



Reconstructing the deep CO₂ degassing behaviour of large basaltic fissure eruptions



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ABSTRACT

The mass of volatiles degassed from volcanic eruptions is often estimated by comparing the volatile concentrations in undegassed glassy melt inclusions with the volatile concentrations in the degassed matrix glass. However, melt inclusions are prone to post-entrapment modification, including diffusive H⁺ loss through the host olivine crystal lattice which lowers the H₂O content of the inclusion, and the degassing of CO₂ into a bubble in response to cooling and crystallisation on the inclusion walls. Such bubbles are very common in olivine-hosted melt inclusions from the AD 1783–1784 Laki eruption, south-east Iceland. We have determined the CO₂ content of these bubbles using micro-Raman spectroscopy, and the CO₂ concentration in the glass by SIMS. Our results show that >90% of the total inclusion CO₂ may be sequestered into the bubble, which demonstrates the importance of measuring the compositions of both vapour bubbles and the glass phase in melt inclusions. We reconstruct the deep degassing path of the Laki magma by using Nb as proxy for the undegassed CO₂ content of the melt inclusions. The substantial CO₂/Nb variation in the Laki melt inclusions (3.8–364) can be explained by concurrent crystallisation and CO₂ degassing in the Laki magmatic system. We calculate the amount of CO₂ lost from individual melt inclusions, assuming CO₂/Nb ≈ 435 for enriched Icelandic mantle and CO₂/Nb ≈ 171 for depleted mantle. Melt inclusions with the greatest saturation pressures have lost the least CO₂ prior to inclusion trapping. At any given saturation pressure, the most enriched melt inclusions have lost the most CO₂, while the most depleted inclusions have lost very little CO₂. Enriched primary melts with high initial CO₂ concentrations are therefore useful for investigating deep degassing behaviour in magmatic systems because a range of melt inclusion saturation pressures are recorded during crystallisation and degassing. Depleted melt inclusions with low initial CO₂ concentrations remain vapour-undersaturated to shallow levels and cannot be used to constrain deep degassing behaviour. The cumulative CO₂ mass release from the Laki magma is determined as a function of pressure and extent of crystallisation. Using an updated petrologic method that takes into account the diversity of primary melts and CO₂ sequestration into vapour bubbles, we calculate the total mass of CO₂ exsolved from the Laki magma to be 304 Mt.

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1. Introduction

The total atmospheric yield of volatile species from volcanic eruptions is often estimated using a petrologic approach, comparing volatile concentrations measured in crystal-hosted melt inclusions with glass from quenched eruption products (e.g. Devine et al., 1984; Sigurdsson et al., 1985; Palais and Sigurdsson, 1989; Sigurdsson, 1990; Métrich et al., 1991; Thordarson et al., 1996). Such calculations rely on the assumption that the melt inclusions with the highest volatile concentrations are representative of pristine, undegassed melts. This assumption is reasonable for soluble

volatile species such as water, sulphur, chlorine and fluorine, which may remain largely dissolved in basaltic magmas to shallow crustal depths (e.g. Schilling et al., 1980; Wallace and Carmichael, 1992; Carroll and Webster, 1994; Webster et al., 1999; Dixon et al., 1995). However, carbon dioxide has limited solubility in basaltic melts, and depending on the initial CO₂ concentration, basaltic liquids may become saturated with a CO₂-rich vapour at depths of 25 km or more (e.g. Pan et al., 1991; Dixon, 1997; Papale, 1999; Shishkina et al., 2010), such that melt inclusions trapped at mid-crustal depths usually do not preserve the initial CO₂ content of the magma (e.g. Fischer and Marty, 2005). This means that, while CO₂ concentrations in melt inclusions can be used to assess the 'erupted' CO₂ budget associated with melt ascent from the magma chamber to the surface, the petrologic method will underestimate

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the total mass of CO₂ degassed from primary mantle melts supplied to the magmatic system and cannot be used to estimate the degree of passive CO₂ release in the lower crust.

1.1. Estimating undegassed volatile concentrations in melt inclusions

Volatile-trace element pairs, such as CO₂ and Nb or H₂O and Ce, have similar partition coefficients in basaltic melts and are expected to exhibit similar geochemical behaviour during the crystallisation of volatile-undersaturated melts (e.g. Michael, 1995; Danyushevsky et al., 2000b; Dixon and Clague, 2001; Saal et al., 2002; Michael and Graham, 2013). This indicates that Nb can be used as a proxy for the undegassed CO₂ content of melts, if the CO₂/Nb of the primary melt is known. This ratio can be estimated from volatile and trace element concentrations measured in rare examples of undegassed melt inclusions, which are expected to exhibit strong positive correlations between volatile species and similarly incompatible trace elements. Such correlations have been reported for CO₂ and Nb for a suite of primitive melt inclusions from the Siqueiros transform fault on the East Pacific Rise (Saal et al., 2002), and were interpreted as evidence that the melt was vapour-undersaturated at the time of inclusion trapping (Saal et al., 2002). Various studies have used CO₂/Nb, H₂O/Ce and other volatile/trace element ratios to estimate magmatic volatile fluxes (Saal et al., 2002; Workman et al., 2006; Cartigny et al., 2008; Koleszar et al., 2009; Shaw et al., 2010; Helo et al., 2011). However, carbon is likely to be heterogeneously distributed in the mantle (e.g. Helo et al., 2011), meaning that the CO₂/Nb estimated for undegassed primary melts varies with location. At the lower end of the global range, the highly depleted Siqueiros melt inclusions record CO₂/Nb = 239 ± 46. At the upper end of the global range, CO₂/Nb values of ~570 have been obtained for MORB glass samples dredged around 14 °N and 34 °N along the Mid-Atlantic Ridge (Cartigny et al., 2008), and CO₂/Nb values of 650–800 have been reported in melt inclusions from Axial seamount, Juan de Fuca Ridge (Helo et al., 2011). The only reported undegassed CO₂/Nb value for Iceland, 314 ± 125, was determined using melt inclusions from the Borgarhraun lava, North Iceland (Hauri et al., 2002). The global variability in CO₂/Nb for undegassed primary melts suggests that CO₂ is slightly more incompatible than Nb during mantle melting. It has recently been suggested that CO₂ has similar geochemical behaviour to Ba or Rb, and CO₂/Ba values of 114 ± 43 have recently been reported for MORB compositions ranging from ultra-depleted to highly enriched (Michael and Graham, 2013). This may indicate that Ba is a better proxy for undegassed CO₂ than Nb, especially since Ba is expected to follow CO₂ more closely during subduction and recycling into the deep mantle (Michael and Graham, 2013).

1.2. Bubble formation in melt inclusions

Volatiles in melt inclusions can be affected strongly by post-entrapment modification and re-equilibration (e.g. Nielsen et al., 1998; Danyushevsky et al., 2000a; Steele-MacInnis et al., 2011; Gaetani et al., 2012). Bubbles in melt inclusions can form in a number of distinct stages. Firstly, shrinkage bubbles form in response to cooling, since the host phenocryst undergoes significantly less thermal contraction than the melt phase (e.g. Roedder, 1979; Lowenstern, 1995, 2003; Schiano, 2003; Métrich and Wallace, 2008). If the melt inclusion is volatile-free, the bubble will be almost a vacuum (Steele-MacInnis et al., 2011). For volatile-bearing melt inclusions, some of the volatiles that were originally dissolved in the silicate melt may exsolve and be sequestered into the bubble (e.g. Roedder, 1979; Skirius et al., 1990; Anderson and Brown, 1993; Cervantes et al., 2002). Bubbles formed by this mechanism are most likely to be formed at the Earth's surface, during

the cooling of erupted magmas. Secondly, bubble growth occurs during post-entrapment crystallisation (PEC) on the melt inclusion walls. This process decreases the internal pressure of the inclusion, which lowers the solubility of volatiles in the melt phase and causes volatile exsolution from the melt (Steele-MacInnis et al., 2011). Thirdly, recent experimental studies have demonstrated that rapid H⁺ diffusion can cause olivine-hosted melt inclusions to re-equilibrate with their carrier liquids within tens of hours at magmatic temperatures of ~1200° (Gaetani et al., 2012; Bucholz et al., 2013). Decompression associated with diffusive H⁺ loss may promote further volatile exsolution, leading to the nucleation of a vapour bubble in a previously bubble-free inclusion, or the growth of a pre-existing bubble. Since CO₂ is relatively insoluble in basaltic melts in comparison to other common volatile species, CO₂ vapour will be preferentially sequestered into the bubble if post-entrapment degassing occurs. The role of shrinkage bubbles in controlling the volatile content of melt inclusions has been largely avoided or ignored in the literature, although there are some notable exceptions (e.g. Kamenetsky et al., 2002; Thomas et al., 2006; Métrich and Wallace, 2008; Hansteen and Klügel, 2008).

1.3. The AD 1783–1784 Laki eruption

The AD 1783–1784 Laki (Skaftár Fires) fissure eruption on Iceland's Eastern Volcanic Zone was one of the largest basaltic fissure eruptions recorded in human history (e.g. Thordarson et al., 2003), and is one of the best-studied small-scale analogues of a flood basalt eruption. The eruption produced 14.7 km³ of lava and ~0.4 km³ dense rock equivalent of tephra from a ~27 km-long fissure that erupted sequentially in 10 en echelon segments (Thordarson and Self, 1993). The eruption was one of the greatest natural atmospheric pollution events of the past ~250 years, loading the atmosphere with 122 Mt sulphur dioxide, ~7.0 Mt hydrochloric acid and ~15.0 Mt hydrofluoric acid in a period of 8 months (Thordarson et al., 1996) and leading to significant climatic effects that were felt across the northern hemisphere (e.g. Thordarson and Self, 2003; Chenet et al., 2005; Oman et al., 2006; Schmidt et al., 2010, 2012).

The presence of vapour bubbles in Laki melt inclusions has been noted in previous studies (e.g. Métrich et al., 1991; Thordarson et al., 1996), making the Laki eruption an ideal case study for investigating the effect of bubble formation on melt inclusion volatile systematics. We present the first study to combine measurements of vapour compositions in these bubbles with volatile concentrations in the glass phase, in order to determine the total volatile content of melt inclusions at the time of inclusion trapping. We use CO₂–H₂O solubility models (Newman and Lowenstern, 2002; Witham et al., 2012) to determine saturation pressures for melt inclusions in order to reconstruct the extent of CO₂ degassing as a function of pressure within the Laki magmatic system, and to assess how CO₂ degassing is coupled to melt mixing and fractional crystallisation for large basaltic fissure eruptions. Finally, we present an updated petrologic method for estimating the total atmospheric yield of CO₂ from magmas, which takes into account both the sequestration of CO₂ into vapour bubbles, and the diversity of parental melts supplied to the magmatic system. This method can be used to assess both the 'erupted' CO₂ budget associated with melt ascent from the magma chamber to the surface, and the mass of CO₂ released through deep, passive degassing in the magmatic system.

2. Sample collection and analytical methods

Fresh, glassy samples of magmatic tephra were obtained from proximal fall deposits around the Laki cone row by digging down

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