and Bachmann, 2010). We 102 first calculate the evolution with temperature and crystallinity of the physical properties of 103 magma and residual melt for granodioritic (i.e. dacitic) magmas with water contents between 1 104 and 6 wt.%. The velocity of melt extraction for crystallinities between 40 and 80 wt.% are 105 estimated using hindered settling (Davis and Acrivos, 1985) and compaction-driven segregation 106 (McKenzie, 1984). To constrain the maximum timescales available for segregation to occur we 107 perform thermal modelling for reservoirs of different volumes and shapes. The aims of this study are (1) to constrain the effect of  $H_2O_i$  on the efficiency of melt extraction, (2) to identify the 108 109 conditions that led to the extraction of residual melt from the Takidani pluton and estimate the 110 timescales of this process, (3) to define the impact of  $H_2O_i$  on the architecture of upper crustal 111 magma reservoirs.

#### 113 **2** Material and methods

## 114 2.1 The Takidani Pluton: Evidences for melt segregation

115 In the following we provide a summary of the main results of a geochemical study previously 116 performed on the Takidani pluton (Hartung et al., 2017), which provides the background and 117 motivation for this study. The Takidani pluton is a well exposed and young pluton (1.6 Ma; 118 Harayama, 1992; Ito et al., 2017), located in the Central Japan Alps. The pluton is vertically 119 exposed over 1800 m (Harayama et al., 2003) from a tectonic contact at the base to a magmatic 120 roof contact with older volcanic rocks (i.e. Hotaka Andesite, Harayama, 1994). Textural, 121 chemical and isotopic evidence of large-scale melt segregation is observed in the upper part of 122 the pluton (Figs. 1, 2; Hartung et al., 2017). The rock textures of the Takidani pluton change from holocrystalline to progressively more porphyritic appearance from the base and centre to 123 124 the roof of the intrusion. Whole-rock geochemistry shows that the rocks immediately below the 125 porphyritic unit are depleted in incompatible elements, while the porphyritic unit is enriched in 126 incompatible elements (Fig. 1; Hartung et al., 2017). Data obtained through quantitative 127 evaluation of minerals by QEMSCAN and electron microprobe analyses (i.e. EPMA) are used to 128 determine the area percent and chemical composition of the matrix components (equivalent to 129 residual melt composition) throughout the upper section of the Takidani pluton, where evidence 130 for melt extraction is observed (Fig. 1; supplementary data: Table 1). These data show 131 progressive enrichment in the residual melt components defined by quartz (Qtz), alkali feldspar 132 (Kfs) and albite-rich plagioclase (Plg<An<sub>30</sub>) from the equigranular granodiorite (GDT) to the 133 porphyritic granite (pGT; Fig. 1). The relative amounts of quartz, albite (Ab), and orthoclase (Or) 134 and therefore the melt compositions, however, do not change across the textural and chemical 135 transition (Fig. 1; supplementary data: Table 1). This suggests that the residual melt had a

chemical composition close to the granitic minimum after emplacement at approximately 200
MPa (Johannes and Holtz, 1996) and that the extraction of the residual melt, now represented by
the porphyritic unit, occurred once magma crystallised sufficiently for the residual melt to
acquire a composition close to the granitic minimum (Fig. 1). Mineral chemistry provides
additional support for the extraction of residual melt once the magma was rheologically locked
(Hartung et al., 2017).

142 Plagioclase phenocrysts in the granodioritic unit below the porphyritic have cores of different 143 chemistry that are overgrown by a distinct rim of common composition, which is consistent with 144 the composition of plagioclase in the matrix of the porphyritic unit (Hartung et al., 2017). An 145 increase in Rb (incompatible during crystallisation of the Takidani granodiorite; Hartung et al., 146 2017) in plagioclase phenocrysts increases by more than a factor of two from the core to the 147 common outer rim. This suggest that once magma crystallised to approximately 40–50 wt.%, 148 plagioclase acquired the same composition indicating rheological locking (Fig. 2). Within this 149 crystallinity range the residual melt was extracted leading to the formation of the porphyritic unit 150 of the Takidani pluton that contains matrix plagioclase with the same composition of the 151 plagioclase rims of the underlying granodioritic unit. 152 Inclusions of orthopyroxene in amphibole, amphibole thermometry, plagioclase composition, 153 and comparison with the phase equilibria experiments of Costa et al. (2004), highlight that 154 amphibole became stable once the magma achieved crystallinities of 40–50 wt.% (Hartung et al., 155 2017). The late appearance of amphibole at low temperatures and core to rim plagioclase 156 chemistry indicate that the Takidani magma was not initially water-saturated (Costa et al., 2004) 157 and contained initially between 3 and 4 wt.% H<sub>2</sub>O.

On the basis of the collected evidence we conclude that the pGT unit represents a lens of residual melt extracted from the underlying GDT granodiorite. The extraction of residual melt from the crystallising magma occurred after the magma became rheologically locked, at which point the residual magma had reached water saturation (Costa et al., 2004; Hartung et al., 2017). The evidence supporting initial water-undersaturation of the magma and melt extraction for the Takidani pluton motivate us to investigate the role of  $H_2O_i$  on the extraction of felsic melt from crystallising magmas.

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## 166 2.2 Rhyolite-MELTS simulations

167 Existing experimental data do not cover the entire range of temperature and water contents 168 required to trace the evolution of residual melt during cooling and crystallisation of magma in 169 the upper crust (Costa et al., 2004; Holtz et al., 2005; Scaillet and Evans, 1999). Thus, we use 170 rhyolite-MELTS (Gualda et al., 2012) to calculate the chemical and physical evolution of 171 residual melt of dacitic (or granodioritic) magma from liquidus to near solidus temperatures and 172 over the entire range of  $H_2O_i$  between 1 wt.% and water saturation. Because granodiorites 173 represent about 95 wt.% of the upper crust (Rudnick and Gao, 2003) with a composition similar 174 to the starting material of Costa et al. (2004), we use the latter for the rhyolite-MELTS 175 calculations. While rhyolite-MELTS it currently not capable of correctly identifying the stability 176 of hydrous phases such as amphibole and biotite, here we use rhyolite-MELTS, especially, to 177 trace the general evolution of residual melt chemistry and magma crystallinity. To test the 178 performance of rhyolite-MELTS we compare the calculated residual melt compositions with 179 residual melts produced experimentally by Costa et al. (2004) using the same starting 180 composition. For all calculations, the confining pressure was fixed at 200 MPa, which are the

181 conditions applied in the experiments of Costa et al. (2004) and comparable to the inferred 182 emplacement depth of many granitic intrusions including the Takidani pluton (Hartung et al., 183 2017). The oxygen fugacity was initially set to the nickel-nickel oxide buffer (NNO) to calculate 184 the liquidus temperature (for different  $H_2O_i$ ), but remained unconstrained during progressive heat 185 extraction. The evolution of the residual melt chemistry as function of temperature and  $H_2O_i$ 186 calculated with rhyolite-MELTS is comparable to that obtained in the experiments of Costa et al. 187 (2004; Fig. 3). Importantly, rhyolite-MELTS can accurately reproduce the evolution of silica 188 content of the residual melt, which together with water plays a dominating role in controlling the 189 viscosity of silicic melts (Hess and Dingwell, 1996). Additionally, the decrease of melt fraction 190 with temperature for different  $H_2O_i$  follows paths that are comparable to those determined 191 experimentally by Whitney (1988; Fig. 4b), hence we consider the rhyolite-MELTS calculations 192 appropriate to compare the evolution of physical properties of hydrous magmas during 193 progressive crystallisation.

194 To quantify the time magma spends within different temperature intervals we perform isobaric 195 rhyolite-MELTS simulations by removing an equal amount of enthalpy (i.e. 1 J g<sup>-1</sup>) from the 196 system at each step (n) starting from the liquidus temperature down to a temperature (T) of about 197 740°C, which corresponds to a residual melt fraction of about 0.1. If a fixed amount of enthalpy 198 is extracted from the magma during solidification, which would be appropriate for quasi-eutectic 199 magma compositions (Gualda et al., 2018), the number of modelling steps within a given 200 temperature interval becomes proportional to the time spent by the magma within a given 201 interval of temperature. This is important to be able to quantify the total time that magma spends 202 within the temperature and crystallinity range at which the conditions are most conducive to melt 203 extraction (Dufek and Bachmann, 2010). Considering that the rate of heat loss will decrease

during magma crystallisation because of the decreasing temperature difference between magma
and host rocks, our approach increasingly underestimates the time spent within a given
temperature range. This is especially the case at intermediate to high crystallinities, where
segregation of interstitial melt takes place. The difference between this scenario and one more
appropriate for non-eutectic magmas, for which the non-linear rate of heat release over time must
be considered, is addressed by thermal modelling.

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## 211 2.3 Thermal modelling

To provide constrains on the timescales of melt extraction, we use thermal modelling to compute the temporal evolution of temperature in instantaneously emplaced cylindrical magma bodies of various volumes, aspect ratios and initial water contents. We solved the two-dimensional axisymmetric formulation of the heat conduction equation, which can be written as:

216 
$$\rho c \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r k \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) + \rho L \frac{\partial X_c}{\partial t}$$
(1)

217 were T is the temperature, t is the time, r is the radial coordinate relative to the symmetry axis, z 218 is the depth, k is the thermal conductivity, L is the latent heat of crystallisation,  $\rho$  is the density, c 219 is the specific heat and  $X_c$  is the fraction of crystals in the magma. For a list of the parameters 220 used in the modelling the reader is referred to Table 2 in the supplementary data. The 221 calculations were performed on a numerical grid using an explicit finite difference discretisation 222 of the above equation. The model considers latent heat of crystallisation, which was 223 implemented using our rhyolite-MELTS results to parametrise the crystal fraction  $(X_c)$  versus 224 temperature for the different initial water contents used in this study (Fig. 5a). As the dependence 225 of  $X_c$  on T is non-linear, the governing equation was solved using an iterative strategy. In all models, we integrated an initial geothermal gradient of 25 °C km<sup>-1</sup> and a temperature-dependent 226

thermal conductivity (*k*) as described in Whittington et al. (2009) for average crust. The upper
(i.e. surface) and lower boundary (i.e. 25–35 km) in the models was set to a fixed temperature
determined by the initial geothermal gradient, while zero flux boundary conditions were imposed
on the left and right side. Cooling due to circulation of hydrothermal fluids around the magma
reservoir was neglected in the modelling and the potential effects of this simplification are
discussed in the following.

233 The setup consists of a 25 x 25 km domain for magma volumes of 100 and 1000 km<sup>3</sup>, and a 35 x 234 35 km domain for intrusions of 10.000 km<sup>3</sup>, using 400 x 400 numerical grid for all simulations 235 (supplementary data: Fig. S1). In each model the magma body was intruded instantaneously at 236 10 km depth at its liquidus temperature and crystallised to near solidus conditions, as determined 237 by the rhyolite-MELTS simulations. To test the effect of reservoir shapes on the solidification 238 history, we ran models with intrusion aspect ratios of 2 and 10 for magma volumes of 100 and 1000 km<sup>3</sup>, while model pluton volumes of 10.000 km<sup>3</sup> were only performed with an aspect ratio 239 240 of 10, due to the large vertical extent of such reservoirs, which makes the application of a single 241 melt fraction-temperature relation unreasonable. Finally, to compare melt segregation velocities 242 to solidification timescales we tracked the temporal propagation of isotherms and melt fraction 243 with depth in the centre of the intrusion.

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#### 245 **3 Results**

# 246 *3.1 Thermal, chemical and physical evolution of dacitic magma*

Water has an important effect on phase equilibria as it depresses liquidus temperatures and
modifies the relationships between temperature, crystallinity, and the physical properties of
magma over a wide range of chemical compositions (e.g. viscosity and density of the residual

250 melt; Blatter et al., 2013; Caricchi et al., 2007; Giordano et al., 2008; Hess and Dingwell, 1996; 251 Lange, 1994; Melekhova et al., 2013; Müntener and Ulmer, 2018; Ulmer et al., 2018; Whitney, 252 1988). Crystallisation of magma leads to a non-linear increase in silica and water content in the 253 residual melt (Fig. 4a-d). Once the residual melt becomes water-saturated, initially water-254 undersaturated magmas join the *T*-melt fraction trajectory of initially water-saturated magma 255 (Fig. 4b). The non-linear relationships between melt fraction and temperature, contributes to 256 modulate both the timescales magma spends at different temperatures, and the temporal 257 evolution of the physical properties of the residual melt during progressive magma 258 crystallisation. In crystallising magma reservoirs, magma spends relatively more time at 259 temperatures at which the rate of crystallisation (i.e. rate of latent heat release) is highest 260 (Caricchi and Blundy, 2015; Marsh, 1981). Thus, if the crystallisation rate increases once 261 rheological locking is achieved, magma will spend relatively more time at these temperature 262 conditions (Huber, 2009, Caricchi and Blundy, 2015). The time is even larger considering that 263 the rate of heat release from magma reservoirs drops with cooling and progressive decrease of 264 the thermal gradient between magma and host rocks.

265 Magmas with lower water contents will achieve rheological locking at higher temperatures with 266 respect to water-rich magmas, which will, in turn, impact viscosity, density and velocity of 267 residual melt extraction (Fig. 5b, c; Bachman and Bergantz, 2004). Our calculations show that 268 regardless of  $H_2O_i$  of magma, residual melt viscosity increases down to melt fractions of 0.5–0.4 269 because of decreasing temperature and increasing silica content. At lower melt fractions (<0.4) 270 and once volatile saturation is achieved, the viscosity of the residual melt remains relatively 271 constant independent of  $H_2O_i$  (Fig. 5b). Relatively dry melts ( $H_2O_i \leq 2$  wt.%) reach a maximum 272 in viscosity before joining the same trajectory of water-richer magmas at lower temperatures

273 (Fig. 5b). The contrast in density between the solid phase and residual melt spans a wide range of 274 values near liquidus conditions (for different  $H_2O_i$ ) but becomes less dependent on  $H_2O_i$  for melt 275 fractions <0.6 (Fig. 5c). The ratio of the difference in density between crystals and residual melt 276 and the viscosity of the residual melt, which is directly related to the velocity of melt extraction 277 (Bachman and Bergantz, 2004), generally increases with water content (Figs. 5b, c). 278 Based on the physical properties of residual melt and magma, the melt extraction velocity is the 279 fastest for initially water-saturated magmas. However, the total time spent by magma at melt 280 fractions <0.6 (i.e. rheologically locked conditions) is inversely proportional to  $H_2O_i$  (Fig. 5d). 281 To assess the relative importance of  $H_2O_i$  on the physical properties of magma and the timescales 282 available for melt segregation to occur, we calculate the velocity of melt extraction by hindered 283 settling (Bachmann and Bergantz, 2004) and compaction (McKenzie, 1984) for granodioritic-284 dacitic magma for melt fraction <0.6. We notice that the formulations used to calculate the 285 velocity of melt extraction are rather simplified, however, such order of magnitude estimates are 286 important to compare the efficiency of residual melt extraction for magmas with different  $H_2O_i$ . 287 We do not consider the presence of excess magmatic fluids (i.e. H<sub>2</sub>O, CO<sub>2</sub>, S) during melt 288 extraction, which can have both positive and detrimental effects on the efficiency of melt 289 extraction (Boudreau, 2016; Caricchi et al., 2018; Parmigiani et al., 2016; Pistone et al., 2015; 290 Sisson and Bacon, 1999).

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292 3.2 Velocities of melt segregation

Hindered settling and compaction-driven segregation describe the relative motion and separation
of liquid (i.e. melt) and solid (i.e. crystal) in a two-phase system (i.e. crystallising magma). The
process of particle settling in a monodisperse suspension is described by hindered settling

(Equation 2). Compaction-driven segregation defines the process of compacting a porous
crystalline mush and resulting melt expulsion (Equation 3). A detailed evaluation of hindered
settling and compaction-driven segregation processes is presented in Bachmann and Bergantz
(2004) and Lee et al. (2015).

To obtain velocities for hindered settling ( $U_{hs}$ ) and compaction-driven segregation ( $V_{comp}$ ) for crystallising dacitic magma we use the physical properties obtained from rhyolite-MELTS simulations. We calculate segregation velocities following the equations provided by Bachmann and Bergantz (2004; and references therein):

304 
$$\boldsymbol{U}_{hs} = \frac{2 \cdot r^2 \cdot g \cdot \Delta \varrho}{9 \cdot \mu} \frac{(1-c)^2}{(1+c_3^3)^{[\frac{5}{3}c(1-c)]}}$$
(2)

305

306 
$$V_{comp} = \frac{\kappa \cdot (1 - \phi) \cdot \Delta \varrho \cdot g}{\mu \cdot \phi}$$
(3)

307 Where *r* is the crystal radius, *g* is gravitational acceleration (9.81 ms<sup>-1</sup>),  $\Delta \rho$  is the density 308 difference between crystal and melt,  $\mu$  is the viscosity, *c* is the crystal fraction,  $\kappa$  is the 309 permeability and  $\phi$  is the porosity. The permeability is calculated from the porosity, crystal 310 radius and permeability coefficient (*K*) after McKenzie (1984):

311 
$$\boldsymbol{\kappa} = \frac{\boldsymbol{\phi}^3 \cdot \boldsymbol{r}^2}{K(1-\boldsymbol{\phi})^2} \tag{4}$$

We consider that porosity is equal to melt fraction and we use *K* values between 50 and 200 (Bachmann and Bergantz, 2004; Pistone et al. 2015). Here we assume monodisperse suspensions of melt and crystals. As magmas contains crystals of different sizes, and this tends to reduce the permeability with respect to monodisperse systems (Bachmann and Bergantz, 2004; Rust and Cashman, 2011), the volumetric rates of melt extraction calculated here for each grain size are maximum estimates. 318 To calculate and compare segregation velocities for dacitic magmas with different  $H_2O_i$ , we first 319 assume that no convection occurs and that magmas are rheologically locked at crystallinities >40 320 wt.%. Based on Equation 2, velocities for hindered settling vary between 3.6 and 0.06 m yr<sup>-1</sup> for 321 water-saturated magmas and melt fractions decreasing from 0.6 to 0.2 (Fig. 6a) using a crystal 322 radius of 3 mm. Velocities for under-saturated magmas are lower (i.e. 1 wt.% H<sub>2</sub>O<sub>i</sub>; Fig. 6a) and decrease from about 0.22 to 0.02 m yr<sup>-1</sup> with increasing crystallinity. Segregation velocities for 323 324  $H_2O_i \ge 3$  wt.% increase in similar fashion as water-saturated melts within the rheologically 325 locked interval (Figs. 5a, 6a). Melt segregation of undersaturated magmas with water content <3326 wt.% occurs on timescales up to one magnitude slower as they become water-saturated at lower 327 temperature and at higher crystallinities resulting in slower melt extraction velocities. The 328 timescales of melt extraction processes strongly depend on the crystal size and vary by more than 329 one order of magnitude when considering crystal radii of 1 to 5 mm (Fig. 6b). 330 We also calculate the velocity of melt extraction considering compaction using a permeability coefficient of 50 and a crystal radius of 3 mm and obtain values between 2.2 m yr<sup>-1</sup> and 5.0 x 10<sup>-4</sup> 331 m yr<sup>-1</sup> for water-saturated magma and between 1.4 x 10<sup>-1</sup> m yr<sup>-1</sup> and 3.9 x 10<sup>-4</sup> m yr<sup>-1</sup> for water-332 333 undersaturated magma. Segregation velocities decrease up to one order of magnitude when 334 applying larger permeability coefficients (i.e. 100-200; Fig. 6d). Velocities of melt extraction for 335 compaction-driven segregation and hindered settling are similar at melt fractions between 0.6 336 and 0.5, however, compaction velocities decrease more rapidly with decreasing melt fraction 337 than hindered settling (Bachmann and Bergantz, 2004; Lee et al., 2015). Below melt fractions of 338 0.2, the velocity of compaction driven segregation has been shown to increase relative to 339 hindered settling (Lee et al., 2015). Our results show that residual melt separates most effectively 340 from highly crystallised dacitic magmas if  $H_2O_i$  is equal or greater than 3 wt.%, independently of

the extraction mechanisms considered (Fig. 6). As phase equilibria and water solubility are both affected by pressure, 3 wt.%  $H_2O_i$  is a threshold value appropriate for confining pressures of 200 MPa. The increase of water solubility with pressure could increase the efficiency of meltextraction at mid to deep crustal levels (i.e. 15–20 km depths), where potentially larger  $H_2O_i$ would result in lower melt viscosity and accelerate segregation processes. At such depths, the rate of heat release from magma reservoir is also lower, which would also increase the time available for segregation of residual melt.

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### 349 3.3 Crystallisation and melt segregation timescales of hydrous felsic magmas

350 The thermal modelling results allow us to constrain the crystallisation timescales of hydrous 351 dacitic magmas at melt fraction between 0.6 and 0.2, where segregation is most efficient (Dufek 352 and Bachmann, 2010). We track the position of the isotherm corresponding to a given melt 353 fraction along a vertical section through the middle of the intrusion. Each isotherm propagates 354 from the base and the top of the intrusion toward its inner portions (Fig. 7a). We define the 355 "maximum segregation timescale" as the time difference between the moment the isotherms 356 corresponding to melt fractions of 0.6 and 0.2 reach the centre of the intrusion (Fig. 7b). 357 The thermal models show that magmas with low  $H_2O_i$  (i.e. <3 wt.%  $H_2O_i$ ) spend substantially 358 more time at rheologically locked conditions than magmas that are initially water-saturated (Fig. 8). For reservoir volumes of 100, 1000, and 10.000 km<sup>3</sup>, the maximum timescales for 359 360 segregation increase from 13 kyr, 61 kyr and 274 kyr for water-saturated magmas to 19 kyr, 84 361 kyr and 349 kyr, and 20 kyr, 94 kyr and 444 kyr for magmas with  $H_2O_i$  of 3 and 2 wt.%, 362 respectively. The timescales available for melt segregation increase by a factor of 4 to 5 with 363 decreasing aspect ratios from 10 to 2 (Fig. 8).

364 To estimate the maximum melt migration distances, we first calculate the average segregation 365 velocity of hindered settling and compaction (Equations 2, 3) for each melt fraction interval of 366 0.1 between 0.6 and 0.2. Secondly, each of these values are multiplied by the time required by 367 the corresponding isotherm to reach the core of the intrusion, once the melt fraction within the reservoir has reached 0.6 (Figs. 7a and 9a,b). Because of the relatively high segregation 368 369 velocities at melt fractions between 0.6 and 0.5, the residual melt can migrate large distances 370 over the duration of isotherm propagation from 0.6 to 0.5 (Figs. 6, 9a,b). For example, maximum 371 segregation distances fall in the order of tens of kilometres for a magma reservoir with a volume 372 of 1000 km<sup>3</sup> (Fig. 9 a,b). The timescales for melt to travel to the roof are fastest for water-373 saturated magma increasing from 600 to 2500 yr for water-saturated magmas and magmas with 374  $H_2O_i$  of 2 wt.%, respectively (Fig. 9b). These values decrease to about 300 to 1200 yr for 375 reservoir volumes of 100 km<sup>3</sup> and increase to about 1300 and 5400 yr for magma reservoirs with 376 a volume of 10.000 km<sup>3</sup> (Fig. 9b). For a reservoir of 1000 km<sup>3</sup>, the time required for the 377 extracted melt to reach the roof by compaction-driven segregation is twice as long and range 378 from 1300 to 5700 yr for water-saturated and magma with 2 wt.%  $H_2O_i$ , respectively.

379

#### 380 4 Discussion

## 381 4.1 Segregation timescales of the Takidani Pluton

Magmas of the Takidani Pluton were initially water-undersaturated with water contents of about 3 to 4 wt.% (Hartung et al., 2017; Costa et al., 2004). During crystallisation and melt evolution the residual liquids would have reached volatile saturation at melt fractions between 0.70 (i.e. 4 wt.%  $H_2O_i$ ) and 0.55 (i.e. 3 wt.%  $H_2O_i$ ) and temperatures around 820°C to 780°C (Fig. 5a,b). At this point an increase of crystallisation rate and release of latent heat of crystallisation would

387 have resulted in near isothermal crystallisation. A potential indication of this process may come 388 from amphibole thermometry, which suggests the crystallisation of this phase occurred almost 389 entirely in between 800 and 750 °C (Hartung et al., 2017). This could support our hypothesis that 390 magma was thermally buffered over long time periods by the release of latent heat of 391 crystallisation. 392 Based on our rhyolite-MELTS simulations and thermal modelling we estimate the maximum and 393 minimum timescales over which melt segregation took place in the Takidani Pluton. We consider 394 that the column of melt (h) is equal to the thickness of the porphyritic unit (270 m; Fig. 1, 395 Hartung et al. 2017). Velocities of hindered settling segregation of magma with  $H_2O_i$  of 3 to 4 wt.% range from 3.1 m yr<sup>-1</sup> to about 0.06 m yr<sup>-1</sup> between 0.6 and 0.2 melt fraction for an 396 397 intermediate crystal radius of 3 mm, which is considered appropriate for granitoids (Bachmann 398 and Bergantz, 2004; Lee et al., 2015) including the Takidani pluton (Fig. 1; Hartung et al., 2017). 399 This would imply that the extraction of the melt producing the porphyritic unit of the Takidani 400 pluton required between about 100 yr and 5000 yr. If we considered a smaller crystal size (i.e. 1 401 mm) the time required for segregation by hindered settling would increase by approximately one 402 order of magnitude to 1000 yr and 45 kyr. Velocities for compaction-driven segregation for the same water contents and crystal size (i.e. 3 mm) vary between 0.5 m yr<sup>-1</sup> and 0.0001 m yr<sup>-1</sup> 403 404 leading to much slower extraction timescales of about 800 yr to 2 Myr. 405 The Takidani Pluton has an exposed horizontal extension of about 14 km and exposed vertical 406 extension of about 2 km, which implies a minimum volume of 300 km<sup>3</sup> considering a cylindrical 407 shape. The results of our thermal models show that the time available for segregation in the 408 Takidani Pluton with magmas containing 3 to 4 wt.% H<sub>2</sub>O<sub>i</sub> varies between 40 and 120 kyr for 409 aspect ratios of 10 and 2, respectively (Fig. 8). The thermal modelling and velocity calculations

410 we performed, show that compaction-driven segregation generally operates at timescales that are 411 larger than estimated solidification timescales and therefore cannot explain the observed 412 segregation in the Takidani Pluton. Hindered settling, on the other side, is a much faster process 413 that can explain the observed melt segregation in the Takidani Pluton. Only in the eventuality 414 that segregation would have started at melt fraction smaller than 0.3, the porphyritic unit of the 415 Takidani pluton would not represent a melt-rich cap near the roof of the pluton. Considering that 416 the base contact of the pluton is tectonic and therefore its volume could be considerably larger 417 than 300 km<sup>3</sup>, our results show that sufficient time was available for melt segregation to occur 418 and to form a melt cap via hindered settling (Fig. 10) or processes that operate at similar 419 timescales (i.e. gas filter pressing; Sisson and Bacon, 1999). Finally, evidence from quartz hosted 420 fluid inclusion studies suggest that the Takidani Pluton developed a liquid-dominated 421 hydrothermal systems at some point in the past (Bando et al., 2003; Sekine et al., 2001). Heat 422 advection associated with hydrothermal circulation is not considered in our calculations but 423 would have increased the rate of heat release depending on the vigour of fluid convection 424 (Delaney et al., 1995). This directly decreases the time available for melt segregation (Dutrow et 425 al., 2001), thus increasing the potential for melt to form isolated melt lenses instead of a large 426 cap on top of magmatic reservoirs.

427

428 4.2 The control of H<sub>2</sub>O<sub>i</sub> on the extraction of residual melt from crystallising magmas
429 Geochemical and petrologic studies show that crystal-poor rhyolites are sourced either from caps
430 at the top of partially crystallised reservoirs (Bachmann and Bergantz, 2004; Hildreth and
431 Wilson, 2007), or from the amalgamation of isolated melt pockets dispersed within a highly
432 crystallised magma body (Wotzlaw et al., 2014, Ellis et al., 2014). The processes responsible for

433 the generation of reservoirs with such distinct architecture are not yet fully understood. The total 434 distance over which melt migrates before the system cools to its solidus temperature, controls 435 the final distribution of crystal-poor felsic melt in magmatic reservoirs. Thus, the maximum 436 amount of rhyolitic melt that can potentially be accumulated depends on  $H_2O_i$  and the 437 temperature of the magma, the volume and shape of the magma body and the temperature 438 difference of the intruding magma and the host rock (i.e. thermal gradient). We have illustrated 439 that the initial amount of water dissolved in magmas affects (1) the physical properties and 440 segregation velocities of residual melts (Figs. 5, 6) and (2) the total amount of time spent within 441 the rheologically locked temperature interval (Figs. 7, 8; Caricchi and Blundy, 2015). 442 Our calculations show that crystal size is an important factor controlling segregation velocities of 443 residual melt (i.e. Bachmann and Bergantz, 2004), while the initial water content affect both the 444 melt extraction velocity and the release of latent heat, which, in turn, modulates the time spent by 445 the magma within different temperature intervals. Volume and shape of the magma reservoir 446 affect the thermal evolution and timescales of reservoir solidification. In case of hindered 447 settling, residual melts of magmas that contain at least 2 wt.%  $H_2O_i$  are extracted sufficiently fast 448 to form melt rich caps at or near the roof of the magma reservoir (Fig. 8) considering an 449 intermediate crystal size of 3 mm. Magma reservoirs that have a minimum volume of 1000 km<sup>3</sup> 450 can facilitate complete melt segregation for smaller crystal sizes (Fig. 10a). On the other hand, 451 smaller magma bodies (i.e. 100 km<sup>3</sup>) with less than 2 wt.%  $H_2O_i$  are less likely to form any melt-452 rich body. Although magmas with 1 wt.%  $H_2O_i$  spent half of their solidification time within the 453 rheological locking temperature window (Fig. 5d), the high viscosity of the residual melt (i.e. 454 low segregation velocity) does not favour the formation of caps or melt-rich lenses of crystal-455 poor rhyolite under the modelled conditions. The boundaries between caps and lenses, thus, may

456 largely depend on the segregation velocity of the residual melt. Slower segregation of the 457 residual melt, for instance through compaction or through hindered settling with smaller crystals 458 (<3 mm radius; Bachmann and Bergantz, 2004), will decrease the probability of forming caps of 459 crystal-poor rhyolitic melt and increase the probability of forming isolated melt-rich lenses. 460 The onset of melt segregation processes can play an important role in the formation of crystal-461 poor rhyolite (Figs. 9, 10). The likelihood of forming a crystal-poor cap is very high when 462 segregation processes start early within the rheologically locked crystallinity window, and 463 segregation velocities remain elevated (Figs. 6a, 10). Unless magmas are extremely dry (1 wt.% 464  $H_2O_i$ ),  $H_2O_i$  of magma does not seem to play a major role in controlling the architecture of the 465 reservoir and melt distribution when melt extraction occurs at high melt fractions (i.e. 0.6; Fig. 466 9a,b). The timescales of melt accumulation, however, are strongly dependent on the type of 467 segregation process (i.e. hindered settling and compaction) and on  $H_2O_i$ : melts containing higher 468 amounts of water migrate faster (Fig. 9a,b) because of their lower viscosities (Fig. 5b). When the 469 onset of melt segregation occurs at relatively low melt fractions (i.e. 0.3),  $H_2O_i$  may influence 470 whether caps or separate lenses are formed. Water-undersaturated magmas may have a higher 471 chance to form melt caps compared to magmas with higher water contents, because of their 472 prolonged cooling timescales at rheologically locked conditions (Fig. 10c). Overall, at low melt 473 fraction timescales for melt segregation are short, which together with low melt segregation 474 velocities decreases the capacity of these magmas to form thick melt-rich bodies near the roof of 475 the reservoir and the formation of isolated melt-rich lenses becomes more likely.

476

477 **5** Conclusions

The interplay between magma water content, the viscosity of residual melt, the density contrast
between residual melt and crystals, and the time spent within the rheologically locked
crystallinity interval, favoured the extraction of residual melt from the Takidani pluton (Fig. 5).
Our time estimates for melt segregation indicate that the extraction of residual liquids in silicic
reservoirs like the Takidani Pluton occurs over centuries and millennia which is consistent with
other studies (Bachmann and Huber, 2018).

484 While  $H_2O_i$  of magma has a significant control on the segregation timescales of magmatic 485 reservoirs, the architecture of magmatic reservoirs is controlled by the magma crystallinity, 486 crystal size and permeability at the onset of melt segregation processes. The results from our 487 models suggest that hydrous felsic magma are prone to form large melt caps at the roof of a 488 magma reservoir if segregation occurs early at intermediate melt fractions (e.g. 0.6–0.5). 489 However, if segregation starts at low melt fractions (i.e. compaction-driven segregation), silica-490 rich melts may not be able to reach the roof of magma reservoirs because of their low 491 segregation velocity and may form isolated melt lenses within highly crystallised magma. A 492 similar reservoir configuration is likely for water-poor magma, as residual melt segregation 493 require longer timescales (Fig. 7). The drainage of isolated crystal-poor lenses of magma during 494 eruption may account for the chemically heterogeneous nature of rhyolitic eruptions in relatively dry systems such as Yellowstone (Ellis et al., 2014; Wotzlaw et al., 2014). We notice that our 495 496 study focuses on felsic magmas characteristic of some of the large eruptions on Earth; the 497 dependency of viscosity and density of magma and residual melt on chemistry does not warrant 498 the extension of our conclusions to systems of significantly different chemistry. 499 The timescales of melt segregation vary by several orders depending on the crystal size and the 500 mechanism that dominates the extraction process (Fig. 6b). The extremely short timescales

obtained from geochronology and geospeedometry (months to decades; Druitt et al., 2012;
Gualda et al., 2018; Wilson and Charlier, 2016), are difficult to explain by gravity-driven
segregation even considering a larger crystal size (>5 mm).

504 Despite limitations, our calculations provide a framework to deduce some of the additional 505 factors that may influence the capacity of magmatic systems to contain lenses or caps of 506 eruptible felsic melt. For example, large systems such as Yellowstone host well-developed 507 hydrothermal systems, which increases the rate of heat extraction from magmatic systems with 508 respect to what we consider here. This directly decreases the time available for melt segregation 509 (Dutrow et al., 2001), thus increasing the potential for melt to form isolated melt lenses instead 510 of large caps on top of magmatic reservoirs. Ultimately the complex feedbacks between magma 511 chemistry and its physical properties and the thermal evolution of magmatic systems require a 512 multidisciplinary approach (Gualda et al., 2018; Holness, 2018; Huber and Parmigiani, 2018) to 513 identify the potential signs of an impending volcanic eruption. Our results suggest that for 514 hydrous felsic magmatic systems  $H_2O_i$  of magma plays an important role on the extraction of 515 residual melt by affecting both the evolution of the physical properties of magma with 516 temperature and the time magma spends within different temperature intervals.

517

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711 FIGURE CAPTIONS

712

713 Figure 1:

714 Evidence for melt segregation in the upper section of the Takidani Pluton. (left) QEMSCAN

715 images collected from the granodiorite (1; GDT) to the porphyritic portion of the pluton (5;

716 pGT). (top right) Variations of Rb and Sr whole-rock content from GDT to pGT. (bottom right)

717 Interstitial residual melt variations represented by quartz (pink), orthoclase (green) and

718 plagioclase with anorthite content <30 wt.% (orange). The fraction of interstitial residual melt

719 (x<sub>melt</sub>), gradually increases from the GDT unit towards the pGT unit. The content of quartz,

orthoclase and albite, normalised to a fraction of 1, remain relative constant and suggest that the

residual melt composition was buffered at the granitic minimum (Johannes and Holtz, 1996).

722

723 Figure 2:

724 Chemical variability in whole rock (a) and mineral chemistry (b). (a) Assimilation and fractional 725 crystallisation (AFC) models (Hartung et al., 2017) performed on whole rock analyses show that 726 compositional diversity is dominantly produced by crystal fractionation. The grey dashed lines 727 and numbers on the side show the amount of melt (i.e. 60 wt.%) and assimilation (i.e. 3.2 wt.%). The evolution of Rb concentration as function of <sup>86</sup>Sr/<sup>87</sup>Sr is calculated for different bulk 728 729 partition coefficients ( $D_{Rb}$ ). (b) Concentrations of Rb in plagioclase phenocrysts (i.e. sample 730 EH70) increase from 0.54 to 1.31 ppm from core to rim, respectively, and point towards a 731 progressive enrichment of the incompatible element Rb in the melt phase through crystal 732 fractionation. The grey dashed lines and number below indicate the amount of melt fraction. 733

734 Figure 3:

- 735 Comparison between rhyolite-MELTS simulations (lines) and the matrix glass compositions
- (circles) measured between 950°C and 800°C by Costa et al. (2004). The colour contouring
- indicates the initial water content  $(H_2O_i)$  of the starting material (Costa et al. 2004). Rhyolite-
- MELTS simulations and experiments are in broad agreement and show the effect of  $H_2O_i$  on the
- rds chemical evolution (i.e. SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, K<sub>2</sub>O) of residual melt with decreasing temperature.
- 740 No experimental data is available below 800°C.
- 741

742 Figure 4:

743 Results of rhyolite-MELTS simulations for dacitic magma with composition similar to the

744 Takidani granodiorite and the starting material of Costa et al. (2004). (a) Evolution of the water

content of residual melts as function of temperature. (b) Relationship between water content of

the residual melt and melt fraction. (c) Silica content of the residual melt versus temperature. (d)

747 Water content versus silica content of the residual melt.

748

749 Figure 5:

Physical melt fraction and physical properties of residual melt and magma calculated with rhyolite-MELTS. (a) Relationship between melt fraction and temperature. (b) Evolution of melt viscosity (logarithmic) as function of melt fraction. (c) Density difference between crystals and the residual melt as function of melt fraction. (d) Fraction of the total time spent by the magma within each interval of melt fraction. This is calculated by normalising the number of simulation step within a melt fraction unit of 0.02 to the total amount of steps between 1.0 and 0.1 melt fraction assuming constant heat loss. 757 Figure 6:

758 Velocities of melt segregation as function of the melt fraction for magma with different  $H_2O_i$  for 759 hindered settling (a, b) and compaction-driven segregation (c, d). (a) Hindered settling velocity 760 for different  $H_2O_i$  and crystal radius of 3 mm. (b) Hindered settling velocity for water-saturated 761 magma containing crystals of different radii (r). (c) Velocity of compaction-driven segregation 762 for different  $H_2O_i$  and magma with crystal of 3 mm radius. (d) Compaction-driven segregation 763 velocity for water-saturated magma, calculated for different permeability coefficients (K) and 764 crystal radius of 3 mm. Segregation velocities are calculated using the physical properties 765 obtained from rhyolite-MELTS simulations.

766

767 Figure 7:

(a) Vertical position of the isotherms, corresponding to different melt fractions (Mf), along a
 vertical section through the centre of the intrusion as function of time. Reservoir thickness of 0

m presents the top. Location of isotherms are shown for a dacitic magma with  $H_2O_i$  of 2 wt.%.

(b) Maximum segregation timescales for magma reservoirs with different  $H_2O_i$ . Isotherm curves

are shown for a magma reservoir of 1000 km<sup>3</sup> with an aspect ratio (AR) of 10. The maximum

segregation timescales increase with decreasing  $H_2O_i$  of magma.

774

776 Figure 8:

777 Maximum timescales available for segregation for different reservoir volumes,  $H_2O_i$  and aspect 778 ratios of the magma reservoir. Curves were obtained by fitting the thermal modelling results 779 using 2<sup>nd</sup> and 3<sup>rd</sup> order polynomials (supplementary data: Table 3).

780

781 Figure 9:

(a, b) Maximum segregation distance for a reservoir volume of 1000 km<sup>3</sup> and aspect ratio of 10

based on hindered settling and compaction velocities corresponding to an intermediate crystal

radius (i.e. r = 3 mm). (c, d) Time required by the residual melt (extracted by hindered settling

and compaction) to reach the roof of the magma reservoir for reservoirs of 100, 1000 and 10.000

km<sup>3</sup>. The y-axis presents the onset of melt segregation for intervals of 0.1 starting at 0.6, 0.5, 0.4
and 0.3.

788

789 Figure 10:

790 Schematic model for the formation of melt caps and melt pockets for different volumes and  $H_2O_i$ 791 for hindered settling. (a) Full segregation is expected in reservoir  $>100 \text{ km}^3$  for crystal radii >1792 mm for the case that segregation processes start at melt fraction of 0.5 or higher. (b) When melt 793 segregation starts at Mf<0.4 only reservoirs that are larger than 100 km<sup>3</sup> or that have low  $H_2O_i$ 794 are expected to form melt caps. (c) Partial melt segregation and the formation of crystal lenses 795 are most likely to occur if segregation occurs at high crystallinity. Solidification of the reservoir 796 without significant segregation is expected for magmas with  $H_2O_i \ll 2$  wt.%. The Takidani 797 Pluton most likely formed a coherent melt cap that may have been erupted.





























