Open-vent volcanoes fuelled by depth-integrated magma degassing

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Abstract

Open-vent, persistently degassing volcanoes—such as Stromboli and Etna (Italy), Villarrica (Chile), Bagana and Manam (Papua New Guinea), Fuego and Pacaya (Guatemala) volcanoes—produce high gas fluxes and infrequent violent strombolian or 'paroxysmal' eruptions that erupt very little magma. Here we draw on examples of open-vent volcanic systems to highlight the principal characteristics of their degassing regimes and develop a generic model to explain open-vent degassing in both high and low viscosity magmas and across a range of tectonic settings. Importantly, gas fluxes from open-vent volcanoes are far higher than can be supplied by erupting magma and independent migration of exsolved volatiles is integral to the dynamics of such systems. The composition of volcanic gases emitted from open-vent volcanoes is consistent with its derivation from magma stored over a range of crustal depths that in general requires contributions from both magma decompression (magma ascent and /or convection) and iso- and polybaric second boiling processes. Prolonged crystallisation of water-rich basalts in crustal reservoirs produces a segregated exsolved hydrous volatile phase that may flux through overlying shallow magma reservoirs, modulating heat flux and generating overpressure in the shallow conduit. Small fraction water-rich melts generated in the lower and mid crust may play an important role in advecting volatiles to subvolcanic reservoirs. Excessive gas fluxes at the surface are linked to extensive intrusive magmatic activity and endogenous crustal growth, aided in many cases by extensional tectonics in the crust, which may control the longevity and activity of open-vent volcanoes. There is emerging abundant geophysical evidence for the existence of a segregated exsolved magmatic volatile phase in magma storage regions in the crust. Here we provide a conceptual picture of gas-dominated volcanoes driven by magmatic intrusion and degassing throughout the crust.

Introduction

Open-vent volcanoes are characterised by their persistent outgassing and mildly explosive activity between major eruptions (Andres and Kasgnoc, 1998; Francis et al., 1993; Rose et al., 2013; Vergniolle and Métrich, 2021). Many are well-studied because persistent low-level activity allows access and collection of extended timeseries of monitoring data. Open-vent volcanoes are found in all tectonic settings and are associated with a range of magma compositions and bulk viscosities (some examples—

not an exhaustive list—are shown in **Fig. 1** and **Table S1**). Open-vent volcanoes may be active over millennia – for example, Masaya, Nicaragua (Stix, 2007), Stromboli, Italy (Allard et al., 1994), Etna, Italy (Allard, 1997), Villarrica, Chile (Witter et al., 2004), Yasur Volcano, Vanuatu (Métrich et al., 2011) and Erebus, Antarctica (Oppenheimer et al., 2011) – or years to decades, such as Soufrière Hills, Montserrat (Christopher et al., 2010) and Fuego (Lyons et al., 2010) and Santiaguito, Guatemala (Holland et al., 2011).

Volcanoes that transition from being 'open-vent' to 'closed vent' over years to decades timescales may be classified as 'persistently restless'. For example, Telica Volcano, Nicaragua, transitions between a 'weak seal' and a 'destabilised' state, which may produce phreatomagmatic eruptions (Rodgers et al., 2015; Roman et al., 2019). Long-dormant volcanoes may also convert to open-system behaviour when they reactivate. Reactivation may initiate explosively, as at Santa María volcano in Guatemala (now the location of the open-vent effusion of the Santiaguito flank volcano) (Lamb et al., 2019), or effusively, as at Soufrière Hills volcano, Montserrat (Wadge et al., 2014). Following the initiation of activity in 1995, Soufrière Hills has outgassed continuously for more than 25 years at the time of writing, despite not being in a state of eruption for much of that time (Christopher et al., 2015).

Eruptions of open-vent volcanoes are typically gas-rich and may be highly hazardous. The nature of the eruptive activity varies with magma composition. Mafic stratovolcanoes exhibiting open-vent behaviour—such as the archetypal Stromboli volcano, Italy—exhibit frequent strombolian eruptions punctuated by large violent explosions, or 'paroxysms' (Bertagnini et al., 2011; Métrich et al., 2005; Rosi et al., 2006). Persistent degassing from mafic lava lakes may persist over decades or longer with accompanying strombolian explosions and/or lake overflows or draining events (e.g., Ambrym, Vanuatu; Erta Ale, Ethiopia; Masaya, Nicaragua) (Bouche et al., 2010; Lev et al., 2019; Moussallam et al., 2021). Open system behaviour in more evolved systems is typically accompanied by episodic explosive activity (typically vulcanian or violent strombolian in style depending on the melt composition; Cashman and Sparks, 2013), effusion of viscous lava flows and domes, and/or gas venting episodes (Edmonds and Herd, 2007). The over-arching characteristics of open-vent activity in all settings, however, are that the outgassing flux of volatiles far exceeds the volatiles that can be supplied from degassing of erupted magma and that high levels of outgassing from a central vent continues between eruptions (Andres et al., 1991). Open-vent volcanoes may therefore be thought of as predominantly gas emitters, with the magma that is supplying the outgassing accumulating endogenously in the crust beneath (Allard, 1997; Anderson, 1975; Francis et al., 1993; Giggenbach, 1992; Giggenbach, 1996). Where in the crust the magma accumulates is, however, an open question.

It has also been shown that open-vent volcanoes are the most prodigious volcanic outgassers of volatiles into the atmosphere, worldwide (Andres and Kasgnoc, 1998; Carn et al., 2016). Additionally, extensive records of the outgassing fluxes of open-vent volcanoes from many decades of in situ measurements (Arellano et al., 2021; Carn et al., 2016; Carn et al., 2017; Fioletov et al., 2016) show that outgassing *between* eruptions dominates the volcanic gas budget (Allard, 1997; Carn et al., 2016; Carn et al., 2017). Indeed, satellite-based global observations of SO_2 flux confirm that persistent, or passive, degassing accounts for ~90% of the global outgassing sulfur flux from volcanoes (Carn et al., 2016; Carn et al., 2017) and that most of the top 20 volcanic outgassers, as quantified from UV sensors TOMS (Total Ozone Mapping Spectrometer) and OMI (Ozone Mapping Instrument), may be classified as ‘open-vent’ (**Table S1; Fig. 1**) (Carn et al., 2017).

Background and aims of this review

Outstanding questions related to outgassing from open-vent volcanoes

The most pressing questions surrounding the outgassing of open-vent volcanoes, and the consequent implications for both monitoring and understanding how these volcanoes work, concern the sources and mechanisms of volatile degassing. Volatiles (e.g., H_2O, CO_2) exsolve from magma upon reaching saturation in the silicate melt or by partitioning into a pre-existing exsolved phase (e.g., sulfur, chlorine) (Aiuppa et al., 2008; Candela, 1997; Cashman, 2004; Edmonds and Wallace, 2017; Edmonds and Woods, 2018; Métrich and Wallace, 2008; Wallace, 2005) (**Fig. 2**). Volatile degassing from melts occurs during decompression (sometimes called ‘first boiling’; **Fig. 2a**); this drives undercooling and crystallisation (Cashman and Blundy, 2000) and, as a result of isobaric cooling and crystallisation, second boiling in magma reservoirs in the crust (**Fig. 2b**). For open-vent volcanoes, where large fluxes of gases are sustained with comparatively little magma erupted (**Table S1**), key questions include (1) the extent to which exsolved volatiles derive from first and/or second boiling, (2) mechanisms of volatile transfer upward through the magmatic system (i.e. as an exsolved magmatic volatile phase (hereafter MVP) or retained within volatile-rich melts, and (3) the effect of the volatile transfer mechanism on resulting volcanic activity (**Fig. 2**).

Degassing in the volcanic conduit

A popular model to explain the high observed outgassing fluxes of water, sulfur, CO_2, and halogen species at mafic open-vent volcanoes is bimodal flow driven by convection, whereby buoyant, volatile-rich magma rises up a conduit and degasses; then the denser, gas-free magma sinks over vertical length scales of several kilometres (Kazahaya et al., 1994; Palma et al., 2011b; Shinohara, 2008; Stevenson and Blake, 1998) (**Fig. 3a**). Convective flow has been reproduced in both analogue and numerical experiments (Beckett et al., 2011; Cardoso and Woods, 1999; Huppert and Hallworth, 2007; Molina et al., 2012). A simple Poiseuille flow model of buoyancy-driven ascent of magma in a conduit is given by:

$$Q_{ascend} = \frac{\pi \Delta\rho_{d-a} g r_a^4}{8\mu_a}, \quad (1)$$

(Kazahaya et al., 1994) where Q_{ascend} is the volume flux of ascending magma, g is the gravitational constant, $\Delta\rho_{d-a}$ is the density difference between the bubble-rich magma at depth and the shallow degassed magma, r_a is the effective conduit radius for ascending magma, and μ_a is the viscosity of ascending magma. If no magma is erupted, then Q_{ascend} must be balanced by the volume flux of descending magma minus the volume of the volatiles released to the surface (Kazahaya et al., 1994; Stevenson and Blake, 1998). SO_2 fluxes of 10^2-10^3 tonnes per day (typical of many of the volcanoes highlighted in **Fig. 1** and **table S1**), for example, require magma fluxes in the conduit of ~1-10 m^3/s. Magma flux, in turn, is controlled by the conduit radius (assuming a cylindrical geometry) and the flow velocity, which is a function of magma viscosity and density. If the gas is transported with the magma, maintaining the same gas supply (assuming similar exsolved gas contents) requires magma with a viscosity of 10^8 Pa s to occupy a conduit approximately ten times wider than magma with a viscosity of 10^4 Pa s.

Critically, however, H_2O-rich magmas do not maintain constant viscosities as they ascend because they undergo extensive decompression-induced degassing and consequent crystallisation (Cashman and Blundy, 2000; Lipman et al., 1985; Métrich and Rutherford, 1998). The addition of crystals may increase the magma viscosity by orders of magnitude (Giordano et al., 2014(Lejeune and Richet, 1995)). Ultimately, it is likely that slowly-rising water-rich magmas will entirely crystallise, as observed in lava domes, and therefore convection is unlikely. In lower viscosity magmas, changing the viscosity contrast between the down- and upwelling liquids can affect the geometry of the exchange flow (Beckett et al., 2014). It has been suggested that magma may overturn at various depths before reaching the surface (e.g., Masaya, Nicaragua; (Aiuppa et al., 2018; Stix, 2007). Regardless of the exact location, however, magma overturn within the shallow system requires that degassed magmas accumulate in subsurface storage regions. At Etna, Italy, for example, there is evidence for endogenous accumulation of degassed magma at a rate of $22.9 \pm 13.7 \times 10^6$ m^3 y^{-1} in a storage region between 3 and 10 km beneath the surface (Coppola et al., 2019); whether this magma crystallised *in situ* or degassed higher in the system is not known; a topic debated by a range of authors in the past (Allard, 1997; Steffke et al., 2011).

Alternative models to explain the outgassing flux from open-vent volcanoes invoke the permeable flow of an exsolved volatile phase through magma in the conduit. In viscous magmas, gas flow is governed by bubble connectivity and the development of permeability. In the absence of crystals, permeability development during decompression of a hydrous melt depends on decompression rate, magma composition (viscosity) and shear (e.g., Giachetti et al., 2019; Hurwitz and Navon, 1994; Lindoo et al., 2015; Okamura et al., 2006; 2008; 2013). Experimentally-determined vesicularity thresholds for permeability development vary from ~30 to 80%, depending on the deformation regime (Okumura et al., 2008). Experimental data suggest that efficient, channelised gas flow may occur at depths of a few kilometres through rhyolite melt containing 5 wt% H_2O when subject to a shear strain >8 (Okumura et al., 2008). Addition of crystals may substantially reduce the percolation threshold for system-scale connectivity during vesiculation and may promote efficient gas loss from conduits even at low gas fractions (Colombier et al., 2020; Collombet et al., 2021; deGraffenreid et al., 2019; Lindoo et al., 2017; **Fig. 3b**). Degassing-induced rheological changes in shallow conduit magma may promote brittle fracturing at the conduit walls, providing transient, highly permeable pathways for gas loss (Gaunt et al., 2014; Gonnermann and Manga, 2003; Rust et al., 2004; Tuffen and Dingwell, 2005) (**Fig. 3c**) and generating low frequency seismicity (Iverson, 2008; Neuberg et al., 2006).

In crystal-rich magmas, gas may be trapped in pore spaces between crystals, where it may accumulate until the overpressure generated overcomes the local yield strength of the crystal framework (Belien et al., 2010; Oppenheimer et al., 2015); this presents a mechanism by which gases may accumulate in crystal-rich plugs and subsequently trigger strombolian eruptions (Oppenheimer et al., 2020; Suckale et al., 2016; Woitischek et al., 2020) (**Fig. 3b**). Gas 'hold-up' (accumulation of gas within the magma) occurs when gas supply from depth is balanced by gas loss from the system and may be implicated as a triggering mechanism for paroxysmal eruptions more generally. For example, paroxysmal eruptions are often preceded by increases in the height of the magma column which may be caused by gas retention; the resulting lava effusion from either flank (Stromboli) or summit (Fuego) vents may then trigger decompression of the shallow conduit (Calvari et al., 2011; Liu et al., 2020b; Ripepe et al., 2015). Similarly, correlations between lava lake surface elevations and gas flux at Villarrica (Johnson et al., 2016), Erta Ale (Bouche et al., 2010) and Masaya (Aiuppa et al., 2018; Williams-Jones et al., 2003) for example suggest that temporal fluctuations in deep (> 1-2 km) gas supply may be important in modulating surface activity at open-vent volcanoes and in advecting heat to maintain an open state.

Degassing throughout the magmatic system

The introduction of gas into shallow (top few km) reservoirs and conduits derived from deeper (>2-3 km) in the system requires a mechanism of deep volatile exsolution. The principal source of that deep MVP is crystallisation and second boiling, which can generate the equivalent of several weight percent for andesite and dacite magmas (**Fig. 2b**). The MVP will initially be CO_2-rich, with increasing water for higher degrees of crystallisation (**Fig. 2b**). Once formed, the MVP can migrate upward and out of the crystal-rich magma reservoir and rise towards the surface (Degruyter et al., 2019; Huber et al., 2010; Parmigiani et al., 2017). Given that the ratio of intrusive to extrusive magmatism is thought to be high in all tectonic settings (from an average of 3:1 to 10:1 or higher in many arc regions); (Crisp, 1984; White et al., 2006) and that plutonic rocks are generally volatile-poor (Bachmann et al., 2007), it follows that the volatiles outgassed persistently by open-vent volcanoes likely have their source, at least in part, in second boiling processes in crustal reservoirs. Evidence for this deep (>2-3 km and perhaps extending into the mid or lower crust in some cases) MVP is provided by the "excess sulfur" emissions accompanying large explosive eruptions of arc volcanoes, which have been explained by sulfur partitioning into substantial accumulations of an exsolved MVP in magma reservoirs (Andres et al., 1991; Rose et al., 1982; Scaillet and Pichavant, 2003; Wallace and Gerlach, 1994; Webster et al., 2011; Zajacz et al., 2012). A deep-derived MVP is also implicated in gas fluxing observed in the volatile systematics of many melt inclusion suites (Blundy et al., 2010; Caricchi et al., 2018; Métrich and Wallace, 2008), as well as in the diffuse degassing of CO_2 along rifts (Foley and Fischer, 2017) and other volcanic centres (Werner et al., 2019). Questions remain as to the MVP source depth and mechanism(s) by which a deep-derived MVP segregates and migrates through crustal magma storage regions.

At low melt viscosities and low crystal fractions, bubbles may accumulate in foam layers at the roof zones of eruptible melt lenses (Jaupart and Vergniolle, 1989; Vergniolle and Jaupart, 1986) and on foam collapse, bubbly plumes may be generated (Degruyter et al., 2019; Parmigiani et al., 2016) (**Fig. 3d**). At intermediate crystal fractions in more evolved systems, the MVP generated through deep (>2-3 km) crystallisation and periodic influx from mafic recharge may rise buoyantly through crystal-rich mush via viscous elongate fingering channels, which produce high permeability pathways for a deep MVP phase to percolate (**Fig. 3e**) (Degruyter et al., 2019; Parmigiani et al., 2016). MVP accumulation in melt-rich caps or lenses may aid eruption of crystal-poor rhyolites (Bachmann and Bergantz, 2004). At high crystal fractions, the MVP may become trapped and accumulate in pore spaces between crystals; it may escape on ductile or brittle deformation (capillary fracturing) when the crystal framework is disrupted (Belien et al., 2010; Oppenheimer et al., 2015; Parmigiani et al., 2016) (**Fig. 3f**).

Aims of this paper

It is clear that a universal paradigm is required that applies to all open-vent volcanoes, of all magma types (high and low viscosity), and which addresses important questions such as how and where the MVP forms and its mode of its transport through the magmatic system. We review outgassing from open-vent volcanoes and lay out the characteristic and generic features common to all settings and all magma compositions. In particular, we examine how new insight into the dynamic nature of crustal magma systems, including conceptual models of volcanic-plutonic systems linked by eruptible melt lenses and unstable volatile-rich fluids (Cashman et al., 2017; Christopher et al., 2015), help us to understand persistent outgassing and gas-rich eruptions from open-vent volcanoes. More specifically, we assess the contribution of unerupted magmas and extensional tectonics to the outgassing fluxes observed at open-vent volcanoes. In considering not only the outgassing characteristics, but also the available evidence for the form and extent of the underlying magmatic system using petrology, geophysics and modeling, we propose a new generic picture for understanding the volatile budget of these volcanoes.

Key observations of open-vent volcanic outgassing

Outgassing fluxes from open-vent volcanoes are decoupled from eruptions

Recent observations of volcanic outgassing from space have highlighted the number and diversity of open-vent volcanoes that emit the overwhelming bulk of volcanic gases into the atmosphere every year (**Fig. 1**; **Table S1**). Global satellite-based monitoring of volcanic gas emissions demonstrate unequivocally that >90% of the global outgassing fluxes of sulfur dioxide are produced during 'passive degassing' from an open-vent, where no eruption is taking place (Carn et al., 2017; Fioletov et al., 2016; Werner et al., 2019). These open-vent volcanoes erupt magmas of a wide range of compositions and rheological properties (**Table S1**), from low viscosity basalt to highly viscous crystal-rich andesite. Moreover, as our understanding of volcanic outgassing increases, it is becoming ever clearer that 'excess' volcanic gas (over that which can be supplied by erupting magma) is the norm, rather than the exception (Andres et al., 1991; Francis et al., 1993). Here we review the gas emission systematics from a number of persistently degassing volcanoes with a wide range in magma compositions and rheological properties, eruptive style and setting.

The flux of sulfur dioxide is commonly used as a proxy for the total volatile flux from a volcano (Aiuppa et al., 2008). In most cases, SO_2 makes up 1-10 mol% of the gas phase from open-vent volcanoes, with the bulk of the gas composed of water and CO_2 in variable proportions. These two major gas species (H_2O and CO_2) are not easily measured, however, owing to their large and variable concentrations in the background atmosphere. SO_2, in contrast, has a distinct and strong absorption in the UV region (Hoff and Millan, 1981) and is not present in the background atmosphere, making it ideal for monitoring volcanoes.

Etna Volcano (Italy) is an archetypal 'open-vent' volcano. It has long been observed that the persistent gas fluxes from Etna are too high to be supplied by the erupting magma (Allard, 1997). SO_2 fluxes between 1975 and 1995 varied from <1000 t/d during quiescent degassing to >10,000 t/d during fountaining (Allard, 1997). Since then Etna has continued to outgas at prodigious rates (Andres and Kasgnoc, 1998; Caltabiano et al., 1994; Salerno et al., 2009), with average SO_2 outgassing rates from 2005-2015 determined from space-based inventories showing an average rate of 2039 t/d (Carn et al., 2017). Approximately 25-30% of that SO_2 flux can be accounted for by decompression-driven degassing of erupted magma (**Fig. 4**). The high rate of volatile outgassing has been attributed to continuous, convective bimodal flow, whereby alkali basalts ascend to shallow pressures, degas and then sink back down into the edifice (Allard, 1997; Burton et al., 2003; Kazahaya et al., 1994), although there is little definitive geophysical or geochemical evidence to support this.

Yasur Volcano (Vanuatu) is a persistent and continuous outgasser with small-scale strombolian activity interspersed with larger paroxysms (Kremers et al., 2012; Métrich et al., 2011; Suckale et al., 2016; Woitischek et al., 2020). Anecdotal and historical evidence suggest that continuous degassing has been taking place for several centuries (Métrich et al., 2011). Frequent, strombolian eruptions eject small volumes of crystal-rich trachybasalt generated in shallow reservoirs by ~50% crystallisation of more primitive alkali basalts (Métrich et al., 2011). SO_2 fluxes ranged from 400-700 t/d during field campaigns in 2006, 2010 and 2018, with much of the SO_2 emitted by passive degassing between explosions (Bani and Lardy, 2007; Ilanko et al., 2020; Métrich et al., 2011; Woitischek et al., 2020). Again, the gas budget cannot be accounted for by degassing of erupted magma (**Fig. 4**); instead, the SO_2 flux requires complete degassing of 0.04-0.05 km^3 per year of unerupted magma, which is ~50 times that erupted (Métrich et al., 2011; Woitischek et al., 2020). If the recent measurements are extrapolated to the past, > 4 km^3 degassed magma has accumulated beneath Yasur over the past 100 years (Métrich et al., 2011).

Manam, a basaltic stratovolcano in the Western Bismarck arc, is one of the most active volcanoes in Papua New Guinea. Continuous outgassing from two summit craters has been sustained at least over the past few decades (Carn et al., 2017; Liu et al., 2020a). Sporadic strombolian eruptions produce low-level ash plumes and are punctuated by occasional paroxysmal eruptions involving lava fountaining, lava flows, pyroclastic density currents, and high ash plumes; five explosive events between August 2018 and June 2019 produced >10 km high eruption plumes. Manam is among the most prolific volcanic outgassers globally, with an average SO_2 flux of 1480 t/d between 2005 and 2015 (Carn et al., 2017); and a 2019 campaign measured fluxes ≤ 7660 t/d over several days (Liu et al., 2020a). Assuming an undegassed magmatic sulfur content of ~2000 ppm, this large SO_2 flux requires around 0.33 km^3 of magma to degas every year, which is likely to be an order of magnitude, and perhaps two, more than the erupted volume (the erupted volumes have not yet been quantified) (**Fig. 4**).

Some of the most prolific and /or persistent global outgassers are lava lake volcanoes (Carn et al., 2017), including Nyiragongo and Nyamuragira (Democratic Republic of Congo), Ambrym (Vanuatu), Masaya (Nicaragua), Erebus (Antarctica), Erta Ale (Ethiopia) and Kilauea Volcano (Hawaii, USA). Degassing from the surface of a lava lake takes the form of vigorous bubbling, low fountains, bubble bursting, gas pistoning and overturn and resurfacing phenomena (Allard et al., 2016; Bani et al., 2012; Bouche et al., 2010; Harris et al., 2005; Oppenheimer et al., 2009; Patrick et al., 2016; Swanson et al., 1979), with upwelling and divergence zones providing evidence for rapid lateral magma motion across the lake's surface (Harris, 2008; Harris et al., 2005; Lev et al., 2019; Pering et al., 2019). These observations, as well as the high gas fluxes (a mean of 7356 t/d SO_2 from Ambrym between 2005 and 2015, with peaks reaching >20,000 t/d SO_2) (Bani et al., 2009) and necessity for a continuous heat source to keep the lake above its solidus temperature, have led to prevailing models of bimodal flow in the conduit to supply both sufficient volatiles and heat (Kazahaya et al., 1994; Oppenheimer et al., 2009; Oppenheimer et al., 2004;

Palma et al., 2011a). Although analogue experiments can reproduce bimodal flow (Palma et al., 2011a; Witham and Llewellin, 2006), we note that simple gas fluxing through the lava lake may supply sufficient heat and volatiles to satisfy observational requirements. For example, degassing at the surface of the Erta Ale (Ethiopia) lava lake occurs at fixed positions that are inferred to be directly above the conduit (Bouche et al., 2010). Here visual, thermal and acoustic observations suggest that spherical cap bubbles rise to burst at the surface; bubbly wakes that detach from the bubble bottom generate small fountains and hold sufficient heat to ensure that the lava lake does not cool over time. In this scenario, a deep (> ~ 1 km) source of gas is required, with no requirement for large-scale vertical bimodal flow. Moreover, the dynamics of bubble behaviour within lava lakes may modulate degassing (Qin et al., 2018).

Intermediate composition magmas can also feed open-system vents, as illustrated by Soufrière Hills Volcano, Montserrat and Santiaguito Volcano, Guatemala. Soufriere Hills erupts high viscosity (10^9-10^{12} Pas) crystal-rich andesite (Melnik and Sparks, 2002). In contrast to the mafic systems, the typical eruptive style is lava dome growth interspersed with episodes of vulcanian activity. SO_2 fluxes here have been sustained since the onset of eruptive activity in 1995 (Christopher et al., 2015; Christopher et al., 2010; Edmonds et al., 2010; Edmonds et al., 2014) and high gas emission rates have continued (to at least 2021) since the cessation of eruptive activity in 2011. SO_2 fluxes have fluctuated between <100 and >2500 t/d throughout the eruption (Christopher et al., 2015; Nicholson et al., 2013), with the highest SO_2 fluxes observed immediately after large dome collapses that exposed the conduit (e.g., July 1998, July 2003) (Herd et al., 2005). SO_2 fluxes were high and sustained during periods of both lava dome growth and prolonged (months-years) periods during which the eruption paused (Christopher et al., 2015; Edmonds et al., 2010). Petrological studies indicate that prior to eruption, sulfur solubility in the rhyolite melt was low (<100 ppm) (Edmonds et al., 2001), consistent with partitioning of sulfur into an exsolved MVP in the shallow storage region beneath the volcano (Clémente et al., 2004; Edmonds et al., 2001; Edmonds et al., 2002). The high bulk viscosity of the magma precludes convective flow as a viable mechanism to supply the outgassing fluxes; sustained degassing during eruptive pauses therefore requires both persistent permeable pathways from depth to the surface and tapping of a substantial pre-segregated reservoir of exsolved volatiles (Christopher et al., 2015).

Bagana Volcano (Papua New Guinea) has exhibited long-lived and continuous degassing, perhaps over centuries (McCormick et al., 2012; McCormick Kilbride et al., 2019; Wadge et al., 2018). Bagana's edifice is built of crystal-rich andesite lava flows and tephra (53-58 wt% SiO_2) and eruptive activity is characterised by the emplacement of steep-sided lava flows, pyroclastic density currents and ash-rich explosions (Bultitude and Cooke, 1981; Wadge et al., 2018). Observations (predominantly by satellite radar) indicate that eruptive activity is strongly pulsatory, with eruptive periods separated by periods of repose, throughout which strong degassing continues (Wadge et al., 2018). SO_2 fluxes at Bagana were first measured in 1983 and reported at > 3,000 t/d (McGonigle et al., 2004). A recent global inventory of volcanic SO_2 emissions measured via satellite-mounted UV sensor (Ozone Mapping Instrument, OMI) reported Bagana's mean SO_2 flux as 1380 t/d for the period 2005-2015 (**Table 1**), placing it 3rd in the global ranking of sustained SO_2 fluxes (Carn et al., 2017). The highest SO_2 fluxes occur during eruptive periods (up to 10,000 t/d) but gas emissions remain high (≤ 2500 t/d) during eruptive pauses (McCormick Kilbride et al., 2019). These high gas emissions cannot be supplied by the erupted magma, which has a time-averaged eruption rate of 1 m^3/s (Wadge et al., 2018) (**Fig. 4**). Instead, the observed rates of degassing from 2005-2015 require around 5-6 times the observed magma flux when reasonable water and sulfur melt concentrations for arc magmas are assumed (McCormick Kilbride et al., 2019).

To summarise, although a model of conduit convection may explain persistent degassing at some volcanoes, it does not supply a universal explanation. In particular, the conduit convection model predicts that high viscosity must be compensated by a larger conduit radius in order to supply gas at similar rate to a lower viscosity system, yet there is no observational evidence for a systematic linear relationship between magma viscosity and conduit radius. This problem is exacerbated by the lower solubility of sulfur in rhyolitic melts (Clémente et al., 2004). Additionally, a convective model requires large accumulations of degassed magma in the shallow crust, which poses a substantial space problem for long-lived open-vent systems. Water-rich magmas may completely crystallise during slow ascent, severely inhibiting return flow. A more parsimonious explanation for high gas flux across all volcano types is that gases are supplied from a mixture of shallow (conduit) and deep (> 1-2 km and perhaps as deep as the mid-crust) sources. Importantly, the flux of gases supplied from deeper magma storage regions to the shallow systems has potential to both modulate and trigger eruptive activity and advect heat; this model also allows for degassed magma accumulations to be distributed over a substantial depth range.

Gas compositions at open-vent volcanoes are consistent with mixing between deep and shallow degassing sources

Additional information about how gases are delivered to the surface and from what depth they are sourced comes from measurements of changes in volcanic gas compositions with eruptive activity. There has been immense progress in quantifying the composition of volcanic gas emissions over the past two decades (specifically the relative abundance of H, C, S and Cl species), principally driven by instrumentation development (Aiuppa et al., 2010; Aiuppa et al., 2006; Liu et al., 2020a; Pering et al., 2020; Shinohara et al., 2008). **Fig. 5** shows a compilation of gas composition data from a range of volcanoes, many of which have open vents (not discriminated on the diagram). Volcanic gases are rich in H_2O and CO_2 and arc volcanoes are typically richer in Cl than rift or intraplate volcanoes. Although we do not consider hydrothermal systems here, we note that gases from volcanoes hosting a large hydrothermal system are typically depleted in S and HCl and rich in H_2O and CO_2. Finally, **Fig. 5** shows the large variability in the molar H_2O/CO_2, C/S and S/Cl ratios measured in volcanic gases at the surface.

Before scrutinising the natural data, it is useful to construct a framework for volcanic gas compositions to understand how gas ratios may evolve during a) decompressional degassing (with some crystallisation) and b) isobaric equilibrium crystallisation in a magma storage region in the crust (second boiling). We use *MagmaSat* (Ghiorso and Gualda, 2015b) to model the solubility of H_2O and CO_2 under different pressure, temperature and oxygen fugacity conditions (as shown in **Fig. 2**). Three examples are considered – Yasur, Stromboli and Soufrière Hills – using appropriate basaltic and andesitic compositions (typical erupted magma compositions for three examples are given in **table S2**, **supplementary material**). For example, to initialise a decompressional degassing model for Yasur trachybasalt (**Fig. 6a**) we use a water content of 1 wt% and a CO_2 content of 0.2 wt%, consistent with petrological studies of melt inclusion compositions (Métrich et al., 2011). Initial melt volatile contents are further modified by crystallisation during magma ascent, which we model using RhyoliteMelts (Ghiorso and Gualda, 2015a). We model chlorine and sulfur exsolution using both a closed and open system partitioning model (see **supplementary material** for details).

We use a suite of D_{Cl} (fluid-melt partition coefficient for chlorine) values collated from the literature (Kilinc and Burnham, 1972; Lesne et al., 2011; Shinohara, 1994; Tattitch et al., 2021; Webster et al., 1999; Webster et al., 2017). D_{Cl} is low (<10) for basaltic compositions and decreases as pressure decreases (Tattitch et al., 2021), although the solubility behavior of Cl is complex and varies with melt composition (Métrich and Rutherford, 1992; Métrich and Rutherford, 1998; Signorelli and Carroll, 2002), fluid composition (Botcharnikov et al., 2004), temperature, oxygen fugacity and pressure (Botcharnikov et al., 2004; Webster et al., 2009); a review is presented in the **supplementary material**. Some studies have postulated an inverse relationship between D_{Cl} and pressure, i.e. that D_{Cl} decreases with increased pressure (Alletti et al., 2009; Shinohara, 2009); this is explained by the large and negative pressure dependence of NaCl partitioning into a melt and the HCl–NaCl exchange reaction between a silicate melt and an aqueous fluid, which favors HCl in aqueous fluids at lower pressures (Shinohara, 2009). These pressure dependencies cause chlorine to appear as HCl in low pressure (~0.1 MPa) volcanic gases and NaCl in high pressure (~50 MPa) fluids. More work is required, however, to fully understand the implications of Cl speciation on fluid-melt partitioning (Shinohara, 2009).

We use D_S (fluid-melt partition coefficient for sulfur) values derived from experiments at high pressure and temperature using natural basalt samples from Masaya and Stromboli (Lesne et al., 2011), which range from 1 to 5 at pressures >200 MPa and >100 for pressures <50 MPa (**Fig. 6a**). Lesne et al. (2011) used synthetic samples based on natural Stromboli melts with an initial volatile inventory representing the most volatile-rich melt inclusions from each volcano. For more evolved compositions, we use partition coefficients derived from experiments (Botcharnikov et al., 2004; Botcharnikov et al., 2015; Webster et al., 2011). These indicate that the fluid-melt partition coefficient for sulfur increases with melt differentiation, reaching values of >500 for rhyolitic melts, and likely increases as the melt water content decreases during decompression (Moune et al., 2009). Model results for Yasur, Stromboli and Soufriere Hills are shown in **tables S3, S4, S5**.

A second set of models (a Yasur example is shown in **Fig. 6b**) simulates isobaric, closed system degassing during crystallisation for four example pressures between 80 and 350 MPa, thus representing magma stored in the crust that undergoes second boiling (details of the model are given in the **supplementary material;** results are shown in **tables S6, S7 and S8**). The melt concentrations of H_2O, CO_2, Cl and S, together with the molar fractions of each in the exsolved volatile phase, are shown in **Fig. 6(v) and 6(vi)** as a function of melt fraction. The observed compositions of the volcanic gas at Yasur are shown in **Fig. 6(iii) and 6(vi)** for comparison (Métrich et al., 2011; Woitischek et al., 2020); and glass compositions are shown in **Fig. 6(ii) and 6(v)** (Métrich et al., 2011). It is important to note that the models shown here incorporate a significant amount of uncertainty; we use them to define the general trends expected for magma degassing under a range of conditions.

The degassing behaviour of Yasur magmas (**Fig. 6**) shows an exsolved volatile phase that evolves from carbon- and chlorine-rich compositions at high pressures, to sulfur- and water-rich compositions at low pressures, consistent with our understanding of the effect of pressure on solubility and partitioning (**Fig. 6a, b; table S3**). Exsolved volatile phase C/S ratios attain a maximum (of > 300 for closed system degassing and ~ 10 for open system degassing) at pressures of 100-230 MPa (**Fig. 6(iii)**). Sulfur is preferentially exsolved over Cl at low pressures, leading to a sharp increase in exsolved volatile phase S/Cl ratios and a sharp drop in the S/Cl ratios of melts at P < ~100MPa (**Fig. 6(ii, iii)**). The ranges in X_{melt}^{S} and X_{melt}^{Cl} thus derived match well with ranges of these elements preserved in melt inclusions and matrix glasses from Yasur (**Fig. 6(ii)**) (Métrich et al., 2011). More generally, model predictions are consistent with published measurements of volatile concentrations in melt inclusion and groundmass glasses at Stromboli, Yasur and Etna (Métrich et al., 2011; Métrich et al., 2010; Métrich and Wallace, 2008; Spilliaert et al., 2006) and observed in the closed system experiments of Lesne et al. (2011).

The exsolved volatile phase is expected to have a molar C/S of 10-100 at pressures >100 MPa, decreasing from ~10 at 80 MPa to ~1 at the surface. Volcanic gases at Yasur have a molar C/S of ~2-3 (Métrich et al., 2011; Woitischek et al., 2020), consistent with gases being sourced from integrated, open system degassing of the entire magma column to a pressure of 80 MPa (< 3 km depth). Open system degassing is expected; the basalt has a low viscosity (<1000 Pa s; (Giordano et al., 2008)) and bursting of large bubbles at the surface is the dominant style of activity (Kremers et al., 2012; Woitischek et al., 2020). Volcanic gases have a molar S/Cl ratio of ~ 0.5 to 30 (Woitischek et al., 2020; Métrich et al., 2011; Oppenheimer et al., 2006); this is again consistent with open system degassing of the entire magma column to a pressure of 80 MPa (**Fig. 6(iii)**).

Importantly, observations of volcanic gases, whilst consistent with models of decompressional degassing, are also consistent with a fraction of the gases being derived from a deep (>2-3 km) exsolved volatile phase generated during prolonged crystallization (**Fig. 6b**). In this scenario, as magma evolves from basalt to trachybasalt (at 80 MPa, after about 50% crystallization) it generates ~ 0.6 wt% exsolved fluids. The exsolved volatile phase is carbon and chlorine-rich at melt fractions ≥~ 0.7 (**Fig. 6(vi))**, then 13rystalliz at molar C/S ratios of ~1.5-2.5 and molar S/Cl ratios of 1-2. These values are consistent with volcanic gas compositions observed at the surface (**Fig. 7a**), which raises the possibility that some, and perhaps a large fraction, of the gases fluxing through the conduit and into the atmosphere may be derived from the fluids produced during equilibrium 13rystallization of basalts at depths of ~3 km or deeper. Indeed, Métrich et al. (2011) concluded from melt inclusion geochemistry that primitive basalts pond at ~ 3 km depth where they fractionate during ~ 50-60% crystallization to form trachybasalts with 56-60 wt% SiO_2. 50% equilibrium crystallization would produce ~1000 ppm exsolved S, ~1000 ppm exsolved Cl and 0.3 to 0.6 wt% H_2O (**Table S2**; **Supplementary Material**); this would require the intrusion of 0.04-0.09 km^3 magma/year, similar to the volume required for the postulated vertical large-scale convection to shallow depths necessary to supply outgassed SO_2 and HCl. Furthermore, the latent heat generated by extensive shallow magma crystallization may be sufficient to thermally buffer the magma reservoir and to maintain a hot conduit (Métrich et al., 2011). Ascent of a deep-derived exsolved volatile phase, possibly with subsidiary melt, could advect heat to the conduit, allowing it to remain at a constant temperature over decadal timescales.

Importantly, fluids generated by second boiling would be relatively water-poor owing to the relatively low water content of Yasur basalts and the high solubility of water in silicate melts. The high water contents of Yasur volcanic gases (Métrich et al., 2011; Woitischek et al., 2020) would thus seem to be good evidence for some decompressional degassing and magma convection. The water content of the volcanic gases is, however, an order of magnitude higher than expected from decompressional degassing alone, which may suggest a contribution from meteoric waters. In summary, it is likely that the volcanic gases emitted to the atmosphere record mixing between exsolved volatiles generated by deep (>2-3 km) isobaric second boiling and by decompression-degassing accompanying convection, with the possible addition of shallow meteoric water, although this is not well constrained (**Fig. 7a**).

Gas data for Stromboli volcano (**Fig. 7b)** illustrate the wide range of gas compositions observed during eruptive activity (Aiuppa et al., 2010; Allard, 2010; Burton et al., 2007; Tamburello et al., 2012). Stromboli's volcanic gases are dominated by H_2O (48 to 98 mol%, mean 80 mol%) and also contain CO_2 (2-50 mol%, mean 17 mol%) and SO_2 (0.2 to 14 mol%, mean 3 mol%). During paroxysms and strombolian explosions, the carbon content of the emitted gases extends to 50 mol% CO_2 with a molar C/S of >10 and up to 47, a low molar H_2O/CO_2 (typically 1-3) and high S/Cl ratios (mean 4.7 ± 0.08).

Between explosions, the gas molar C/S is <15, H_2O/CO_2 is 1.5 to 6.5 and S/Cl is 1-1.5. Stromboli is fed by magmas with a much higher volatile content than at Yasur, as evidenced by studies of melt inclusions (Métrich et al., 2010). As an approximation of the Stromboli system, we use a starting basalt composition (**Supplementary Table S1**) with 3 wt% H_2O, 2 wt% CO_2, 0.2 wt% Cl and 0.25 wt% S (Métrich et al., 2010) for the modeling (details given in **supplementary material**).

As for Yasur, and consistent with previous studies (Aiuppa et al., 2010; Allard, 2010; Métrich et al., 2010), we find that decompressional degassing of the exsolved volatile phase for Stromboli compositions causes the C/S ratio to decrease from >100 at pressures between 240 and 100 MPa to ~1-2 at the surface (**Fig. 7b; table S4**). Also similar to Yasur, the volcanic gas molar S/Cl ratio increases with decreasing pressure from <0.1 at depth to 1-10 at the surface, governed by the relative partitioning behaviour of Cl and S with pressure (Lesne et al., 2011; Tattitch et al., 2021). The fluids generated during isobaric crystallization (second boiling), in contrast, initially have low C/S and S/Cl but converge on C/S ~5-8 and S/Cl ~ 1-2 after 50% crystallisation.

The high molar CO_2 content of the gases during strombolian explosions and paroxysms suggest triggering by a deep-derived gas phase (Aiuppa et al., 2010; Allard, 2010; Burton et al., 2007; Métrich et al., 2010), with the gases emitted during quiescent degassing fed by more shallowly-equilibrated gases. However, a comparison of gas compositions to a decompressional degassing model (**Fig. 7b**) shows that the SO_2/HCl systematics (Burton et al., 2007) are not obviously consistent with such an interpretation. Indeed, decompressional degassing models predict 'deeper'-equilibrated gases to have a lower S/Cl than shallow-equilibrated gases; this trend reflects the decrease in the fluid-melt partition coefficient for Cl with decreasing pressure, in tandem with a dramatic increase in the fluid-melt partition coefficient for sulfur (Lesne et al., 2011). As noted above, we have only a limited understanding of the chlorine systematics in volcanic gases. However, a likely explanation is that degassing during paroxysms is more 'closed' than during persistent degassing, consistent with the higher observed molar S/Cl as well as the high molar C/S. The observed S/Cl ratio of ~ 2 of the quiescent plume at Stromboli (Burton et al., 2007), which accounts for the bulk of the outgassing flux (Allard et al., 2008), is equally consistent with an exsolved volatile phase being generated by decompression degassing or by second boiling processes at depth, or a mixture of both sources (**Fig. 7b**).

Petrological studies provide additional constraints on the Stromboli magmatic system. Stromboli is fed by primitive, volatile-rich high K_2O (HK) basalts with 49-51 wt% SiO_2 and CaO/Al_2O_3 > 0.6 (Métrich et al., 2010) stored at depths of 7-10 km beneath the summit (Bertagnini et al., 2003; Métrich et al., 2010). Large paroxysms erupt this low-density CO_2-rich HK basalt as 'golden pumice' (Pichavant et al., 2009; Rosi et al., 2000), with little evidence for mixing with shallow-stored magma, consistent with rapid and primarily closed system decompression (Métrich et al., 2021; Pichavant et al., 2009). Eponymous strombolian activity, in contrast, ejects crystal-rich, degassed shoshonitic basalt (51-54 wt% SiO_2) stored at 2-4 km beneath the summit and produced by 20-30% fractional crystallisation of HK basalts at depth (Landi et al., 2004; Métrich et al., 2010; Métrich et al., 2001; Métrich et al., 2005; Vergniolle and Métrich, 2021). Deep and shallow magmas mix only during smaller paroxysms (LaFelice and Landi, 2011a). CO_2-rich fluids derived from ponding and crystallising basalts at depth, in contrast, flux through the shallow system, dehydrating the overlying magma and promoting extensive crystallisation within the shallow conduit (Landi et al., 2004; Métrich et al., 2001). Resulting crystal networks may trap rising fluids to form gas pockets; the release of these accumulated gases when they overcome the forces holding the crystals together (the effective yield strength) may explain the 'normal' strombolian activity (Barth et al., 2019; Belien et al., 2010; Oppenheimer et al., 2015; Suckale et al., 2016; Woitischek et al., 2020) that produces highly degassed, crystalline and high viscosity bombs, remnants of the degassed 'plug' (Caracciolo et al., 2021; Gurioli et al., 2014; Lautze and Houghton, 2007). Triggers for paroxysmal activity, in contrast, are debated. One suggestion is that they may be triggered by rapid (days) ascent of HK magma (La Felice and Landi, 2011; Métrich et al., 2010; Métrich et al., 2021) caused by increases in overpressure in the deep storage area or by the greater buoyancy of gas-rich basaltic magma (Allard, 2010; Métrich et al., 2005; Métrich et al., 2021). A 'top-down' trigger has been suggested for paroxysms preceded by high gas hold-up and lava effusion, which promote decompression of the shallow conduit (Calvari et al., 2011; Ripepe et al., 2015). These contrasting scenarios raise interesting questions about the role of deep volatiles in modulating eruptive activity.

Volcanic gas compositions measured at Mount Etna (Italy) reveal that paroxysmal phases of ash emission and lava fountaining during 2001 (Aiuppa et al., 2002) and mid- and late November 2002 (Aiuppa et al., 2004) were accompanied by volcanic gases with low molar S/Cl ratios (< 1) and high SO_2 fluxes (15,000 t/d). Conversely, a trend of increasing S/Cl ratios and decreasing SO_2 flux accompanied the transition of volcanic activity toward mild strombolian activity and finally passive degassing with minor effusive activity. A sulfur and halogen degassing model developed to explain these trends (Aiuppa, 2009) suggest that the S/Cl ratio in the gas phase increases by decompression degassing as magma nears the surface because of the increasing preference of Cl for the melt and of S for the gas (see **Fig. 7b**), as observed in geochemical studies (Spilliaert et al., 2006). The Cl-rich gas emitted during the paroxysms may therefore represent a deeper exsolved volatile phase, perhaps generated through second boiling processes at depth. Such a fluid phase would fuel the development of deep, volatile-rich melts co-existing with the Cl-rich exsolved volatile phase implicated in driving paroxysms at Etna. Evidence of high S/Cl ratios in volcanic gases during fountaining (Allard et al., 2005), in contrast, may record a large shallow influx of undegassed magma accompanied by relatively shallow degassing at low pressures.

Now that gas geochemical monitoring is commonplace, and often automated, trends prior to explosive eruptions at open-vent volcanoes are increasingly well characterised. Pulses of CO_2 are often observed prior to paroxysms and other forms of explosive eruption, manifest as increases in the C/S ratio days to weeks prior to eruption (Aiuppa et al., 2017; Aiuppa et al., 2007; de Moor et al., 2017). At Villarrica volcano, Chile, for example, an increase in volcanic gas C/S after January 2015 preceded the 3 March 2015 paroxysm (Aiuppa et al., 2017). The same pre-eruptive period saw an increase of >50 m in the height of the persistent lava lake (from 27 February; Johnson et al., 2018), suggesting increased gas hold-up. Similar signals preceded explosive activity at Turrialba Volcano, Costa Rica, in 2014 and 2015, where pulses of deeply derived CO_2-rich gas ($C/S_{total} > 4.5$) have been observed up to 2 weeks before eruptions (de Moor et al., 2016). These signals of ‘deep-derived’ exsolved volatiles, arriving at the surface in the absence of (or preceding) magma, provide further evidence of a significant, exsolved and segregated exsolved volatile phase at depth that is capable of fluxing up through the shallow plumbing system prior to and during explosive eruptions, including paroxysms, supported by studies of gas fluxes and scoria textures, which illustrate degassing-driven mingling between deeper hotter melt and degassed, more crystallined magma derived from the upper parts of the conduit (Gurioli et al., 2008).

At intermediate open-vent volcanic systems, evolved melts with high fractions of exsolved volatiles may dominate the magma reservoir and the contribution of second boiling to the exsolved volatile phase may be far more significant. Although few long time series of volcanic gas compositions exist for these systems, one exception is Asama Volcano in central Japan, a persistently degassing volcano that erupts every few years (Shinohara et al., 2015). Here, periods of high gas flux coincide with periods of eruptions and elevated seismic activity. Low SO_2 emission rates characterise periods of low eruptive activity. SO_2/HCl ratios in the gas are high during eruptive periods and lower during eruptive pauses, a pattern consistent with eruptive periods dominated by decompressional degassing (Shinohara et al., 2015). There is no clear variation in C/S between active and inactive periods (Shinohara et al., 2015). In contrast, Soufrière Hills Volcano, which erupted crystal-rich andesite episodically between 1995 and 2011, showed a clear pattern of molar S/Cl > 1 during eruptive pauses and S/Cl < 1 during eruptive episodes dominated by lava dome building that remained consistent over many years of observation (Christopher et al., 2010; Edmonds et al., 2001). These gas characteristics can be explained by cessation of gaseous HCl flux during eruptive pauses whilst a near-constant (or slowly declining) SO_2 flux is sustained (Christopher et al., 2010). The data are insufficient to assess whether systematic temporal variations in molar C/S exist.

Volcanic gas compositions arising from models of decompressional degassing versus isobaric second boiling are compared with observations in **Fig. 8**. The Soufrière Hills andesite is crystal-rich with a rhyolitic carrier liquid; fluid-melt partition coefficients for chlorine and sulfur are estimated to be ~ 20-30 and ~200-500, respectively, at the pressures of storage prior to eruption; with decreasing pressure, D_{Cl} decreases to ~ 1 and D_s increases to >1000 (Tattitch et al., 2021; Webster et al., 2011) (**Tables S5, S8**). Bulk chlorine and sulfur contents are poorly constrained; we use 0.15 wt% for Cl (informed by melt inclusion studies; (Edmonds et al., 2001) and 0.3 wt% S for initializing the isobaric degassing models at

F=1. In general, the deep MVP generated by second boiling has an initially high molar C/S ratio, which then decreases and converges on a composition of ~ 1-2 after ~ 60% crystallisation at a range of pressures (**Table S8**), and an initially low molar S/Cl ratio that increases and converges on values between 2 and 3 (**Fig. 8a**). These values yield Cl and S melt concentrations of 700-1000 ppm Cl and 50-100 ppm S after 90% crystallisation, consistent with melt inclusion studies of SHV rhyolitic melt inclusions (Edmonds et al., 2001) (**Table S3, supplementary material**). After the 90% crystallisation required to generate rhyolite melt, there is 4 - 7 wt% exsolved water-rich MVP (**supplementary material table S8**) with a molar C/S of ~1. Rhyolitic melts beneath Soufrière Hills Volcano are therefore likely to have significant fractions of MVP that must be migrating to the surface, even during eruptive pauses, to supply the outgassing flux (Christopher et al., 2015).

Rhyolitic melt starting with 8 wt% H_2O, 1 wt% CO_2, 0.1 wt% Cl and 0.01 wt% S (consistent with the melt concentrations measured in melt inclusions, **table S3**) subjected to slow degassing-induced crystallisation yields a Cl-rich volcanic gas (**Fig. 8a; table S5**), consistent with the S-poor melt. Variable mixing between the deep (> 2-3 km) MVP generated during second boiling and a decompression-derived MVP during eruptions could thus yield a volcanic gas with a low S/Cl ratio during eruptive periods (contributions from both deep- and decompression-derived MVP) and a high SO_2 flux with a high S/Cl ratio during eruptive pauses (contributions dominated by the deep MVP generated through second boiling), which is precisely what we observe (Christopher et al., 2010; **Fig. 8b**). This example clearly demonstrates the importance of a segregated deep MVP in sustaining outgassing at more evolved open-vent volcanoes; this mechanism may be generic to other, similar volcanic systems globally (e.g., Tungurahua and Reventador, Ecuador; Bagana volcano, Papua New Guinea).

Geophysical evidence for decoupled flow of an exsolved magmatic volatile phase in the crust

Seismicity related to shallow degassing and eruption

Low frequency (LF, or long period, LP) earthquakes are a common feature of active volcanoes (McNutt and Roman, 2015). When LP earthquakes are closely spaced in time, the signals may merge to form a continuous tremor signal. LP earthquakes (and tremor) are thought to be caused by fluid pressurisation, including the resonant response of fluids in conduits or dykes (Chouet, 1996; Neuberg et al., 2000). Very-long-period (VLP) and ultra-long-period (ULP) events detected using broadband seismometers originate at shallow depths (≤ 1.5 km) and are associated with eruptions or vigorous fumarolic activity (McNutt and Roman, 2015; Sanderson et al., 2010). Although the specific interpretations of VLP and ULP events vary, there is general agreement that they provide evidence of short-term deformation accompanying eruptive activity (Chouet et al., 1999; James et al., 2006; Oppenheimer et al., 2020; Ripepe et al., 2015; Suckale et al., 2016). Recent studies highlight links between seismic signals and degassing flux at many open system volcanoes (Zuccarello et al., 2013). A direct link between VLP signals and strombolian activity was first identified at Stromboli volcano: very long period (VLP) signals sourced from a few hundred metres depth in the conduit were thought to originate from the rise and bursting of large slugs of gas within the conduit (Chouet et al., 1999). More recent data clearly show a VLP signal preceding each event together with synchronous thermal and SO_2 flux signals accompanying each explosion (Gurioli et al., 2014; Tamburello et al., 2012). Although the form of gas transport up the conduit linked to these seismic (and infrasound) signals has long been interpreted as a rising gas slug (Blackburn et al., 1976; Jaupart and Vergniolle, 1988; Ripepe et al., 2001), an alternative model calls for gas accumulation in, and release from, a crystal-rich, shallow plug (Del Bello et al., 2015; Gurioli et al., 2014; Oppenheimer et al., 2015; Suckale et al., 2016). Correlations between VLP events and degassing have also been observed at Etna (Zuccarello et al., 2013), Merapi (Hidayat et al., 2002), Asama (Kazahaya et al., 2011) and Erebus (Aster et al., 2008). Similarly, episodic explosive activity modulated by accumulation and release of a gas phase beneath a rigid or semi-rigid plugs may explain shallow (~300 m) VLP signals at Fuego (Waite et al., 2013) and inflation-deflation cycles and periodic explosions at Santiaguito (Bluth and Rose, 2004; Johnson et al., 2014).

Seismicity and strain signals related to migration of volatiles at depth

Deeper geophysical signals related to the movement or pressurisation by an MVP are limited. MVP-related seismic signals in the upper crust have been observed at Popocatepetl volcano, where VLP signals accompany volcanic degassing bursts at a depth of ~1.5 km. One interpretation is that these signals record the opening of an escape pathway for an exsolved volatile phase that accumulated because of second boiling in a shallow sill (Chouet et al., 2005). Sharper pressure transients associated with expanding gas pockets may generate VLP signals to depths of ≤ 3 km (Arciniega-Ceballos et al., 2008). Another example of upper crustal movement of MVP comes from Soufrière Hills, where strain signals observed during vulcanian explosions and gas emission events record inflation of a shallow conduit and near-simultaneous contraction of deeper magma reservoirs (> 5 km depth) (Hautmann et al., 2014). This strain pattern has been interpreted as rapid upward migration of a buoyant MVP, initiated by a sudden destabilisation of large pockets of already segregated fluid in the magma reservoirs (Christopher et al., 2015; Hautmann et al., 2014; Linde et al., 2010).

Deep long period earthquakes (DLPs) associated with volcanoes have been observed in the mid-lower crust or mantle (Aso and Tsai, 2014; Melnik et al., 2020; Wech et al., 2020). Although their origin is enigmatic, some studies have linked DLPs to an exsolved MVP. A striking example is Mauna Kea, Hawai'i, where more than a million DLPs have been recorded in the past 19 years (Wech et al., 2020). These events are not linked to eruptions but have been ascribed to the second boiling of deep (near-Moho) magma intrusions. Other interpretations of DLPs include thermal stresses set up by cooling magmas (Aso and Tsai, 2014) and rapid changes of magmatic pressure in the lower crust caused by rapid nucleation and growth of gas bubbles in response to the slow upwelling of volatile-saturated magma (Melnik et al., 2020).

The latter explanation for the Klyuchevskoy volcanic group relates to primary melts that may contain ≤ 4 wt% H_2O and 0.6 wt% CO_2, which would cause volatile saturation at 800 MPa (~30 km). Alternatively, these DLPs may record pressurisation of a deep reservoir and consequent transfer of the magma towards the surface. The relatively fast upward migration of long-period activity at Klyuchevskoy (months) suggests that a hydraulic connection is maintained between deep and shallow magmatic reservoirs (Shapiro et al., 2017) and the upward transport includes a large fluid component (Koulakov et al., 2012).

Seismic tomography within the crust beneath volcanoes yields a picture of how melt versus MVP-rich areas may be distributed (Kuznetsov et al., 2017; Londoño et al., 2018; Vargas et al., 2017). A porous medium saturated with gas has a low compression modulus that yields low velocity P-waves but no decrease in S-wave velocity (a low Vp/Vs ratio). High P-wave velocities and low S-wave velocity (high Vp/Vs ratios) may, in contrast, indicate the presence of melts, i.e. an active magma reservoir (Kuznetsov et al., 2017). In this way, repeat tomographic studies can monitor temporal changes in the structure of magmatic systems. At Nevado del Ruiz, Colombia, for example, the distribution of low and high Vp/Vs regions changes on yearly timescales (Londoño et al., 2018; Vargas et al., 2017). Nevado del Ruiz is an open-vent volcano with considerable fluxes of SO_2 emitted continuously (Lages et al., 2018). Here a high Vp/Vs anomaly 2-4 km prior to 2010 is interpreted as a volatile-rich melt reservoir, the lower boundary of which moved upward in 2011-2012 and was replaced by a region of low Vp/Vs, interpreted as a gas-rich region undergoing second boiling; this was associated with intense, persistent outgassing at the surface (Vargas et al., 2017). Tomographic studies of Mt. Spurr, an intermittently open-vent volcano, show finger-shaped seismic anomalies with a high Vp/Vs ratio beneath the location of intensive fumarolic activity in 2004-2005 that are interpreted to represent separate conduits of magma and volatiles (Koulakov et al., 2018). A shallow (0-2 km) region of low Vp/Vs directly above is interpreted as a large-scale degassing event, whereby gases were segregated and migrated up to the surface (Koulakov et al., 2018). Although limited, these studies illustrate the potential for future monitoring of volatile and melt distributions beneath open volcanic systems.

Open-vent persistent volcanic outgassing is promoted in complex, extensional tectonic regions

Open-vent volcanoes that generate high outgassing fluxes (**Fig. 1**) are often located in regions of complex tectonics and local extension. The correspondence between the locations of open-vent volcanoes and major crustal extensional structures highlights the role of tectonics in promoting magma intrusion, MVP segregation, and MVP migration to the surface. Although the processes that modulate MVP behaviour are not well known, the association of persistent degassers with extensional regions suggests that a) extension leads to high intrusive/extrusive magma ratios and therefore provides large upward fluxes of exsolved volatiles through second boiling; b) extension may promote the gravitational segregation of low density

MVP phases in shallow reservoirs, allowing their migration and outgassing; and/or c) faults and shear zones in extensional regions may become permeable pathways for deep fluids (**Fig. 9**).

Many persistently degassing open-vent volcanoes —Popocatepetl (Mexico), Fuego and Pacaya (Guatemala), Turrialba and Poas (Costa Rica), Telica and Masaya (Nicaragua)— are located within grabens along the Central American Volcanic Arc (CAVA) and Trans-Mexican Volcanic Belt (TMVB). For example, the open-vent volcano Masaya lies within a large arc-parallel basin – the Nicaraguan graben – that contains Lake Nicaragua and Lake Managua (Morgan et al., 2008) (**Fig. 1; 9a**). Masaya exhibits cycles of intense outgassing that coincide with lava lake activity (Delmelle et al., 1999; Stoiber et al., 1986) (Delmelle et al., 1999; Stoiber et al., 1986) but few eruptions—there has been no major effusive activity since 1965 (Harris, 2009)—although Masaya has a history of large basaltic plinian eruptions (at 6 ka, 2.1 ka, 1.9 ka; (Pérez et al., 2020; Walker et al., 1993; Williams, 1983). Presently, there is a lava lake at Masaya and evidence for a shallow subvolcanic reservoir (Aiuppa et al., 2018); petrological studies have reconstructed the equilibration pressure of the super-hydrous melts responsible for explosive activity to below the seismic Moho (Pérez et al., 2020). The extension rate in Nicaragua has been estimated from initiation of arc splitting and dating of volcanic products (Plank et al., 2002). The observed crustal thickness of 30-35 km greatly exceeds the ~10 km expected for the estimated 100 km extension over 15 Ma, suggesting that intrusive magmatism has infilled the space created by extension at a rate of 90-180 $km^3/km/Ma$ (Morgan et al., 2008). Moreover, the estimated intrusive flux for Nicaragua is ~100 times the estimated volcanic output rate (Carr et al., 2003). This intrusive/extrusive ratio is much larger than the global average, which is ~ 5:1 (with a range of 1:1 to >35:1) (Crisp, 1984; White et al., 2006). Over the entire arc, regions of greatest extension also have the highest magma productivity and the strongest geochemical slab signature (as demonstrated by geochemical indices Ba/La and Yb/La; (Burkart and Self, 1985; Carr et al., 2003). Nicaragua, specifically, has the largest magma productivity (intrusive and extrusive together), the highest rates of extension and slab flux and the strongest slab melting and source melting signals (Carr et al., 2003). Although it is unclear whether the large magma fluxes are a cause or an effect of upper plate extension, the large fluxes of intrusive magmas beneath the Managua graben allow ample opportunity for extensive second boiling and decompression-degassing and production of a deep exsolved MVP. Venting of these deep-derived fluids advects heat to the shallow system and maintains a hot conduit.

Ambrym, a top-ranking volcanic open-vent outgasser (**Fig. 1, 4**) located in the New Hebrides arc (**Fig. 9b**), is situated in the transition zone between a compressional regime in the central arc (Calmant et al., 2003) and an extensional regime in the south (Beier et al., 2018). The relative motion between the central and neighbouring northern and southern arc segments, respectively, is accommodated by dextral strike-slip zones (Pelletier et al., 1998). Ambrym, with its 12 km wide caldera and the resurgent domes of Marum and Benbow, is located exactly at the transition from regional subsidence to strike-slip faulting

(Picard et al., 1994). Changes in the stress field from compression to extension (plus rotation) has created a complex polybaric magmatic system (Beier et al., 2018), including accumulation of large intrusive volumes, crystallisation in shallow reservoirs and resulting large fluxes of exsolved volatiles that contribute to the persistent outgassing observed at Ambrym (Allard et al., 2016).

Etna has developed on the margin of the Hyblean plateau, the foreland to the Late Tertiary Maghrebian-Calabrian thrust belt, a compressional regime that started extending at ~ 0.5 Ma (Hirn et al., 1997; Laigle and Hirn, 1999) (**Fig. 9c**). Crustal-scale normal faults imaged by reflection seismology extend over 20 km; their size, depth, location and evidence of activity suggests that these faults are the source of large earthquakes, which are associated with enhanced volcanism in time and space (Hirn et al., 1997). The specific location of Etna might be related to extension within a narrow zone of active normal faulting that stretches from the Hyblean Plateau in eastern Sicily to northern Calabria (Monaco et al., 1997). A high seismic velocity zone with a lateral dimension of ~6 km has been imaged beneath the summit at 9-18 km depth (Hirn et al., 1997). This body probably comprises cumulates produced from intrusive magmas, fragments of which are occasionally erupted as cognate xenoliths (Corsaro et al., 2014). This cumulate body likely contains significant volumes of volatile-rich melts generated through second boiling as well as regions dominated by an exsolved volatile phase. These fluids may mix with intruding basalts and ascend to shallow levels in the plumbing system shortly before eruptions, contributing to the large and persistent outgassing fluxes of Etna.

Persistently outgassing volcanoes in extensional (continental) regions include Erta Ale, Oldoinyo Lengai, Nyiragongo and Nyamuragira in the East African Rift and Erebus in the West Antarctic Rift system. A global link between outgassing and tectonics was suggested by Tamburello et al. (2018) to explain the distribution of high CO_2-emitting volcanic areas, which are focused in the extensional regions of arcs and in continental rifts. However, oceanic regions of extension are conspicuous for their lack of persistent volcanic outgassers. Iceland, for example, sits astride the Mid-Atlantic Ridge and has frequent eruptions but has no lava lakes or persistently outgassing conduits. Instead, frequent eruptions follow short periods of unrest (including increased outgassing) and return rapidly to closed-system behaviour once the eruption is over, although diffuse CO_2 degassing between eruptions may be linked to magma intrusions at depth (Ilyinskaya et al., 2018). In this respect activity is more similar to other ocean islands such as Reunion, Galapagos or the Canary islands, where sulfur-rich degassing occurs during, but not between, eruptions (Di Muro et al., 2016).

Depth-integrated magma degassing drives persistent outgassing and eruptive activity at open-vent volcanoes

A conclusion that can be drawn from the data and models presented above is that open-vent volcanoes may be thought of as predominantly gas, rather than lava, emitters. A corollary is that degassed magmas accumulate in the crust beneath open-vent volcanoes, thereby growing the crust endogenously. Open-vent volcanoes often occur in regions of crustal extension, which yield the accommodation space for large volumes of intruded magmas that ultimately form dry plutons once they crystallise, exsolve and lose their volatiles. Open-vent volcanoes are active for decades to millennia; their longevity may be controlled by the tectonics of the crust, which may cause different arc segments to 'switch on and off' over time (de Moor et al., 2017). Eruptions of open-vent volcanoes may be triggered by the ascent of segregated exsolved volatiles that flux through the shallow system, or by volatile-rich melts that migrate rapidly from deeper levels in the crust, exsolving large volumes of volatiles as they ascend. Therefore, although traditionally it has been assumed that magma is the 'carrier' for advecting volatiles—requiring mass balance in the upper crust to account for open-vent outgassing fluxes (i.e., the convection model)—we have shown instead that large volumes of intruded magma at depth, stored at multiple levels throughout the crust, provide a potential source of segregated exsolved volatiles, which inevitably must contribute to the large outgassing fluxes at open-vent volcanoes. Our model both removes the necessity for the volatiles to be supplied by continuous, large-vertical-scale bimodal flow and alleviates the space problem caused by the need to store large volumes of degassed magma within the shallowest parts of the crust.

For basalt-dominated open-vent volcanoes (**Fig. 10a**) with basalt or alkali basalt lava lakes or open-vents (e.g. Stromboli, Yasur, Villarrica, Masaya, Fuego), volatiles may be delivered to the atmosphere through a combination of deep and shallow mechanisms, both consistent with the volcanic gas compositions observed at these volcanoes (**Figs 5-7**). Primitive basalts (which may be saturated in an exsolved volatile phase even at mantle depths in some cases) typically undergo ≥ 50% crystallisation in the crust to produce basaltic andesites or trachybasalts that dominate the shallow storage regions beneath these volcanoes. Exsolved volatiles generated through second boiling may migrate via capillary flow in crystal-rich mush in mid- and upper crustal magma storage regions, accumulating and segregating, perhaps giving rise to pockets of exsolved volatiles that may ascend rapidly to the surface and trigger paroxysms (**Fig. 3**). Primitive melts may be drawn into the conduit in the wake of these pockets of exsolved fluids. Although conduit convection may allow magmas to ascend to near atmospheric pressure, outgas and then sink, convection likely acts in tandem with the fluxing of deep-derived exsolved volatiles through the shallow conduit system. Together these processes may explain much of the outgassing volatile flux, as exemplified by Stromboli (**Fig. 7b**). The balance between convective degassing and deep MVP fluxing likely differs between volcanoes depending on both the depth of magma storage and crystallisation and the total volatile content of the magma. Volatile-rich magmas stored at relatively shallow depths are likely to produce a large volume of exsolved volatiles during even modest amounts of crystallisation. We note that this concept of exsolved volatiles being integrated over large depth ranges in the crust to supply open vent outgassing is consistent with geochemical evidence from volcanic rocks that suggest that melt as well as crystals in magmas mingle over similarly large depth ranges (Cashman and Edmonds, 2019; Ruth et al., 2018). A high magma intrusion rate will buffer the melt composition in the subvolcanic reservoir to produce monotonous erupted compositions and long-lived outgassing. The latent heating generated by extensive subvolcanic crystallisation combined with the rise of deep-derived exsolved volatiles (which efficiently advect heat) may produce sufficient heat to maintain hot conduits and lava lakes.

For intermediate composition open-vent volcanoes (dominated by andesites and dacites) (**Fig. 10b**) (e.g. Bagana, Soufrière Hills, Santiaguito, Anatahan), magma crystallisation over long timescales generates extensive regions of mush. Crystallisation of basalts at lower crustal depths may generate low viscosity hydrous or even 'superhydrous' basaltic andesite or andesite melts, as inferred for Kamchatka (Goltz et al., 2020). These melts may be further enriched in incompatible elements (including volatiles) upon mixing with highly evolved water-rich melt lenses in deep crustal mush. Petrological and experimental studies suggest mid-crustal water contents of 5-11 wt% in basaltic andesites from the Lesser Antilles (Edmonds et al., 2016; Melekhova et al., 2017). These volatile-rich melts may rise up to the mid and upper crust through percolation along grain boundaries or by channelised reactive flow. Intrusion of volatile-rich basaltic andesite into shallower, more evolved mush-dominated reservoirs can induce partial melting, gas sparging (Bachmann and Bergantz, 2006) and/or trigger gravitational destabilisation or eruption (Christopher et al., 2015) . The volatile-rich melts may generate substantial fractions of exsolved volatiles in mid and upper crustal mush-dominated reservoirs, which may accumulate and segregate from their rhyolitic melt lenses over millennia. These volatile-rich lenses may be later tapped by eruptions and drive persistent and long-lived volcanic outgassing. Importantly, bimodal flow and convective degassing are precluded in volcanoes dominated by crystal-rich, hydrous intermediate-composition magma because of extensive decompression-induced crystallisation and resulting high bulk viscosity of the magma. In these systems, persistent degassing requires volatile migration that is independent of magma migration.

A generic model for the degassing regime at open-vent volcanoes brings together our understanding of magmatic crystallisation, mixing and storage processes with our observations of volcanic gas flux and composition at open-vent volcanoes. Intrusive, unerupted magmas crystallising at a range of crustal depths generate a substantial exsolved volatile phase, which is fluxed into the overlying system and up through conduits. Volatile fluxing advects heat and brings with it small volumes of primitive melts that replenish the melt resident in the shallow magma storage and conduit systems. Although basalt-dominated reservoirs may also experience subsidiary convection, convection is unlikely in more evolved, mush-dominated magmatic systems, where the outgassing flux will instead be dominated by the fluxing of a deep-derived MVP generated through second boiling. In this model, large bodies of crystal-rich mush generated through extensive crystallisation remain *in situ* at a range of depths, with no requirement for magmas to convect to atmospheric pressure and back down again. Volcanic gases emitted from these volcanoes are the integrated products of the degassing of melts at a range of crustal depths that have undergone various degrees of crystallisation and mixing.

Conclusions

1. Open-vent volcanoes produce *large outgassing fluxes*, much greater than can be supplied by erupting magmas. Open-vent volcanoes may be thought of as gas vents connecting the mantle and/or crust to the atmosphere.
2. Open-vent volcanoes produce *explosive and gas-rich eruptions*, e.g. violent strombolian, vulcanian, paroxysms, that are triggered by the rise of volatile-rich melts and/or fluxing of segregated exsolved volatiles from deeper mush-dominated magma storage regions.
3. *Volcanic gas compositions at open-vent volcanoes* are likely derived from a mixture of exsolved volatile produced from decompressional degassing, whereby magmas degas during their ascent to atmospheric pressure; and isobaric (or polybaric) second boiling in the crust, which generates a substantial volume of exsolved volatiles during crystallization.
4. High fluxes of deep exsolved volatiles are sourced from the *second boiling of intrusive magmas* in the mid to lower crust. These deep-derived exsolved volatiles flux through shallow volcanic systems, advecting heat, sustaining persistent degassing and triggering eruptions. These processes are particularly important for more evolved, water-rich volcanic systems.
5. *Bimodal flow and magma convection* may operate in low viscosity basaltic systems, which brings magma up to near atmospheric pressure to outgas and then sink back down, but this mechanism acts in tandem with fluxing by a deeper-derived volatile phase. Convection is not likely to be important in more water-rich, more evolved volcanic systems, due to the extensive degassing-induced crystallisation in the conduit, which will stall magma return flow by viscous inhibition.
6. Intrusion and degassing of magma into the crust beneath open-vent volcanoes is accommodated by *extensional tectonics* and the extension plays a role in allowing exsolved fluids to migrate up to the shallow volcanic systems. The location and longevity of open-vent volcanic outgassing and activity is likely controlled by tectonics.
7. Open-vent volcanic outgassing is an integrated product of the *degassing of a vertically-protracted magmatic storage and transport system*, not merely a shallow magma reservoir. A great challenge for volcano monitoring in the future will be to detect and understand both geochemical and geophysical signals from the mid and lower crust to enhance eruption forecasting.
8. Accurate measurements of outgassing volatile and magma fluxes from individual volcanoes and from volcanic regions may greatly improve existing estimates of *intrusive/extrusive magma fluxes and their link to tectonics*.

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Conflicts of interest/Competing interests

There are no conflicts of interest to report.

Availability of data and material

All data generated in this paper through modeling are available in the Supplementary Material.

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Figure captions

Fig. 1 Global distribution of open-vent volcanoes, listed in **Supplementary table S1**, encompassing a broad range of magma compositions and tectonic settings. [1]Average SO_2 flux (2005-2015) from Carn et al (2017)

Fig. 2 Schematic illustration of a volcanic system to illustrate potential sources of outgassing volatiles. Magmas degas volatiles in response to (A) decompression during transit to the surface. The plot shows the concentrations of water (red) and carbon dioxide (blue) in a basaltic melt during decompression to the surface from a pressure of 200 MPa (modelled using MagmaSat; (Ghiorso and Gualda, 2015b). The green shaded area shows the amount of exsolved volatile phase produced during degassing. In this case the basalt has a bulk concentration of 2 wt% H_2O and a range in CO_2 concentrations from 0.1 to 1 wt%. The exsolved volatile phase thus produced may outgas to the atmosphere during eruptions. Magmas also degas in response to (B) crystallisation in magma reservoirs in the crust. Crystallisation drives up the concentrations of volatiles in the residual melt and causes the formation of a substantial exsolved volatile phase after differentiation of the magma to highly evolved compositions. In this closed system degassing model involving crystallisation occurring at pressures of 80 to 350 MPa, a primitive basalt begins (at F=1) with 1 wt% H_2O and 0.1 wt% CO_2. After 50% crystallisation, the magma has reached basaltic andesite composition, and after 80%, approximately dacite composition

Fig. 3 Conceptual frameworks to understand magmatic volatile phase (MVP) segregation from magmas in conduits (a, b, c) and in reservoirs (d, e, f) that may be relevant to open-vent volcanic systems. In conduits, (a) convection is driven by density differences, with volatile-rich melts ascending, vesiculating, outgassing, then sinking (Kazahaya et al., 2004; Palma et al., 2011b; Shinohara et al., 1995; Stevenson and Blake, 1998). Crystals are generated by degassing-induced crystallisation in degassed, sinking melts (Beckett et al., 2014). (b) For open-vent volcanoes exhibiting strombolian activity, volcanic gases may accumulate in shallow crystal-rich plug made up of degassed and crystallised magma (Barth et al., 2019; Belien et al., 2010; Gurioli et al., 2014; Oppenheimer et al., 2015; Suckale et al., 2016; Woitischek et al., 2020); explosions may be caused by overpressure in the gas pockets overcoming the local yield strength in the crystal pack. (c) At low confining pressures and high magma viscosities, there may be sufficient strain at the conduit walls to induce brittle failure, with gas loss along permeable channels (e.g. Santiaguito, Mount St Helens 2004-2006) (Dingwell, 1996; Edmonds and Herd, 2007; Gonnermann and Manga, 2003; Tuffen and Dingwell, 2005). In crustal magma reservoirs, it has been proposed that the MVP may segregate under different regimes depending on magma crystal content. (d) in crystal-poor melt lenses the dominant regime may be buoyant bubble rise, the timescale for which is governed by the density difference between melt and MVP, the melt viscosity and the bubble size (Parmigiani et al., 2016). (e) In more crystal-rich mobile mushes, the MVP may rise buoyantly by viscous fingering, forming interconnected channels which may allow potentially much faster MVP segregation (Parmigiani et al., 2016). (f) In crystal-rich, melt-poor mush the MVP may become trapped in pore spaces, becoming mobilised once a critical overpressure is reached inside the pores, which may induce capillary fracturing (Degruyter et al., 2019; Parmigiani et al., 2016)

Fig. 4 A plot of mean outgassing SO_2 flux (from Carn et al., 2017) against magma flux for a range of volcanoes. The magma flux required to supply the SO_2 flux is shown as a black square and the time-averaged eruption rate is shown as an open square. Estimates of degassing and erupted magma flux are sourced from: Ambrym (Allard et al., 2016); Manam (Liu et al., 2020a); Bagana (McCormick Kilbride et al., 2019; Wadge et al., 2018); Etna (Allard, 1997; Allard et al., 2006); Yasur (Métrich et al., 2011); Masaya (Zurek et al., 2019); and Stromboli (Allard et al., 2008). The individual studies use a combination of melt inclusion evidence and observed gas fluxes to infer the flux of degassing magma; and geological evidence to infer the magma eruption rate; please see papers for detail

Fig. 5 A review of volcanic gas compositions. (a) H_2O–CO_2–S_t (i.e. SO_2 + H_2S) and (b) (b) H_2O–HCl–S_t (i.e. SO_2 + H_2S) gas compositions for a range of volcanoes, made up of both direct sampling (Fischer, 2008; Hammouya et al., 1998; Symonds et al., 1994) and Multigas data (Aiuppa et al., 2008; Aiuppa et al., 2015; Burton et al., 2007; Sawyer et al., 2008; Shinohara and Witter, 2005). Red shaded regions = arc volcano emissions; blue = hydrothermal emissions; and yellow = intraplate/rift emissions

Fig. 6 Models reconstructing the degassing of volatiles during decompression degassing and during isobaric crystallisation of alkali basalt melts at Yasur Volcano, Vanuatu. (a) Decompression of a trachybasalt from Yasur is accompanied by the exsolution of water and CO_2 (using Magmasat; (Ghiorso and Gualda, 2015a) from initial values of 1 wt% and 0.2 wt% respectively, based on melt inclusion and volcanic gas studies (Métrich et al., 2011; Woitischek et al., 2020). Fluid melt partition coefficients for Cl and S are shown in (i), melt volatile concentrations in (ii) and exsolved volatile phase composition in (iii). Open and closed system degassing models are considered, where open system degassing incorporates integration of the gas phase of the magma column at each pressure step. (b) Isobaric crystallisation leads to second boiling through enrichment of the melt in volatiles. Shown here are fluid-melt partition coefficients for Cl and S (i), melt volatile concentrations (ii) and the composition of the exsolved volatile phase (iii) for crystallization models (from a melt fraction, F, of 1 to a melt fraction of 0.1) at pressures of 80, 160, 240 and 350 MPa. The pressures of crystallization are marked to show how the range in compositions links to pressure. Details of the models are given in **supplementary material**. Observed glass compositions (Métrich et al., 2011) and volcanic gas compositions (Oppenheimer et al., 2006; Woitischek et al., 2020) are marked on (a) and (b)

Fig. 7 Composition of the exsolved volatile phase with pressure and melt fraction for (a) Yasur, Vanuatu and (b) Stromboli, Italy. Shown in solid black lines in both (a) and (b) are the decompressional degassing models for open and closed system degassing, marked with some of the pressure steps, in MPa. In dashed and red lines (see legend) are the isobaric second boiling models to describe the exsolved volatile phase produced during equilibrium crystallisation and degassing at various pressures, marked in red with the melt fraction remaining (1 to 0, where 1 is the case where there is no crystallisation, 0 denotes fully crystallised). A yellow box marks the compositions of volcanic gases observed at the surface (Aiuppa et al., 2010; Allard, 2010; Oppenheimer et al., 2006; Woitischek et al., 2020)

Fig. 8 Mixing between a deep-derived MVP, generated through extensive second boiling, and an MVP derived from decompressional degassing may explain the gas systematics at Soufrière Hills Volcano, where high SO_2 fluxes and low S/Cl are observed during dome building and high SO_2 fluxes and high S/Cl during eruptive pauses. (a) the molar S/Cl of the MVP varies with S outgassed (in wt% of the melt+exsolved volatile phase) for isobaric degassing during second boiling and for decompressional degassing of S-poor rhyolite. Melt fraction remaining, F, is marked onto the trajectories for isobaric second boiling. Note the composition of the 'deep' MVP in equilibrium with rhyolite will differ if different bulk magma compositions of sulfur and chlorine are used, but the relative trends shown in the Fig. will remain the same. (b) The volcanic gas compositions at the surface may be explained well by a mixing model whereby a deep MVP generated through second boiling mixes with an MVP generated during decompressional degassing and crystallisation of sulfur-poor crystal-rich andesite (with a rhyolitic melt phase). Depending on the relative sizes of the two MVP reservoirs, the effect of mixing on the volcanic gas composition changes. For equal-sized reservoirs in terms of mass of the MVP phase per unit of magma, a scenario might be envisaged whereby during dome-building the shallow MVP dominates, generating Cl-rich gases; and during eruptive pauses (open-vent degassing) the deep MVP dominates, generating high S/Cl gases and a high SO_2 flux

Fig. 9 Open-vent volcanoes are often in complex, extensional tectonic settings. (a) Masaya, Telica, Fuego and Pacaya, in the Central American Volcanic Arc, are closely located in regions of local crustal extension, associated with the Nicaraguan Depression (Masaya, Telica) and the rotational block tectonics of Guatemala (Fuego, Pacaya). (b) Ambrym is located at the boundary between a compressional and extensional regime in the Hebrides Arc. (c) Mount Etna, Italy, is located in an extension region stretching from Eastern Sicily to the south of Italy (see text for detail). Maps generated using GeoMapApp

Fig. 10 Schematic diagram to illustrate the principal mechanisms of magma degassing at persistently active open-vent volcanoes. a) At basalt-dominated volcanoes, magmas rise to shallow storage regions in the crust to form shallow basic plutons. Some magma may rise and convect in the conduit. The exsolved volatile phase that outgasses quasi-continuously from the volcano is sourced from a mix of deep (second boiling) and shallow (convective degassing) sources. Volcanic activity at these volcanoes is dominated by gas-driven strombolian activity and paroxysms, and there may be a semi-stable lava lake. b) At andesite and dacite-dominated volcanoes, magmas undergo multi-level fractionation in the crust to form evolved melts which rise to shallow storage regions, exsolving a substantial exsolved volatile phase through second boiling. The persistent outgassing observed at these volcanoes is sourced principally from the second boiling process, which takes place during the solidification of hybrid and felsic plutons at depth. In both cases, magma intrusion and open-vent outgassing is promoted by crustal extension, which provides accommodation space for magma intrusion at depth and for the gravitational segregation of lower density exsolved volatile phases to the upper parts of the storage region. Deep generation of superhydrous melts may advect volatiles up to subvolcanic reservoirs