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2	Volatiles in volcanic systems
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9	Magmas contain only a small mass fraction of volatiles, yet its role in magma dynamics and
10	eruption style is fundamental. Magmatic volatiles partition between liquid, solid and gas
11	phases and in doing so change magma bulk density and compressibility, which have
12	consequences for magma buoyancy and volume. An exsolved fluid phase, which may be
13	distributed unevenly through reservoirs, contains sulfur and metals, which may be
14	transported into the atmosphere or into porphyry deposits. We review the controls on
15	volatile solubility and the methods to reconstruct the volatile budget of magmas, focussing
16	on the exsolved gas phase. We consider the role of exsolved and evolving fluids on magma

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# **19 INTRODUCTION**

dynamics and on eruption style.

Volcanic eruptions, in all their diverse forms, are driven by overpressure, buoyancy and degassing. Subaerial eruptions are associated with vast clouds of magmatic gases, a process that has shaped our hydrosphere over Earth's history. Volcanic eruption deposits, when viewed at high magnification using a scanning electron microscope, are dominated by vesicles: bubbles frozen in place by quenching during eruption (**figure 1**). There can be no doubt that in order to understand volcanic processes, the mechanisms and consequences of magmatic volatile degassing are paramount.

27 Magmatic volatiles are chemical constituents in melts that have relatively low molecular 28 masses and partition into a gas phase at low pressures. The primary magmatic volatiles that drive 29 volcanic eruptions are water (H<sub>2</sub>O) and carbon dioxide (CO<sub>2</sub>). Volatiles are only present in small 30 amounts (a few weight per cent) in magmas, but their influence belies their weight. The presence 31 of dissolved volatiles in silicate melts controls the presence, abundance and composition of 32 crystal phases that grow in the magma during cooling and decompression; e.g. hydrous phases 33 such as amphibole and plagioclase with more Ca-rich compositions crystallize in melts with high 34 dissolved  $H_2O$ . In addition, the density of hydrous melts is lower relative to anhydrous melts 35 (Ochs III and Lange, 1999), which may promote magma ascent through the crust (owing to36 greater buoyancy) as well as convection in magma reservoirs.

37 Close to the Earth's surface, in the crust, melts typically reach saturation with respect to 38 vapor (or gas) in silicate melts, such that volatiles exsolve to form a gas phase disseminated in the 39 magma as bubbles. Exsolution may occur through "second boiling" in magma chambers - a 40 result of prolonged cooling and crystallization - and this increases pressure in the chamber 41 (because the surrounding country rocks are relatively incompressible) and may trigger eruptions 42 (Tait et al., 1989). Volatile exsolution also results from the decompression that occurs during 43 magma ascent to the surface ("first boiling"), and this exsolution causes tremendous expansion of 44 bubbly magma, causing it to accelerate up the conduit. The rate and efficiency of exsolution and 45 gas loss during ascent, as well as the rheological properties of the magma, largely control 46 eruption style.

47 The vapor phase (referred to as "gas" at low pressures, and "fluid" at high pressure) in 48 magma reservoirs in the crust is predominantly made up of CO<sub>2</sub> at depth, becoming more H<sub>2</sub>O-49 rich at lower pressures. Magmatic gases also contain a myriad of other chemical species; chief of 50 which are sulfur and halogens. Sulfur partitions strongly into the gas phase at low pressures 51 (Scaillet et al., 1998). In relatively oxidised and lower temperature arc magmas in reservoirs in 52 the upper crust, most of the sulfur in the magma is likely to exist in the gas phase, rather than 53 dissolved in melt (Wallace, 2001). A consequence of this is that explosive volcanic eruptions, 54 which evacuate these reservoirs rapidly, typically release far more sulfur (as sulfur dioxide) than 55 can be accounted for by the amount dissolved in melt inclusions (tiny fragments of melt trapped 56 in growing crystals at depth, prior to eruption). Chlorine, which also partitions into fluids, forms 57 complexes with metals, a key part of the process of formation of Cu porphyry and Au epithermal 58 deposits in the shallow plumbing systems of some arc volcanic systems (Williams-Jones and 59 Heinrich, 2005).

60 The presence of vapor bubbles in magma changes its bulk physical and rheological 61 properties in important ways. In particular, bubbles make magma compressible. Compressible 62 magma responds to injections of new magma, or evacuation of magma during eruption, by 63 contracting or expanding in response, effectively behaving like a "magma sponge" (Rivalta and 64 Segall, 2008), and this behaviour has consequences for eruption longevity and duration (Huppert 65 and Woods, 2002), and volcano monitoring (see Biggs and Pritchard, this issue). The exsolution 66 of H<sub>2</sub>O from melts removes a "network modifier", resulting in the lengthening of chains of silica 67 polyhedra in the melt, increasing melt viscosity. The interplay between exsolution, viscosity

change and outgassing (physical separation of the gas from the magma) during magma ascent iscritical for determining eruption style (Dingwell et al., 1996).

The behavior of exsolved gases in long-lived, vertically-extensive, mush-dominated magmatic systems beneath volcanoes remains poorly understood and is the focus of much recent research. Gas bubbles may be retained in crystal-rich magmas by capillary forces at low gas fractions, may be transported through quasi-brittle fractures at high gas fractions, and might modify the bulk rheological properties of crystal mushes, rendering them able to respond (by mingling and reorganisation) on relatively fast timescales to magma recharge (Huber et al., 2011).

77 The reconstruction of volatile budgets in magmas that feed volcanic eruptions has received much attention from the perspective of understanding climate impacts (Thordarson and 78 79 Self, 1993), mantle volatile systematics (Dixon and Clague, 2001), magma storage conditions 80 (Wallace and Gerlach, 1994) and volcanic processes (Blundy and Cashman, 2005). We have 81 various tools at our disposal for determining volatile concentrations in melts and their exsolution 82 history, including melt inclusion geochemistry, the distribution of volatiles in solid phases, phase 83 equilibrium experiments and thermodynamic models. Evaluating how exsolved fluids are 84 generated and distributed in magma reservoirs, however, is more challenging. Owing to their 85 buoyancy, gases may migrate and segregate from their source magmas. Long-lived reservoirs, 86 which are subject to sporadic recharge and mingling of magmas, may develop complex reservoir 87 architectures over time, with segregated regions of melts, mushes and exsolved gases 88 (Christopher et al., 2015). In this article we review evidence for the formation, distribution and 89 form of exsolved gases in magma reservoirs and their consequences for volcanic processes, both 90 prior to and during eruptions.

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# 92 THE ABUNDANCE AND DISTRIBUTION OF EXSOLVED GAS IN MAGMA93 RESERVOIRS

94 Vapor saturation of silicate melts occurs when the sum of the partial pressures of the 95 dissolved volatiles in a melt is equal to the confining pressure, in which case a multicomponent 96 gas phase will be in equilibrium with the magma. The solubility of the volatiles CO<sub>2</sub> and H<sub>2</sub>O is 97 mainly controlled by pressure (figure 2), and the much lower solubility of  $CO_2$  compared to  $H_2O$ 98 causes gases at higher pressures to be more  $CO_2$  rich and those at lower pressures to be more  $H_2O$ 99 rich. Bubbles are the consequence of vapor-saturation of magmas, and their nucleation and grow 100 in silicate melts accommodates the exsolving vapor phase. Bubble nucleation may be 101 homogeneous (in melt) or heterogeneous (on crystals) and requires volatile supersaturation to overcome surface tension. The extent of supersaturation is usually small and easily achieved in
 decompressing or crystallizing magmas, except in the case of homogeneous nucleation of bubbles
 in crystal-free rhyolitic magma, where strong melt supersaturation in volatiles may develop
 (Mangan and Sisson, 2000).

106 Other volatiles such as sulfur and halogens partition into the exsolved gas phase to 107 varying extents. Sulfur and chlorine partitioning behaviour is well understood for a wide range of 108 oxidation states and melt compositions (Zajacz et al., 2012). In general, experiments indicate that 109 sulfur partitions strongly into the gas phase, particularly for more reducing conditions below the 110 sulfate-sulfide transition (figure 2), caused by the lower solubility of sulfur when it exists as sulfide  $(S^{2-})$  than when it occurs, under more oxidising conditions, as sulfate  $(S^{6+})$ . Saturation of 111 the melt with Fe-sulfide melt or solid or with anhydrite (at more oxidising conditions) limits the 112 113 sulfur concentration in the co-existing gas phase. Chlorine partitions much less strongly into a gas 114 phase at magma chamber conditions than sulfur, and partitioning into the gas phase is much more 115 pronounced for more silica-rich compositions (figure 2) (Zajacz et al., 2012). These chlorine-rich 116 gases are important for transporting metals to the sites of hydrothermal ore deposits. 117 Experimental data are consistent with the idea that for magmatic systems characterised by long-118 lived, evolved magmas recharged by underplating mafic magmas, the gas phase coexisting with the more evolved magma will be more chlorine-rich, whilst the gas phase supplied by the mafic 119 120 magmas will be sulfur-rich, to a degree that depends on oxidation state and whether saturation 121 with respect to a solid sulfur-bearing phase has been reached.

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# 123 Observations constraining vapor-saturation of melts in the crust

124 Mafic, primitive melts in arc settings contain an average of 4 wt% H<sub>2</sub>O (Plank et al., 125 2013) in melt inclusions and have been inferred to contain 3000 ppm to 1 wt% CO<sub>2</sub> (from melt 126 inclusions and from modelling) (Blundy et al., 2010; Wallace, 2005) before significant 127 differentiation occurs. Hotspot primitive basalts contain 0.5 to 1.5 wt% H<sub>2</sub>O, and mid-ocean ridge basalts 0.3-0.5 wt% H<sub>2</sub>O, based on analyses of submarine pillow-rim glasses and in melt 128 inclusions (Dixon et al., 2002). The CO<sub>2</sub> contents of these primitive melts are less well 129 130 constrained (due to degassing of melts prior to eruption and prior to entrapment as melt 131 inclusions) but may reach levels similar to arc basalts, implying that vapor saturation may occur 132 in some settings soon after melts are generated in the mantle and certainly by the mid-crust. The 133 deep fluids released by magma intrusion in arcs have long been linked to flux melting and assimilation of lower crustal rocks and the generation of intermediate magmas (Annen et al., 134

2006) as well as to the linked processes of lower crustal dehydration and formation of granuliteterrains.

137 There is strong evidence that magmas stored in the mid to upper crust prior to eruption are 138 usually vapor-saturated as a consequence of magma differentiation, prolonged storage, and 139 recharge or underplating by CO<sub>2</sub>-rich mafic magma (Wallace, 2001). For some volcanic systems, 140 the geochemistry of melt inclusions has been used to reconstruct the exsolved vapor phase; chief 141 among these is the Long Valley Caldera system in California, USA, through studies of melt 142 inclusions hosted by quartz in the Bishop Tuff (Wallace et al., 1999). Volatiles and trace element 143 concentrations in the melt inclusions are consistent with gas-saturated crystallization. 144 Calculations of the amount of crystallization that took place, based on trace element variations in 145 the melt inclusions, suggest that the magma chamber was zoned with respect to exsolved gas, 146 varying from  $\sim 1 \text{ wt\%}$  exsolved gas near the bottom of the body (at  $\sim 250 \text{ MPa}$ ) to  $\sim 6 \text{ wt\%}$  near 147 the top (at  $\sim 150$  MPa) (Wallace et al., 1999).

148 Another approach to quantify the abundance of exsolved fluids in volcanic magma 149 reservoirs is to compare the mass flux of volcanic gases with the flux of magma erupted. At 150 Soufriere Hills Volcano, Montserrat, measurements of the gas composition and flux during the 151 eruption (1995 to 2011) permitted estimates to be made of 2-8 wt% exsolved gas in the magma 152 prior to eruption (Edmonds et al., 2014), similar to estimates from the Long Valley system 153 (Wallace et al., 1999). This quantity of exsolved volatiles in the magma would impart a 154 significant compressibility to the magma, which would result in only very muted volume changes 155 in the magma reservior in response to either recharge or eruption (figure 3). This muted 156 deformation is in fact observed: the volume decrease (deflation) during periods of eruption 157 measured using a network of GPS receivers around the volcano is only around one tenth of the volume erupted (Elsworth et al., 2008). The presence of significant exsolved volatiles also has the 158 159 effect of greatly increasing eruption longevity owing to the greater compressibility, leading to 160 eruption of a larger mass to relieve the same overpressure (Huppert and Woods, 2002) (figure 3).

161 Observations of large, sulfur dioxide clouds accompanying explosive volcanic eruptions 162 have been proposed to require a pre-eruptive gas phase (containing sulfur) in the magma 163 reservoir (Wallace, 2001). Notably the eruptions associated with the largest sulfur clouds per 164 erupted unit volume of magma appear to be intermediate arc magmas (andesites and dacites) 165 (Wallace, 2001), such as the eruption cloud shown in **figure 3**, from Kasatochi Volcano, Alaska, 166 in 2008. The degree to which sulfur partitions into the gas phase is maximised when the magmas 167 are silica and H<sub>2</sub>O-rich, relatively Fe and alkali-poor, and at low temperature, conditions met by 168 the vast majority of explosive arc eruptions over the last few decades. The opportunity to observe 169 a contrast to this sulfur-rich case may have presented itself recently: the plinian eruption of 170 Chaiten Volcano, Chile, in 2008 was associated with a remarkably sulfur-poor cloud (Carn et al., 171 2009) accompanying the eruption of crystal-poor rhyolite. This low abundance of gaseous sulfur 172 may have been due to the magma being relatively oxidised (i.e. sulfate-dominated), such that 173 sulfur fluid-melt partitioning was inhibited in the presence of anhydrite, as suggested by recent 174 modelling (Masotta et al., 2016). The presence of exsolved gas and the conditions under which it 175 is generated therefore affects directly the sulfur loading of the atmosphere accompanying 176 explosive eruptions. This is of great interest because sulfate aerosol interacts with solar radiation 177 in the stratosphere, causing tropospheric cooling which, for the largest eruptions, may be severe, 178 with mean global cooling of perhaps a few degrees Centigrade over a few years for VEI 6-7 179 eruptions, resulting in implications for climate and our environment.

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# 181 Dynamics of gas-rich magma in the crust and eruption triggering

182 The fluid mechanics of bubbly magma in the crust and the role of bubbles in magma 183 mixing, eruption triggering and the dynamics of magma reservoirs have been studied using analogue materials and numerical modelling. It has long been recognised that mafic magmas 184 185 underplating more silicic, viscous magmas can produce a range of dynamical behaviors. If the 186 mafic magma is vapor-saturated, bubbles may accumulate at the magma interface, lowering the 187 bulk density of the mafic magma, which can induce overturn, mafic enclave formation, or bubble 188 rise up through the interface, depending on the viscosity contrast between the two magmas. It has 189 been suggested that the heating and remobilization of crystal-rich magma bodies in the arc crust 190 may take place by "gas sparging", a process of mafic underplating, quenching and outgassing of a 191 fluid phase that advects heat up through the pore spaces of a sub-solidus crystal-rich magma, thus 192 partially melting it and perhaps triggering it to erupt on the timescales of a few months for relatively small-scale systems (10s of km<sup>3</sup> in size) (Bachmann and Bergantz, 2006). 193

194 A great deal of attention has been devoted to the microphysics of multi-phase crystal mushes in the crust and in particular how a gas phase might be trapped and/or might migrate 195 196 through a crystal-rich mush. The presence of an exsolved gas phase has implications for bulk 197 mush rheological properties: experiments have shown that the presence of only a few weight per 198 cent of exsolved gas reduces the effective bulk viscosity of a crystal-rich magma substantially 199 and induces shear-thinning behaviour, where the viscosity decreases with increasing strain rate. 200 The presence of significant proportions of gas bubbles could make mushes more mobile in 201 response to magma recharge events, potentially allowing large volumes of magma to be tapped in 202 large eruptions; while the removal of such a gas phase (through outgassing) could result in

"viscous death" and the formation of plutonic bodies. Experiments to investigate the mobility of 203 204 such a gas phase through a partially molten mush show that bubbles that are of a similar size to 205 the crystals may move upward through a mush under compaction by splitting and moving around 206 crystals, whilst larger bubbles become trapped in the pore spaces. Injection of gases into crystal-207 rich suspensions in analogue materials shows that gas migration occurs by a range of processes, 208 including viscous fingering and quasi-brittle failure, suggesting that under conditions of high gas 209 or high crystal contents, mushes may behave in a brittle way with a yield strength, allowing gases 210 to migrate relatively rapidly through them (figure 4) (Oppenheimer et al., 2015). These 211 experiments show that crystal mushes and complex magmatic systems in the crust have the 212 ability both to "hold" gas in the liquid layers but also to allow rapid outgassing under different 213 conditions.

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# 215 EXSOLUTION OF VOLATILES DURING ERUPTION

216 Eruptions may be triggered when overpressures exceed the tensile strength of the country 217 rocks, allowing magma to ascend along fractures towards the surface. These overpressures may be caused by magma recharge or second boiling. As magma ascends, pre-existing bubbles will 218 219 grow, or a new population of bubbles may nucleate (for high decompression rates), owing to the 220 continued lowering of H<sub>2</sub>O solubility in silicate melts at low pressures (figure 2). Bubble growth 221 during magma decompression is limited by two main factors: the rate of diffusion of volatiles 222 through melt into bubbles and the rate of viscous deformation of melt as bubbles expand. For high melt viscosities ( $\sim$  10<sup>9</sup> Pas), viscous retardation may limit expansion of bubbles, causing the 223 224 development of overpressure and perhaps Vulcanian explosive activity.

225 This growth of the gas phase driven by H<sub>2</sub>O exsolution has multiple immediate 226 consequences: the exsolution of H<sub>2</sub>O from the silicate melt raises both the melt viscosity 227 (Dingwell et al., 1996) and the solidus temperature, inducing a rapid burst of crystallization; and 228 the magma's bulk density is drastically lowered as a result of gas bubble expansion, particularly 229 in the uppermost few km of the conduit where pressure drops over several orders of magnitude, 230 causing acceleration of the magma up the conduit (through conservation of mass). This 231 combination of processes makes for a rich variety of possible volcanic eruption styles (figure 5), 232 which are dependent on the interplay between magma decompression rate and the rheological 233 properties of the melt. Interestingly, and somewhat counter-intuitively, the outcomes are not very 234 sensitive to the total volatile content of the magma, except perhaps in the case of Strombolian 235 activity (see below).

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# 237 Influence of volatile exsolution and outgassing on volcanic eruption styles

238 For basaltic eruptions, which involve relatively low viscosity melt, eruption style is 239 governed by ascent rate and the dynamics of two phase flow (Houghton et al., 2016). At low 240 ascent rates, bubbles rise through melts in the conduit, inducing an eruptive spectrum from 241 quiescent degassing to effusive activity to mild Strombolian eruptions, accompanied by persistent 242 tropospheric gas plumes. This range of activity also broadly includes magma convection in a 243 conduit, supplying volatiles to the atmosphere through outgassing of rising and bursting bubbles 244 on the surface of the lava lake, followed by sinking of the denser degassed magma (Kazahaya et al., 1994). 245

246 Explosive styles of basaltic volcanic eruptions fall mainly into the categories of Hawaiian 247 and Strombolian (figure 5). Observations, analogue experiments and textural studies (Houghton 248 et al., 2016) demonstrate that these styles display a continuum in eruption intensity and 249 magnitude, with Strombolian eruptions discrete in duration (typically < 100 seconds), with mass fluxes of  $10^2$ - $10^4$  kg/s and Hawaiian eruptions more long-lived (typically > 2 hours), with mass 250 251 fluxes of  $10^4$ - $10^6$  kg/s (Houghton et al., 2016). The differences in duration and vigor are caused 252 by fluctuations in eruption rate driven by the pressure regime in the magma chamber, which has 253 non-linear consequences for two phase flow in the conduit. At low magma ascent rates, rising gas 254 slugs and bubbles dominate, with their mass growth limited by diffusion; Strombolian activity 255 may be related to the bursting of segregated single slugs or trains of bubbles. At Stromboli, Italy, 256 it has been shown that more crystalline magma at the top of the conduit may act as a plug, 257 promoting gas accumulation at shallow depths. Mixing between hotter, primitive melts and more 258 evolved crystal-rich magma might be facilitated by the turbulent rise of gas bubbles and slugs 259 (Lautze and Houghton, 2007). At higher magma ascent rates, continuous vesiculation and 260 expansion causes inertia-dominated magma fragmentation (Namiki and Manga, 2008) and 261 Hawaiian fountaining, which may persist for as long as overpressures in the reservoir remain 262 elevated. Violent Strombolian activity, which is common at basaltic cinder cones, involves simulaneous explosive activity at the top of the cone and lava effuision from vents near the base. 263 264 This style of activity occurs occurs in more H<sub>2</sub>O-rich basaltic magmas at mass fluxes of  $10^4$ - $10^5$ kg/s and is thus intermediate between Strombolian and sub-Plinian regimes (Pioli et al., 2008). 265

For high viscosity magmas, magma decompression rate (governed by magma chamber overpressure) and the rheological properties of the magma control the style of eruption. Here the viscous retardation of bubble growth generates overpressure in bubbles in silica-rich magmas. Magma fragmentation is driven by overpressure overcoming the tensile strength of the surrounding melt, with the fragmentation wave stopping at some critical overpressure threshold at 271 depth in the conduit. For the case of Vulcanian eruptions, the high bulk viscosity of the magma 272 precludes conduit refilling on the timescale of the eruption, rendering the eruption discrete in 273 duration and limited in magnitude. For lower bulk viscosity magmas and large magma chamber 274 overpressures, plinian eruptions, with magma column heights of 10s of km, often penetrating the 275 stratosphere, are driven by continuous magma fragementation and magma flow, refilling the 276 conduit on timescales of eruption (figure 5). The primary mode of magma fragmentation here 277 might be brittle failure caused by rapid strain rates experienced by the rapidly vesiculating and 278 expanding magma (Papale, 1999).

279 At low magma ascent rates, when melt relaxation can keep pace with bubble growth, 280 permeable bubble networks can develop as the melt vesiculates. This gives magma the ability to 281 effectively outgas volatiles both upward into the atmosphere and laterally into shear-fragmented 282 conduit margins and country rocks, thus preventing magma fragmentation and explosive 283 eruptions. It is thought that effective magma permeabilities for outgassing can develop at 284 porosities of around 30% but perhaps at much lower porosities for sheared magmas (Rust and 285 Cashman, 2004). Under these conditions, magmas erupt effusively in the form of steep-sided lava 286 flows or domes.

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# 288 FUTURE PERSPECTIVES

289 It has been proposed that andesitic volcanoes are essentially "ventholes" that allow excess 290 subducted volatiles to be recycled to the surface, where the term "venthole" implies the presence 291 and rise of a free volatile phase all the way down to the zones of arc magma generation 292 (Giggenbach, 1996). While this may be an extreme view, the data for sulfur dioxide emissions 293 from volcanoes discussed above suggests that in most cases, magma bodies in the mid to upper 294 crust not only are vapor saturated but actually need to become gas charged before they can erupt, 295 unless an eruption is "prematurely" triggered by tectonic or fault activity. Indeed, vapor 296 saturation in crustal magma bodies can be seen as the inevitable consequence of the high CO<sub>2</sub> 297 contents of most mantle-derived mafic magmas combined with the relatively low solubility of 298 CO<sub>2</sub> at crustal pressures such that recharge or underplating by basaltic magma causes fluxing of 299 CO<sub>2</sub>-rich gas to occur (Wallace, 2003). The results of this can be seen at many scales – where the 300 differences between systems reflect the distribution, geometry and size of magma bodies in the 301 crust. Within this framework, long term periods of eruption and unrest at volcanoes such as 302 Montserrat or Popocatépetl can be viewed as essentially intrusive events, in which the mass of 303 erupted magma, while potentially devasting to the environment around the volcano, is quite small in comparison to the likely masses of both stored, differentiated magma and mafic, recently 304

intruded magma at depth. Explosive eruptions of much larger magma bodies, by contrast, contain
within them the exsolved gases released from underlying mush zones and recharging mafic
magmas (Parmigiani et al., 2016), and might reflect thousands or even tens of thousands of years
of gas accumulation (Christopher et al., 2015).

309 In light of this, the future for understanding volatiles in magmatic systems is to develop 310 methods for tracking the movement of magmatic gas and fluids independent of magma 311 movement. Methods for doing this include (1) tracking of the concentrations and fluxes of 312 volatile components of very different solubilities, using melt inclusions and volcanic emissions, 313 (2) phase equilibrium experiments and textural studies that can reveal processes such as gas 314 fluxing, (3) seismic, ground deformation and other geophysical methods such as magnetotelluric data, and (4) use of volatile isotopes like <sup>210</sup>Po that can reveal information about the volumes and 315 timescales of degassing magma bodies. 316

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#### 416 Figures

Figure 1: Photomicrographs of volcanic rocks illustrating the prevalence and importance of 417 418 the exsolved gas phase in driving magma expansion and ultimately, volcanic eruptions. The 419 images show products from a diverse range of eruption types for which the style and extent of gas 420 loss are a critical control on eruption style: a) backscattered electron image of pantelleritic pumice from Pantelleria, Italy (vesicles are black); b) transmitted light photomicrograph of a lava 421 422 dome rock from Soufriere Hills Volcano, Montserrat, showing rounded white vesicles, tabular 423 plagioclase, dark brown pleiochroic hornblende and light brown glass; c) backscattered electron 424 image of scoria erupted at Stromboli, Italy. Large, coalesced vesicles are black, plagioclase 425 phenocrysts grey and olivine (on left of image), white.

426 Figure 2: Solubilities of volatiles in silicate melts and the controls on the development of an 427 **exsolved vapor phase**. a)  $H_2O$ - $CO_2$  saturation model, showing the  $CO_2$  and  $H_2O$  concentrations 428 stable in a silicate melt for a range of pressures (Newman and Lowenstern, 2002); b) The solubility of sulfur-bearing phases (in the absence of degassing) in silicate melt as a function of 429 430 oxygen fugacity, showing the fields of typical MORB (in which dominantly sulfide is stable) and arc basalts (in which anhydrite becomes stable) (Jugo, 2009); c) schematic diagram illustrating 431 432 the controls on the partitioning of sulfur between vapor and melt in silicate magmas, illustrating 433 the importance of melt composition and oxidation state on the fluid-melt partition coefficient for 434 sulfur; d) evolution of the melt volatile contents of a typical water-poor ocean island basalt and a 435 water-rich arc dacite with pressure, showing the contrasing behaviour of sulfur in each case. The 436 yellow shaded area is the typical depth range for the magma reservoirs feeding eruptions. Panel 437 on right shows how the gas volume fraction evolves with decompression for each case.

Figure 3: Some physical and chemical consequences of gas-rich magma reservoirs. a) volcanic eruptions are typically associated with large clouds of SO<sub>2</sub>-rich gases. An AIRS image here shows the SO<sub>2</sub> cloud associated with the 2008 eruption of Kasatochi, Alaska (credit F. Prata); b) the presence of gas bubbles in magmas prior to eruption causes magma to be compressible, which results in only muted deformation being observed at the surface prior to and after eruptions, owing to the buffering effect of the compressible gas phase on volume in response to pressure.

**Figure 4**: Gas transport and storage in crystal-rich, recharging magma reservoirs, after (Parmigiani et al., 2016). a) a schematic of a magma reservoir, illustrating that gas may be

- transported at a higher rate (see b) through crystal-rich mushy layers (by fracturing) than through
- 448 liquid-rich layers (by buoyant bubble rise), leading to gas accumulation in liquid-rich regions.
- 449 Eruptions tapping these liquid- and gas-rich regions would be associated with large gas clouds.

450 Figure 5: The effects of outgassing style and magma ascent rate on volcanic eruption style.

451 Images show a representative range of volcanic eruption styles, with styles associated with rapid

452 magma decompression at the top and slow magma decompression at the bottom. Viscosity

453 increases from left to right.







a: Satellite image (AIRS) of a SO<sub>2</sub> cloud associated with the Okmok (2008) eruption

b: An exsolved gas phase in the magma reservoir causes **large sulfur-rich gas clouds** and **muted syn-eruptive ground deformation** 





