

Understanding Degassing and Transport of CO₂-rich Alkalic Magmas at Ross Island, Antarctica using Olivine-Hosted Melt Inclusions

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ABSTRACT

Volatiles play an important role in magmatic and volcanic processes. Melt inclusions are a powerful tool to study pre-eruptive volatiles, but interpretation of their H₂O and CO₂ variations can be difficult. The H₂O and CO₂ contents of melt inclusions from nine basanites from Hut Point Peninsula, Mt Terror and Mt Bird on Ross Island, Antarctica, were studied to understand better the behavior of volatiles in the magmas and to provide insight into magma transport and storage processes. Ninety olivine-hosted (Fo_{78–88}) melt inclusions were examined along with the composition of the associated bulk-rock samples. The H₂O (0.4–2.0 wt %) and CO₂ (0.2–0.9 wt %, or 0.2–1.8 wt % after correction for vapor bubbles) variations in the melt inclusions cannot be explained by equilibrium degassing. A strong correlation between melt inclusion radius and H₂O content for Hut Point samples indicates that diffusive loss of H⁺ has occurred. Based on vapor saturation pressure trends, it is inferred that a magma reservoir existed below Hut Point at a depth of ~18 km, and by modeling diffusive loss of H⁺ for melt inclusions, it is shown that the magmas ascended from this depth in less than a year. Melt inclusions from Terror and Bird lack evidence of diffusive loss of H⁺ and there are no strong chemical indicators of CO₂ fluxing. Compositional heterogeneities in melt inclusions indicate that magma mixing occurred, making it difficult to interpret H₂O and CO₂ trends. Melt inclusions from these volcanoes were entrapped polybarically, inconsistent with entrapment in a single storage region. Published analyses of 54 olivine-hosted (Fo_{53–83}) melt inclusions in seven samples from Erebus volcano on Ross Island were re-examined for comparative purposes. Low H₂O (~0.1 wt %) and CO₂ (0–0.2 wt %) contents and the evolved compositions of these indicate that Erebus magmas undergo shallow (<6 km) crystallization before eruption, probably in a shallow storage region. Magmas from the surrounding volcanoes show no sign of shallow storage.

Key words: alkali basalt; melt inclusion; Polar Rock Repository; Ross Island; volatile; degassing; magma ascent; magma chamber; diffusion modeling; olivine

INTRODUCTION

Studies of magmatic volatiles offer insight into melt generation, magma transport and evolution, and global volatile cycles (e.g. Grove & Baker, 1984; Allard *et al.*,

1991; Gaetani & Grove, 1998; Dixon *et al.*, 2002; Wallace, 2005). Such efforts require knowledge of the behavior of volatiles in the pre-eruptive magmas, which is a challenge because magmas degas during their

ascent to the surface and retain only a small fraction of their initial volatile contents after eruption. The direct measurement of pre-eruptive volatile concentrations is possible through the study of glass inclusions, known as melt inclusions (MIs). These small (tens of microns) blebs of silicate glass represent melt entrapped in growing crystals during various stages of magma ascent and differentiation, thereby providing a window into the changing physicochemical states of magmas prior to their eruption.

One important goal of MI studies is to elucidate magmatic degassing processes, which may be reflected in the compositions of polybarically entrapped MIs (Wallace, 2005), provided that proper consideration is given to post-entrapment cooling and bubble growth (Steele-MacInnis *et al.*, 2011). Mafic magmas are often considered to follow equilibrium degassing paths (Sparks *et al.*, 1994), which are predicted using models (Newman & Lowenstern, 2002; Papale *et al.*, 2006) and decompression experiments (Lesne *et al.*, 2011). Interestingly, MIs often show variations in H₂O and CO₂ contents that are inconsistent with model predictions (Blundy *et al.*, 2010). The inconsistency is most commonly attributed to processes that affected the magma prior to MI entrapment. Studies often invoke CO₂ fluxing, or buffering of magmatic volatiles by the addition of a CO₂-rich vapor. Disequilibrium degassing has also been suggested (e.g. Gonnermann & Manga, 2005; Pichavant *et al.*, 2013). Recent efforts have underscored the importance of MI-specific mechanisms in causing the inconsistency, such as post-entrapment diffusive loss of H⁺ (e.g. Gaetani *et al.*, 2012; Bucholz *et al.*, 2013; Mironov *et al.*, 2015). Most investigations of the enigmatic H₂O and CO₂ behavior have focused on H₂O-rich arc magmas and/or evolved magma compositions, whereas studies of primitive alkalic systems have been mostly confined to Mt Etna (e.g. Métrich & Clocchiatti, 1989; Spilliaert *et al.*, 2006; Collins *et al.*, 2009). Alkalic magmas provide a critical test for CO₂-fluxing because they typically have high magmatic CO₂ concentrations, inherited from their generation by low degrees of partial melting and possible enriched mantle sources (Sun & McDonough, 1989; Dasgupta *et al.*, 2007).

We report volatile, major and trace element concentrations in olivine-hosted MIs from basanitic tephra and hyaloclastites from Ross Island, Antarctica. Our aim is to understand the behavior of H₂O and CO₂ through studying variations recorded in MIs. These variations are used to investigate ascent timescales and depths of crystallization and mixing. Previous studies of MIs from Ross Island focused on evolved magmas from Erebus and demonstrated the CO₂-rich nature of the magmas and the deviation of H₂O and CO₂ contents from equilibrium degassing paths (Eschenbacher, 1998; Oppenheimer *et al.*, 2011). This study focuses on basanites from Hut Point Peninsula, Mt Terror, and Mt Bird. The new results are compared with the published data from Erebus to evaluate the plumbing system and distribution of volatiles in magmas at Ross Island.

GEOLOGICAL SETTING

Ross Island (Fig. 1) is an alkalic volcanic center located within the West Antarctic Rift System, a broad region of thinned Antarctic continental crust characterized by extension, high heat flow, and volcanism (Behrendt, 1999). North of Ross Island is the Terror Rift, where extension in the West Antarctic Rift System has been focused from the late Miocene to the present (Hall *et al.*, 2007). Finotello *et al.* (2011) used receiver functions to determine crustal thicknesses of 19–27 km below Ross Island (Fig. 1), similar to ~18–25 km crustal thicknesses within the Terror Rift (Watson *et al.*, 2006). Magmatic activity is typically attributed to extensional tectonics (e.g. Rocchi *et al.*, 2005; Cooper *et al.*, 2007) or upwelling of deep mantle (e.g. Kyle *et al.*, 1992; Sims *et al.*, 2008; Gupta *et al.*, 2009).

Over the last ~4 Myr, eruptions of ~4520 km³ of basanite to phonolite, with minor benmoreite and trachyte,

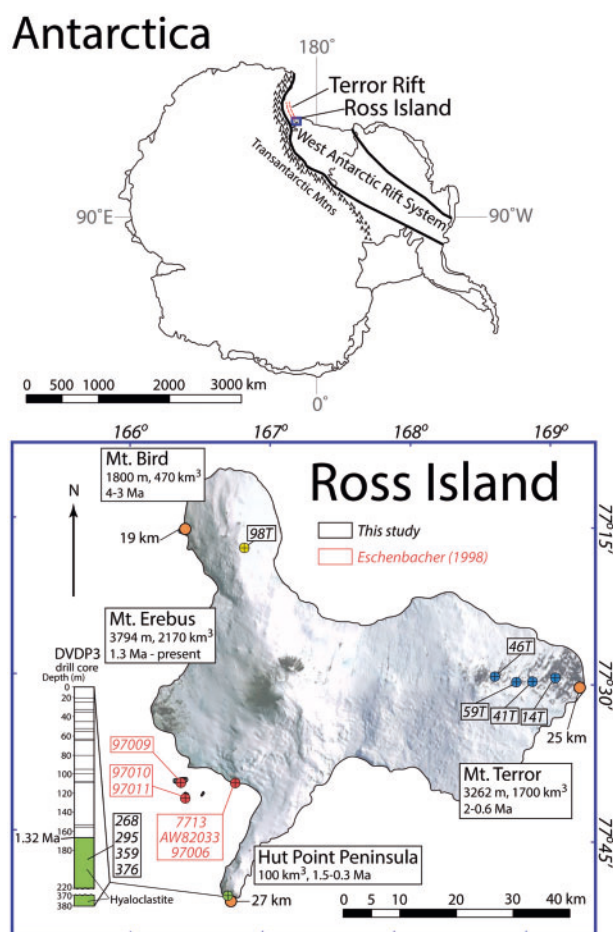


Fig. 1. Ross Island sample location map. DVDP 3 sample numbers correspond to sample depths (in m) in the core. Crustal thicknesses (orange circles) are from Finotello *et al.* (2011). Volumes and ages of the four volcanic centers are from Kyle (1990a) and Esser *et al.* (2004), and the age of the top of the hyaloclastite unit in DVDP 3 is inferred from a correlative unit in DVDP 2 (Kyle *et al.*, 1981a). Samples from Erebus, which include samples from a sea cliff and outlying islands, are those of Eschenbacher (1998). The Antarctica base map and image of Ross Island are courtesy of the U. S. Geological Survey.

have built the four coalesced volcanic centers that form Ross Island—Mount Bird, Mount Terror, Hut Point Peninsula, and Mount Erebus—which are hereafter referred to as Bird, Terror, Hut Point, and Erebus (Armstrong, 1978; Kyle *et al.*, 1992; Esser *et al.*, 2004). Volcanism began at Bird, lasting from ~ 4 to 3 Ma (Kyle, 1990b). Mostly basanitic eruptions formed the 1800 m high shield volcano, which has parasitic basanitic and phonolitic scoria cones populating its flanks (Wright & Kyle, 1990a). Volcanism at Terror (3230 m) occurred from 2 to ~ 0.6 Ma (Kyle, 1990a), forming a mafic shield volcano (Wright & Kyle, 1990b). Numerous basanitic cones and phonolite domes are scattered about the flanks. Hut Point volcanism spanned ~ 1.5 – 0.3 Ma (Kyle, 1990a). Hut Point is a 20 km long, 2–4 km wide line of mostly basanitic scoria cones and lavas with a 228 m tall endogenous phonolite dome near its southern terminus (Kyle, 1990b). Erebus, the only active volcano on Ross Island, has had three phases of construction (Esser *et al.*, 2004). Mafic eruptions from 1.3 to 1.0 Ma built the proto-Erebus shield. A cone building phase, distinguished by eruptions of evolved phonotephrites and tephriphonolites, followed from ~ 1.0 to 0.25 Ma. Most of the modern edifice formed since 250 ka with the eruption of tephriphonolite to phonolite lava flows and minor pyroclastic material. Englacial tephra deposits in the East Antarctic ice sheet provide evidence of at least two large, possibly Plinian, eruptions at Erebus (Harpel *et al.*, 2008; Iverson *et al.*, 2014). No large-scale explosive eruptions have been attributed to Hut Point, Terror, or Bird.

SAMPLES

The studied suite includes nine basanite samples: four rapidly quenched marine hyaloclastites from Hut Point, four tephtras from Terror, and one tephtra from Bird (Fig. 1). The Hut Point hyaloclastite samples were cored during the Dry Valley Drilling Project (DVDP) in 1973. We examined MI samples from 268, 295, 359, and 376 m depths of DVDP hole 3, all part of a 214 m thick basanitic hyaloclastite consisting of palagonitic tuffs and breccias with interspersed pillow fragments (Kyle, 1981a). Olivine was sampled from the palagonitic matrix. To characterize the host magmas from which the MIs formed, pillow fragments were sampled nearest the depths from which the MI samples came (267–38, 306–33, 359–02, and 376–85 m). Mafic lapilli tuffs and tuff breccias were collected from scoria cones on Terror (14 T, 41 T, 46 T, 59 T) and Bird (98 T). Loose olivine was picked from the ash size fraction. *In situ* bombs and blocks from agglutinates and lava flow samples were collected near the sampled tephtra deposits. These samples, along with fresh lapilli clasts from the tephtras, were used to examine the host magma. Further descriptions of the samples can be found in [Supplementary Data Electronic Appendix 1](#) (supplementary data are available for downloading at <http://www.petrology.oxfordjournals.org>).

ANALYTICAL METHODS

Analytical methods summarized here are detailed in [Supplementary Data Electronic Appendix 2](#).

Major and trace element analyses of bulk samples

A total of 37 samples of fresh lapilli clasts from tephtra, pillow fragments in hyaloclastites and lavas or agglutinates were crushed in a quartz swing mill. They were analyzed for bulk-rock major element and Sr concentrations at Michigan State University using a Bruker S4 PIONEER wavelength-dispersive X-ray fluorescence (XRF) spectrometer; trace element contents were measured by inductively coupled plasma mass spectrometry (ICP-MS) on a Thermo Scientific ICAP Q quadrupole ICP-MS system connected to a Photon Machines G2 193 nm excimer laser ablation system. Analytical methods followed those of Rooney *et al.* (2012). Replicate analyses of BHVO-1, used as a check standard, have precisions (1σ) of $\leq 0.4\%$ for all elements except P_2O_5 , for which precision was 3.6%. For trace element analyses, JB-1a and BHVO standards were measured to assess analytical precision. Reproducibility was $<1\%$ for Sc, Ni, Rb, Zr, Ba, La, and Ce; $<3\%$ for V, Cr, Co, Y, Nb, Cs, Pr, Eu, Gd, Tb, Ho, Er, Yb, Lu, Ta, Pb, Th, and U; and $<4\%$ for Nd, Sm, Dy, Tm, and Hf.

Mineral and glass analyses

Mineral and glass analyses were obtained from 90 doubly polished olivine-hosted MIs, 29 matrix glasses, and 268 minerals (olivine, pyroxene, kaersutite, spinel, feldspar, apatite, leucite).

Fourier transform infrared spectroscopy

FTIR spectroscopy was performed on 82 MIs at the University of Oregon using a Thermo-Nicolet Nexus 670 FTIR spectrometer interfaced with a Continuum IR microscope. Quantitative measurements of H_2O and CO_2 (as CO_3^{2-}) were obtained by measuring absorbance spectra with 120–150 scans between 4000 and 1000 cm^{-1} . In most cases, three replicate spectra were collected on each MI. Vapor bubbles, when present, were avoided using the masking aperture in the FTIR system. A single absorbance spectrum of each host olivine was collected and compared with the MI spectra to check for incomplete intersection of the MI. For six MIs (41 T-21, 59 T-9, 59 T-29, 59 T-31, 359-8, 359-10) that were incompletely intersected, the maximum peak height of second-order Si–O overtones occurring between 1600 and 2100 cm^{-1} , which corresponds to olivine in the FTIR spectra, was measured on both host and MI spectra, and a ratio of the two was used to correct (by reducing) the thickness of each inclusion used in the Beer–Lambert calculation. We obtained three reflectance spectra of the host to calculate wafer thickness, following the method of Wysoczanski & Tani (2006). Total H_2O and CO_2 concentrations were calculated using the Beer–Lambert law with a value of 63 L

$\text{mol}^{-1}\text{cm}^{-1}$ for ϵ_{3520} (total H_2O) (Dixon *et al.*, 1997) and the compositionally dependent model of Mandeville *et al.* (2002) for ϵ_{1430} and ϵ_{1515} (carbonate). Analyzed MIs were 13–69 μm thick. Measured H_2O and CO_2 concentrations have precisions (1σ) of $<10\%$ and $<12\%$, respectively, which include uncertainties in glass thickness, density, and absorbance.

Electron microprobe analysis

All 90 MIs and their host olivines were analyzed for their major element and S, Cl, and F compositions using a CAMECA SX-100 electron microprobe in a single session at the CAMCOR facility, University of Oregon. Generally, three replicate analyses were performed on each MI and each olivine crystal. Spot analyses of MIs did not overlap and were performed near their centers. Olivine analyses were conducted within no more than a few tens of micrometers of the MI but no less than 5–10 μm .

Glasses were analyzed using a 10 μm beam diameter, 15 kV accelerating voltage and 10 nA for K (80 s), Al (80 s), Na (80 s), Si (80 s), Mg (80 s), Ca (80 s), S (80 s), and Fe (150 s), and 50 nA current for Mn (90 s), S (80 s), Cl (120 s), Ti (100 s), P (160 s), and F (160 s). Si, Fe, Mg, and Ca in olivine were analyzed with a 10 μm diameter beam, 15 kV accelerating voltage, 30 nA current and a count time of 80 s for all elements. Synthetic and natural minerals, oxides, and glasses were used as calibration standards and were analyzed twice to monitor instrument drift over time. Data were reduced using Probe for EPMA 10.0.7 software. Si, Al, Na, and K were corrected for the effects of alkali migration (Humphreys *et al.*, 2006) by extrapolating intensity–time plots to time zero. A fixed value of 1 wt % H_2O was used for the matrix correction of all MIs. Replicate glass analyses of unknowns have averaged precisions (1σ) of $\leq 3.5\%$ for SiO_2 , TiO_2 , Al_2O_3 , FeO, MgO, CaO, Na_2O , K_2O , P_2O_5 , S, and Cl, and $\leq 9.5\%$ for MnO and F.

Matrix glass and non-host minerals were analyzed and imaged on a Cameca SX-100 at New Mexico Tech. The analytical conditions and precision are provided in Supplementary Data Electronic Appendix 2.

Laser ablation ICP-MS of MIs

Trace element abundances were measured in 81 MIs using a Photon Machines G2 short pulse length 193 nm ArF Excimer laser coupled to a Thermo Xseries2 quadrupole ICP-MS system at the W. M. Keck Collaboratory for Plasma Spectrometry, Oregon State University (see Kent *et al.*, 2004). Helium was used as the carrier gas. A beam diameter of 40 μm or (rarely) 30 μm was used with a pulse frequency of 7 Hz and shot count of 225. Special care was taken to avoid analyzing crystal inclusions in MIs. The USGS GSE-1 G glass was measured as a calibration standard and USGS glass standards BHVO-2 G and BCR-2 G were measured to estimate accuracy and precision. Elemental concentrations were determined using Ca as an internal standard with abundances being

those obtained from electron microprobe analyses (corrected for post-entrapment processes, as described below). Precisions (1σ), which include uncertainty in calibration and internal standard measurements, are $\leq 5\%$ for Sc, Rb, Sr, Ba, La, Ce, Nd, and Sm; $\leq 10\%$ for V, Zn, Y, Zr, Gd, Dy, Yb, Hf, Pb, and Th; $\leq 16\%$ for Li, Ni, Cu, Nb, Er, and U; and $\leq 23\%$ for Cr and Cs.

CORRECTION OF MI DATA FOR POST-ENTRAPMENT PROCESSES

Post-entrapment crystallization and diffusive Fe–Mg exchange

Post-entrapment crystallization (PEC) of olivine along the MI–host interface is common in natural MI suites, but the effects of this process are often difficult to discern optically (Kent, 2008). PEC has the effect of depleting elements compatible in olivine and enriching those that are incompatible. A secondary result of PEC is disequilibrium between the host, PEC rind, and MI, which can result in diffusive Fe–Mg exchange between the MI and host (Danyushevsky *et al.*, 2000; Gaetani & Watson, 2000). Although the effects of PEC are most pronounced near the melt–olivine interface, in many cases the center may be affected as well (Newcombe *et al.*, 2014), so positioning analyses near the center of the MI reduces, but often fails to eliminate, the effects of PEC.

The effects of PEC were examined by comparing the measured host Fo with a predicted equilibrium olivine Fo content, calculated using the measured MI composition and appropriate values for Fe redox state and Fe^{2+} –Mg partitioning (K_D). The ferric–ferrous ratio was estimated using the model of Kress & Carmichael (1991) and assuming oxygen fugacity on the quartz–fayalite–magnetite (QFM) buffer, which is appropriate for magmas from Erebus (Kyle, 1977; Kyle *et al.*, 1992) and in the range for global mid-ocean ridge basalt (MORB) (Cottrell & Kelley, 2011) and ocean island basalt (OIB) (Mallmann *et al.*, 2011). Using the olivine–melt model proposed by Danyushevsky (2001), calculated K_D values are 0.29 ± 0.01 . In most cases, MIs examined in the present study are in equilibrium with a lower Fo olivine than the host, indicating that PEC has occurred, which is reflected in lower than expected concentrations of MgO (Fig. 2a). Diffusive Fe^{2+} exchange was assessed by comparing MI and bulk-rock FeO_t values. At the same extent of differentiation, FeO_t concentrations of the MIs are lower than bulk-rock values (Fig. 2b), which is consistent with diffusive loss of Fe^{2+} from the MIs.

MI major and volatile element compositions were corrected for the effects of PEC and diffusive Fe–Mg exchange using the Petrolog3 software (Danyushevsky & Plechov, 2011). The FeO_t contents of MIs were assumed to fall on a liquid line of descent defined by a polynomial regression of the bulk-rock data (Fig. 2b). This type of Fe correction is an oversimplification because it relies on the assumption that the bulk-rock composition is indicative of the melt originally entrapped to form the MI.

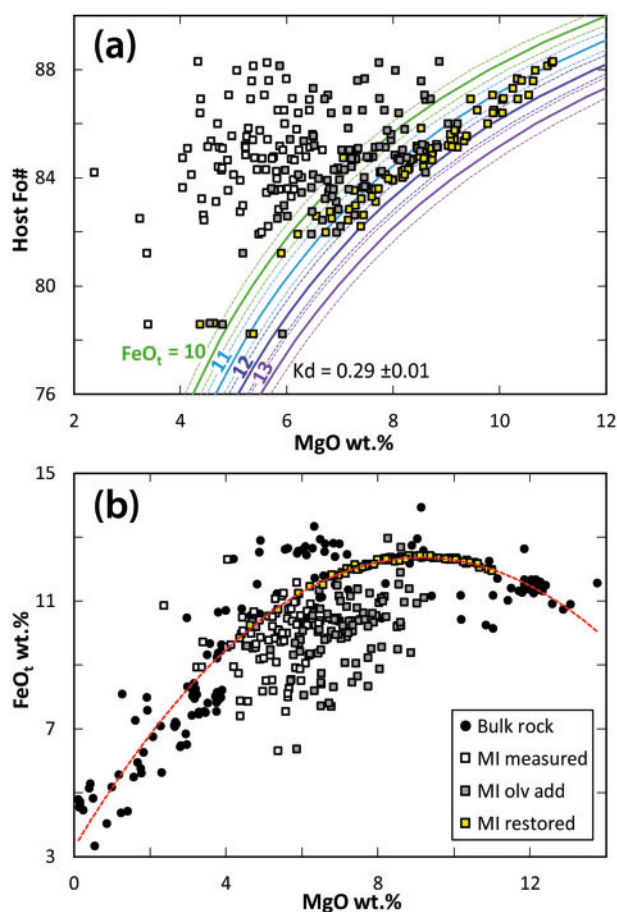


Fig. 2. Plots of olivine host composition (a) and bulk-rock and melt inclusion (MI) FeO_t content (b) vs MgO content for Ross Island, illustrating the effects of post-entrapment modification of MIs. Originally measured, olivine addition and restored compositions are all shown for MIs. (a) Comparison of host olivine compositions with MgO content of MIs, which indicates that most MIs have compositions that fall below the equilibrium line for appropriate values of FeO_t and K_d . (b) Total iron as FeO (FeO_t) vs MgO content for MIs and bulk-rock samples, showing that most uncorrected MI compositions and MI compositions with olivine addition (and no Fe correction) fall below the FeO_t of the liquid line of descent, consistent with diffusive loss of Fe^{2+} from MIs. The red dashed line is a polynomial regression of the bulk-rock data, which was used to infer the original FeO_t of the MIs. Data from this study (Supplementary Data Table EA3.12), Cole & Ewart (1968), Goldich *et al.* (1975), Kyle (1976, 1981b) and Stuckless *et al.* (1981). Compiled data are reported in Supplementary Data Table EA3.14.

However, the effect of the correction on most elements, apart from MgO and FeO_t , is relatively minor, and corrected MI compositions better coincide with bulk-rock and modeled evolutionary trends (Figs 2–4).

In the present study, PEC corrections require olivine additions of up to 21.9 wt % (average 11.8 wt %) and the difference between measured FeO_t and the final, corrected FeO_t is as much as 5.6 wt % (average 2.4 wt %). Subtraction of olivine is required for one MI (98T-4), which needed 0.4 wt % olivine subtracted. There were two MIs (14T-1 and 98T-4) that require the removal of FeO_t , meaning the PEC corrected MI compositions fell above the bulk-rock value of FeO_t . These corrections are

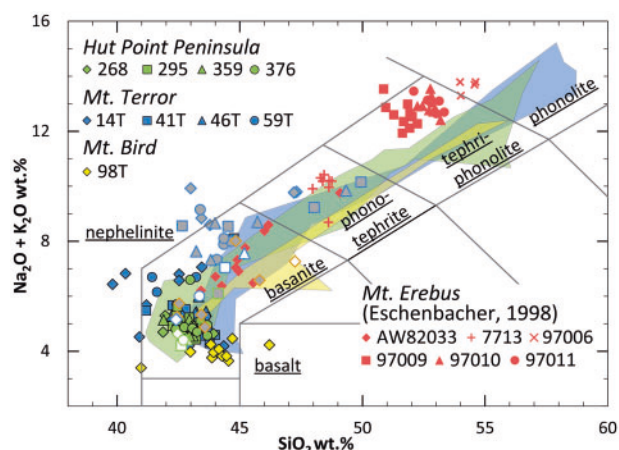


Fig. 3. Total alkalis-silica diagram for melt inclusions (MIs) from Ross Island, matrix glasses (gray fill), and bulk-rocks associated with MI samples (white fill). All compositions are normalized to 100% and volatile-free. Bulk-rock arrays from Bird, Terror, and Hut Point are overlapping and MI compositions occupy the least evolved end of the array. Erebus MIs are from Eschenbacher (1998). Fields are whole-rock analyses from Hut Point (green), Terror (blue), and Bird (yellow), which include the same data as in Fig. 2.

minor, requiring subtractions of 0.4 and 0.3 wt %, respectively.

The MI compositions reported here have been corrected for PEC and Fe–Mg exchange. MI compositions from Eschenbacher (1998) were also corrected for the effects of PEC and Fe–Mg exchange using the same methods as described above. Uncorrected analytical results of MIs from this study are given in Supplementary Data Electronic Appendix 3 Table EA3.1, and restored compositions of MIs from this study and Eschenbacher (1998) are given in Electronic Appendix 3 Table EA3.2 and EA3.3, respectively.

Vapor bubble formation

Vapor bubbles are common in MIs (Lowenstern, 1995). They form through a combination of PEC, which results in a net volume decrease, and melt contraction caused by cooling, which is due to the greater thermal expansion of the melt compared with olivine (Roedder, 1979; Lowenstern, 1995; Wallace *et al.*, 2015). These effects cause a pressure drop inside MIs that results in nucleation and growth of a vapor bubble. Diffusive loss of H^+ is also likely to play a role in vapor bubble formation and growth (Bucholz *et al.*, 2013). Decompression of the external melt has been shown to have relatively minor effects on the internal pressure of the MI (Tait, 1992). In most cases when vapor bubble growth occurs, the largest effect on the melt is decreased dissolved CO_2 concentration (Steele-MacInnis *et al.*, 2011; Moore *et al.*, 2015), owing to its low solubility relative to other major volatile species (H_2O , S, Cl, F).

All but eight MIs contain vapor bubbles. The number (0–4) and size (9–59 μm diameter, 21 μm average) of the bubbles is variable. The vapor bubbles occupy 1–22 vol. % of the MI (Supplementary Data Table EA3.4);

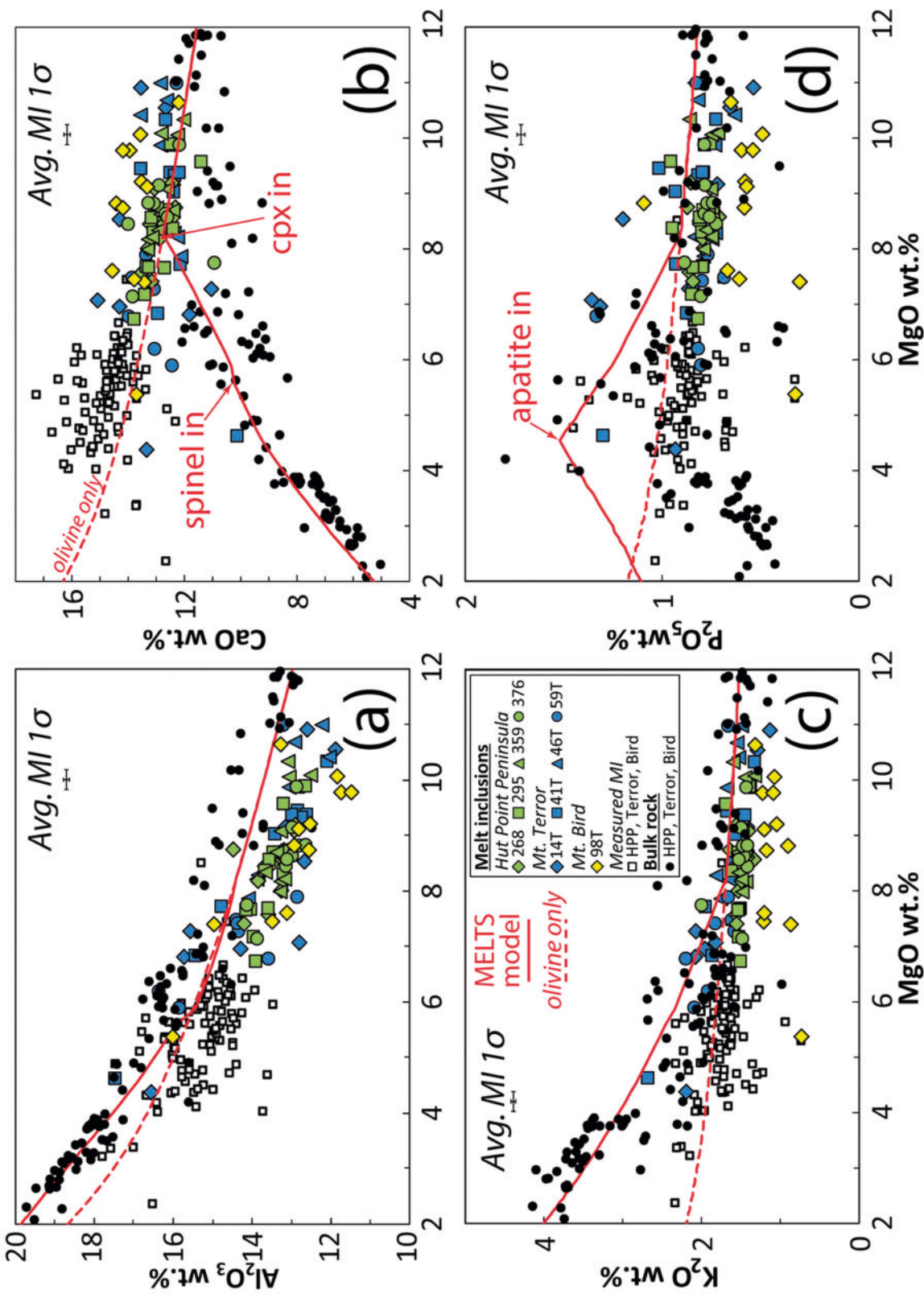


Fig. 4. Variations of Al_2O_3 , CaO, K_2O , and P_2O_5 abundance vs MgO content for melt inclusions (MIs) and bulk-rock samples from Ross Island. MI compositions are largely consistent with olivine-only fractionation. MIs have slightly high values of CaO and lower values of Al_2O_3 relative to measured bulk-rock compositions. Lines are modeled fractional crystallization paths obtained using MELTS (Ghiorso & Sack, 1995), with DVP2-105.53 as the starting melt composition (Kyle, 1981b). The conditions of the calculation are given in Supplementary Data 2. The dashed path is olivine-only fractionation. Bulk-rock data are the same as in Fig. 2.

these volumes were estimated assuming ellipsoid and sphere geometries for MIs and vapor bubbles, respectively, with the third axis (not viewable in prepared MI wafers) assumed to be equal to the length of the shorter measured axis. Vapor bubbles that are $< \sim 3\text{--}5$ vol. % of the MI are likely to have formed by shrinkage (Moore *et al.*, 2015; Wallace *et al.*, 2015). Larger bubbles are probably due to the co-entrapment of an exsolved vapor phase with the MI (Wallace *et al.*, 2015; Aster *et al.*, 2016), which is consistent with the highly variable volume per cent of vapor bubbles. Variable extents of post-entrapment diffusive H^+ loss, inferred from $\text{H}_2\text{O}/\text{Ce}$ in relation to MI size and location (see section below), are also likely to have played a role in the wide size range of bubbles (Buchholz *et al.*, 2013).

The effects of vapor bubble growth have been evaluated by using the computational approach of Wallace *et al.* (2015), which assumes MI shrinkage to be due to the combined effects of cooling and crystallization. To perform the calculation, the pre-eruptive vapor bubble volume percentage was estimated by the method of Riker (2005), which is based on phase equilibria and the densities and coefficients of thermal expansion for tholeiitic melts and olivine. Temperature estimates for the pre-eruptive MIs were calculated using the measured MI compositions. Original trapping temperatures were determined using the PEC/Fe-loss corrected MI compositions. For both, we used equation (4) of Putirka *et al.* (2007). The measured volume of the MI was then used to determine the pre-eruptive volume of the vapor bubble. The moles of CO_2 in the bubble were calculated using the Redlich–Kwong equation of state with the bubble volume, modeled pressure, mole fraction of CO_2 in the vapor phase [calculated using the method of Newman & Lowenstern (2002)], and temperature. The vapor phase was calculated to be dominantly CO_2 (>85 mol % in nearly all cases). The effect of vapor bubble formation on H_2O is assumed to be negligible. Vapor bubbles were found to contain 3–55% (average 36%) of the total CO_2 . Uncertainties in the estimated initial CO_2 concentrations are based on the error contributed from both the FTIR measurements and vapor bubble addition, which is based on error introduced by the thermometry.

RESULTS

Rock textures and mineral assemblages

Rock textures described here are discussed further in [Supplementary Data Electronic Appendix 1](#). Mineral analyses are reported in [Electronic Appendix 3 Tables EA3.5–3.9](#).

All the Hut Point samples exhibit similar textures and mineral assemblages. Olivine ($\text{Fo}_{75\text{--}90}$) is euhedral and generally abundant (~ 10 vol. %). No other macrocryst phases are present. However, there is microlitic augite and plagioclase feldspar in the groundmass.

Samples from Terror and Bird have similar phenocryst assemblages and disequilibrium textures; rare

xenocrysts are present. Olivine (mostly $\text{Fo}_{72\text{--}90}$) is common (≤ 14 vol. %) and variably resorbed. There are rare grains of high-Fo olivine ($\geq \text{Fo}_{90}$) that are large (> 3 mm), highly fractured, and partially dissolved. These grains have anomalously low CaO concentrations (0.07–0.20 wt %), indicating that they are likely to be of higher pressure origin (e.g. Larsen & Pedersen, 2000). Kaersutite is less common (≤ 7 vol. %). Most grains are reversely zoned, with rims $\sim 20\text{--}270$ μm thick observed in backscattered electron (BSE) images; the cores are commonly rounded ([Supplementary Data Fig. EA5.1](#)), implying that magma mixing has occurred. Clinopyroxene, orthopyroxene, plagioclase feldspar and alkali feldspar are also present, showing varying degrees of zonation and dissolution textures. Orthopyroxene is likely to be xenocrystic because it is not an expected crystallizing phase in silica-undersaturated, alkalic rocks (Miyashiro, 1978).

Melt inclusions

Melt inclusion morphologies

MI and host olivine morphological data and photomicrographs are reported in [Supplementary Data Electronic Appendix 3 Table EA3.4](#) and [Electronic Appendix 4](#), respectively. The MIs are fully enclosed and naturally glassy. Diameters of MIs range from 26 to 131 μm (average 61 μm). Of the 90 analyzed MIs, 35 contain crystal inclusions. In most cases, the crystals are of spinel and they are located at the MI–host interface. MIs with mineral inclusions are not outliers on compositional plots apart from a single MI from 376 (e.g. [Figs 3 and 4](#)). It is therefore unlikely that the mineral inclusions are daughter minerals crystallized from trapped melt. They are instead more likely to have been co-entrapped with melt as part of the process of MI formation (Roedder, 1979).

Olivine hosts of MIs

All MI-bearing olivines are euhedral and mostly free of matrix glass. Most have polyhedral morphology and contain spinel inclusions. During microprobe analysis, we observed no compositional zonation in BSE images. Olivines range in diameter from 0.4 to 1.9 mm (average 0.94 mm) and have overlapping compositions of $\text{Fo}_{78\text{--}88}$, with most between Fo_{82} and Fo_{88} ([Fig. 2a](#)). Olivine hosts have CaO concentrations of 0.22–0.38 wt % ([Supplementary Data Table EA3.10](#)), indicating that none belong to the high-Fo, low-CaO olivine population discussed above.

Major element compositions of MIs

When evaluating the compositions of MIs it is important to consider the possibility of boundary layer enrichment or depletion during trapping, which occurs when melt adjacent to a growing crystal becomes enriched in incompatible elements and depleted in compatible elements prior to entrapment (Faure & Schiano, 2005; Kent, 2008). This process is important when the crystal growth rate outpaces the diffusion of chemical species

into and out of the boundary layer melt. Boundary layer entrapment is unlikely to be important for our samples because we know of no natural datasets that show this process and MIs in hosts with slow growth morphologies were intentionally selected, although we do note that inferences of crystal growth rate based on morphology have recently been called into question (e.g. Welsch *et al.*, 2014). Additionally, slowly diffusing chemical species (e.g. P_2O_5) are not well correlated with melt inclusion size, which would occur during the entrapment of boundary layers because larger MIs are expected to be less affected by boundary layer processes.

Major element and volatile compositions of MIs, corrected for post-entrapment effects and normalized to 100%, are presented in [Supplementary Data Electronic Appendix 3 Table EA3.2](#), along with the modeling results. Matrix glass and bulk-rock analyses are given in [Electronic Appendix 3, Tables EA3.11 and EA3.12](#), respectively. Silica (39.0–45.4 wt %) and alkali ($Na_2O + K_2O$; 3.3–8.0 wt %) contents of MIs indicate that nearly all have basanite compositions (Fig. 3). MI compositions plot at the least evolved end of bulk-rock and matrix glass compositional arrays. The Hut Point MIs exhibit remarkably minor chemical variation despite spanning 108 m of the basanite hyaloclastite unit. Bulk tephra (and bulk-rock samples from the vicinity of MI-bearing tephra) have compositions that are relatively less (Hut Point samples, 14 T, and 59 T) or more (Terror, except 14 T and 59 T, and Bird samples) evolved than the associated MI compositions. The latter relationship is inconsistent with a simple model in which MIs formed from a parental melt, represented by the bulk-rock, which ascended in a closed system. It is much more likely that these samples were affected by open-system processes, including the addition of MI-bearing olivines to an evolved melt or mixing between more and less evolved magmas.

MI major element trends are broadly consistent with olivine crystallization and inconsistent with significant crystallization of the other observed macrocryst phases (e.g. clinopyroxene, kaersutite, feldspars, apatite; Fig. 4). Small yet distinct differences are exhibited in the behavior of major elements at each volcano. Olivine is the most abundant macrocryst phase in the Hut Point samples and MI compositions lie near the olivine-only fractional crystallization path, although bulk-rock trends appear to show clearly the effect of clinopyroxene fractionation (e.g. Fig. 4b). Terror MI compositions follow evolutionary trends similar to those from Hut Point, but the Terror MI compositions show greater scatter and some may reflect clinopyroxene fractionation (Fig. 4b–d). The Bird sample (98 T) has MIs with compositions that are also generally consistent with olivine-only fractionation, although there are two MIs (98 T-4 and 98 T-12) with low values of K_2O and P_2O_5 (Fig. 4c and d). These MIs also have trace element compositions that are anomalous, and they are interpreted to have a different origin.

When compared with the bulk-rock compositional array, MI compositions tend to plot at low values of Al_2O_3 and high values of CaO. Anomalous compositions

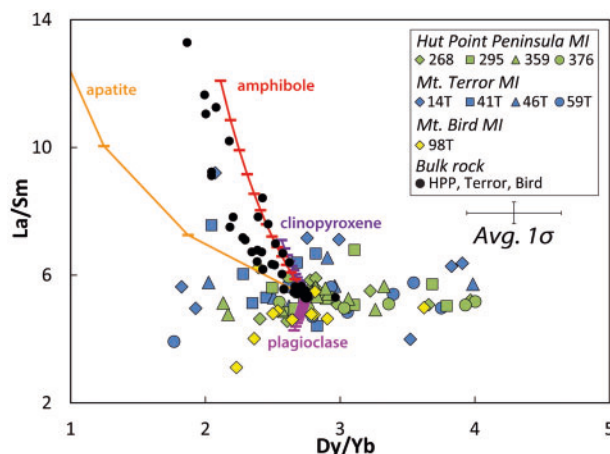


Fig. 5. La/Sm vs Dy/Yb for melt inclusions (MIs) and bulk-rock samples from Ross Island, showing that MI compositions have significant scatter in Dy/Yb. Each line shows a liquid line of descent that starts with the composition of DVDP2-105.53, a representative parental melt (Kyle, 1981b) undergoing fractional crystallization of the labeled phase. The ticks show 5 wt % crystallization increments. Mineral–melt partition coefficients used in the models are from McKenzie & O’Nions (1991).

such as these have been previously observed for MIs hosted in high-Fo olivines (e.g. Schiano *et al.*, 2000; Danyushevsky *et al.*, 2004). A thorough examination of the origin of high-Ca MIs is beyond the scope of this work, but there would appear to be two possible explanations. First, MIs may be related to a parental melt composition different from that from which the bulk-rock samples were derived. Second, the melts trapped as MIs may have been modified by grain-scale clinopyroxene assimilation (Danyushevsky *et al.*, 2004).

Trace element compositions of MIs

Trace element compositions of MIs are reported in [Supplementary Data Electronic Appendix 3, Table EA3.13](#). The MI trace element patterns are broadly consistent with those of the bulk-rock samples (i.e. chondrite-normalized rare earth element patterns are similar; Fig. EA5.2). Additionally, MI and bulk-rock compositions have similar ratios of trace elements with similar partitioning (Fig. EA5.3). Kyle (1981b) investigated samples from the DVDP cores and suggested that the compositions are consistent with descent from a single parent with a composition of DVDP2-105.53. The MIs show significant scatter in trace element concentrations, which is less apparent in the bulk-rock samples (Fig. 5). Some of the variation in MI trace element composition may be due to crystal fractionation (e.g. La/Sm; Fig. 5), but other variation (e.g. Dy/Yb) cannot be explained in this way. It should be noted that trace element ratios are unaffected by the corrections and normalizations that have been applied to the MI major and trace element compositions.

Volatile element compositions of MIs

Volatile concentrations in the MIs are highly variable, in the range of 0.4–2.0 wt % for H_2O , 0.09–0.24 wt % for S,

0.04–0.19 wt % for Cl, and 0.05–0.13 wt % for F (Fig. 6). CO₂ concentrations (0.16–0.86 wt % measured; 0.19–1.83 wt % after vapor bubble correction) are amongst the highest values reported for silicate MIs, although similar or greater values have been theorized (based on gas chemistry) to be present in undegassed magmas at other alkalic volcanoes (Allard *et al.*, 1997; Gerlach *et al.*, 2002; Burton *et al.*, 2007).

H₂O and CO₂ abundances are not positively correlated with K₂O content, indicating that the variations in these elements cannot be explained by simple incompatible behavior during fractional crystallization (Fig. 6a and b). This provides evidence that the melts were vapor-saturated and had already partially degassed at the time of trapping, and/or that variable extents of post-entrapment diffusive loss of H⁺ has occurred. In contrast, all MIs have S and Cl contents that generally correlate with K₂O abundance (Fig. 6c and d), which indicates that these volatiles behaved incompatibly during fractional crystallization and did not strongly partition into a vapor phase. At the lowest degree of differentiation, MIs from Hut Point, Terror, and Bird have slightly different concentrations of S and Cl (Fig. 6c and d). There is no correlation between melt H₂O content and trace element enrichment (e.g. Hartley *et al.*, 2015).

DISCUSSION

H₂O and CO₂ variations

A central goal of this study is to determine the behavior of H₂O and CO₂ in Ross Island magmas, which can be used to investigate magma storage, ascent, and differentiation. A first-order control on volatile behavior is degassing, an inevitable consequence of magma ascent owing to the strong dependence of volatile solubility on pressure (Kadik *et al.*, 1972). Equilibrium degassing is commonly assumed for most mafic magmas because relatively high diffusivities of volatiles (owing to high temperatures) promote vapor–melt equilibrium during degassing (Sparks *et al.*, 1994). During degassing, differences in the relative solubility of each volatile species result in fractionation, strongly affecting the CO₂/H₂O ratio of the melt and vapor phases (Dixon, 1997). Degassing models predict that an ascending magma will show a steep decline in the CO₂/H₂O ratio, owing to the low solubility of CO₂, until shallow depths where more significant H₂O degassing begins (Newman & Lowenstern, 2002; Papale *et al.*, 2006). These models are supported by the experiments of Lesne *et al.* (2011) and the volatile concentrations of MIs formed in some magmatic systems (e.g. Roggensack *et al.*, 1997; Liu *et al.*, 2006).

The H₂O and CO₂ concentrations of MIs from Hut Point, Terror and Bird are inconsistent with modeled equilibrium degassing paths (Fig. 7). The inconsistency exists within single samples and at the volcano scale. The trend of H₂O depletion (or CO₂ enrichment), relative to equilibrium degassing paths, has been recognized in many MI suites (e.g. Spilliaert *et al.*, 2006; Johnson

et al., 2008; Vigouroux *et al.*, 2008; Collins *et al.*, 2009; Roberge *et al.*, 2009; Blundy *et al.*, 2010; Witham, 2011; Oppenheimer *et al.*, 2011). In the following sections, several mechanisms are explored that can explain the trend, including diffusive loss of H⁺ from MIs (Roedder, 1979), disequilibrium degassing (Mangan & Sisson, 2000; Gonnermann & Manga, 2005; Pichavant *et al.*, 2013), CO₂ fluxing (Rust *et al.*, 2004; Spilliaert *et al.*, 2006; Johnson *et al.*, 2008; Vigouroux *et al.*, 2008; Collins *et al.*, 2009; Blundy *et al.*, 2010), and mixing with a degassed magma (Dixon *et al.*, 1991; Wallace & Anderson, 1998; Witham, 2011).

Volatile loss (or gain) by diffusion between MIs and carrier melt

The H₂O contents of sealed MIs can re-equilibrate with the external magma on relatively rapid timescales (Chen *et al.*, 2011; Gaetani *et al.*, 2012). Disequilibrium between a sealed MI and its carrier melt is caused by a change in the H₂O content of the carrier melt, most probably owing to degassing during shallow magma ascent or water loss during CO₂ fluxing. The extent of MI re-equilibration is largely a function of the time and length scales and the diffusivity of H⁺ in the olivine host (Qin *et al.*, 1992). Time, which is a function of magma ascent rate and the last depth at which the H₂O contents of a MI equilibrated, is a limiting factor, and H⁺ re-equilibration is often incomplete when MIs are quenched. Otherwise MIs would not record H₂O contents greater than atmospheric pressure equilibrated values (~0.1 wt %). A side effect of diffusive H⁺ loss from a MI is a decrease in internal pressure that leads to increased transfer of CO₂ into a vapor bubble (Bucholz *et al.*, 2013; Moore *et al.*, 2015; Aster *et al.*, 2016).

Previous research has demonstrated that there is a strong link between MI radius and the extent of H₂O re-equilibration (e.g. Qin *et al.*, 1992; Chen *et al.*, 2013; Lloyd *et al.*, 2013). The relationship is primarily due to the control of MI size on the surface area to volume ratio. Additionally, it has been shown that near-rim MIs re-equilibrate more rapidly (Ruprecht *et al.*, 2010). To investigate diffusive loss of H⁺, the relationship between the size and position of MIs and their H₂O content has been evaluated (Fig. 8). To eliminate the effects of variable amounts of fractional crystallization that may have occurred prior to MI trapping, the H₂O contents of the MI were normalized to their Ce contents; Ce was chosen because it exhibits partitioning like that of H₂O (Michael, 1995; Danyushevsky *et al.*, 2000). Hut Point MIs show a positive correlation between H₂O/Ce and MI radius (Fig. 8a), indicating significant diffusive loss of H⁺ from many smaller MIs. Additionally, those MIs that are closer to the edge of the host crystal tend to have reduced H₂O concentrations, consistent with diffusive loss of H⁺. The observed correlations are not especially well pronounced, but this is not surprising given that the extent to which diffusive re-equilibration occurs is a complex function of the shape, size, and position of the

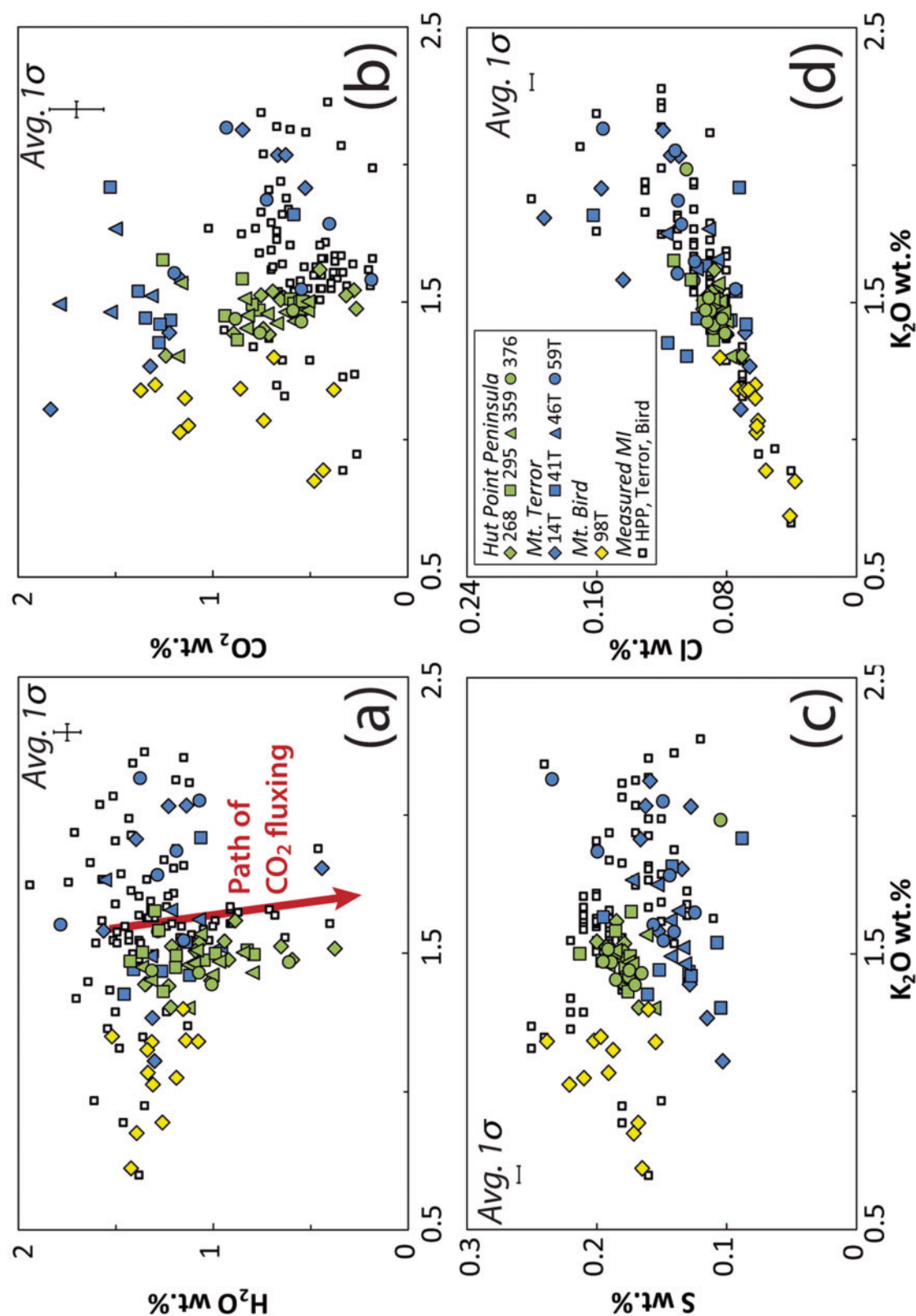


Fig. 6. Variation of K₂O and volatile contents in melt inclusions (MIs) from Ross Island. (a) The lack of a positive correlation between H₂O and K₂O indicates that simple incompatible behavior has not occurred. MI compositions are generally inconsistent with the path of CO₂ fluxing, which is calculated by assuming a loss of 1 wt % water results in a 38°C liquidus elevation (Medard & Grove, 2008) or 0.1 wt % increase in melt K₂O assuming olivine-only fractionation results (see text for details). (b) CO₂ compositions are plotted with vapor bubble addition (see text for details). The lack of correlation between CO₂ and K₂O abundances indicates that variation is not controlled by crystal fractionation. (c, d) Correlation with K₂O content suggests that variations in volatile abundances are caused by crystal fractionation.

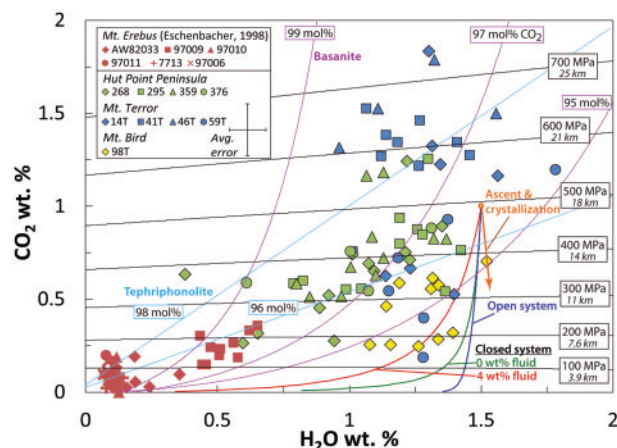


Fig. 7. H_2O vs CO_2 contents of melt inclusion (MIs) from Ross Island corrected for vapor bubble growth. Variation in MI composition is not consistent with equilibrium degassing paths (shown in orange). Erebus MI data are from Eschenbacher (1998). Isobars (black) and isopleths (purple) were calculated following the method of Iacono-Marziano *et al.* (2012), using an average of Hut Point, Terror, and Bird MI compositions. Degassing paths were modeled using VolatileCalc (Newman & Lowenstern, 2002) with the same averages, except for the path of ascent and crystallization, which is from fig. 29c of Blundy & Cashman (2008). Light blue isopleths are for more evolved compositions, which were calculated following Duan (2014), using the averaged composition of 97009 (tephriphonolite).

MI and the shape, size, and MI density of the host. In contrast to Hut Point samples, MIs from Terror and Bird lack evidence for significant diffusive loss of H^+ (Fig. 8a and b). There are a few MIs with small radii and anomalously high values of $\text{H}_2\text{O}/\text{Ce}$ (>150), which may have taken up H^+ by diffusion (Koleszar *et al.*, 2009; Hartley *et al.*, 2015). It is unlikely that much of the diffusive loss of H^+ occurred post-eruptively (i.e. Lloyd *et al.*, 2013). All the samples are expected to have quenched rapidly. Additionally, the samples that show the most pronounced effects (Hut Point samples) are hyaloclastites, which are likely to have cooled more rapidly than the samples of ash from Terror and Bird.

MI compositions converge on an $\text{H}_2\text{O}/\text{Ce}$ ratio of ~ 130 (Fig. 8a), which is considered to be the original ratio prior to diffusive water loss. This value is considerably lower than the $\text{H}_2\text{O}/\text{Ce}$ ratio of 200 that is associated with depleted MORB mantle (DMM) sources, indicating the involvement of a low $\text{H}_2\text{O}/\text{Ce}$, enriched component in the source (Dixon *et al.*, 2002). Further evidence for an enriched component in the source comes from isotopic and trace element studies that indicate mixing between HIMU and DMM sources at Ross Island (Sun & Hanson, 1975; Kyle *et al.*, 1992; Sims *et al.*, 2008).

Disequilibrium degassing

The rate at which individual volatile components move from melt to growing vapor bubbles during exsolution is controlled by the diffusivity of each component. Volatiles with low diffusivities, such as CO_2 (Watson, 1994), transfer from the melt to the vapor phase more

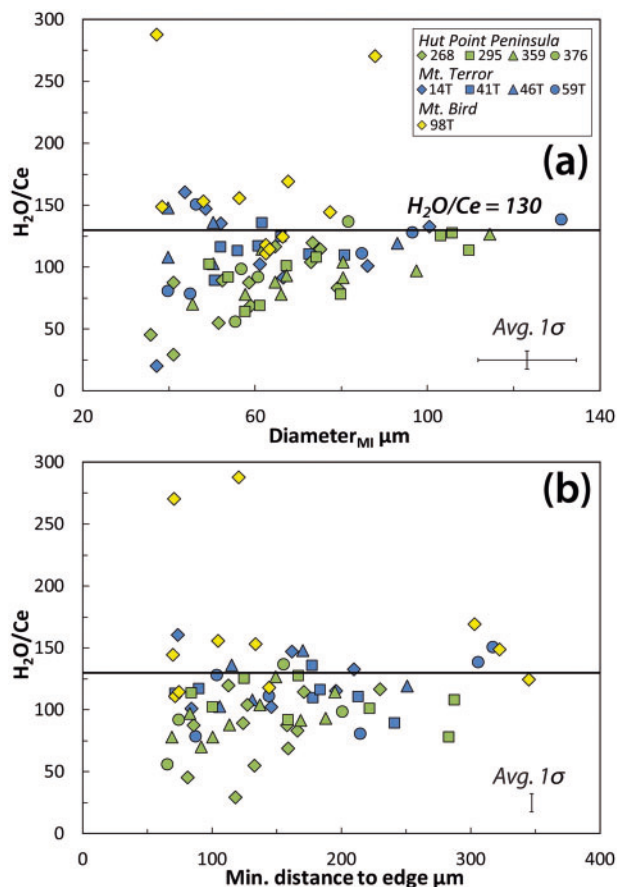


Fig. 8. Comparison of $\text{H}_2\text{O}/\text{Ce}$ with melt inclusion (MI) size and position for Ross Island samples; $\text{H}_2\text{O}/\text{Ce}$ converges on a value of ~ 130 at increasing values of MI diameter and minimum distance to edge of the host crystal. In Hut Point samples, diffusive loss of H^+ is indicated by the overall tendency for low values of $\text{H}_2\text{O}/\text{Ce}$ to be in smaller inclusions and those closer to an edge of the host. (a) The reported diameter is the average of the long and short axes of the intersected MI. (b) The minimum distance to edge is measured from the center of the MI to the nearest crystal face.

slowly and can become significantly super-saturated in a melt during disequilibrium degassing. This process is typically associated with mid-ocean ridge basalts (e.g. Sarda & Graham, 1990) and felsic magmas (e.g. Mangan & Sisson, 2000; Gonnermann & Manga, 2005). The experiments of Pichavant *et al.* (2013) show that other mafic magmas can also be susceptible. Experiments demonstrate that this process becomes important when vesicle number densities are low because the volatile exsolution is limited by the associated time and length scales of diffusion (Pichavant *et al.*, 2013).

One way to evaluate the likelihood of disequilibrium degassing is to consider bubble nucleation during exsolution. From experimental studies, there is no consensus about whether high alkali contents promote (e.g. Gardner, 2012) or inhibit (e.g. Bagdassarov *et al.*, 2000) bubble nucleation. Vapor bubbles occupy a variable percentage (1–22%) of the MI volume in the Ross Island samples (Supplementary Data Table EA3.4). The

significant variation, even in a single sample, of vapor bubble volume per cent indicates that a separate vapor phase was probably co-entrapped with some MIs, perhaps causing a defect at the crystal–liquid interface and resulting in MI formation (e.g. Roedder, 1979). If some of the vapor bubbles were in fact co-entrapped, it would follow that vesicle number densities were high in the carrying magma, which favors equilibrium degassing behavior. It is also important to note that the experiments of Pichavant *et al.* (2013) were largely crystal-free, and bubble nucleation was therefore homogeneous. A more likely case for a crystal-bearing natural system, such as the Ross Island one, is heterogeneous bubble nucleation. Heterogeneous bubble nucleation requires a lower degree of volatile super-saturation (Mangan & Sisson, 2000), and it follows that disequilibrium conditions are favored during homogeneous nucleation. Although the possibility of disequilibrium degassing cannot be completely ruled out, we note that high inferred vesicle number densities and the likelihood of heterogeneous bubble nucleation indicate that this process may not have been significant at Ross Island.

CO₂ fluxing

CO₂ fluxing (also called CO₂ flushing) is the most commonly invoked mechanism to explain non-equilibrium degassing behavior (e.g. Rust *et al.*, 2004; Spilliaert *et al.*, 2006; Johnson *et al.*, 2008; Vigouroux *et al.*, 2008; Collins *et al.*, 2009; Blundy *et al.*, 2010). CO₂ fluxing occurs when a CO₂-rich vapor emanating from depth percolates through magma. Exchange of H₂O and CO₂ between the vapor and melt results from chemical disequilibrium; dissolution of CO₂ is less pronounced owing to its slow diffusion rate (Yoshimura & Nakamura, 2010). H₂O diffuses about an order of magnitude faster (Watson, 1994), and significant transfer of water to the vapor can occur (Yoshimura & Nakamura, 2010). The fluxing vapor can be generated by conduit degassing (e.g. Rust *et al.*, 2004), degassing of deep melts (e.g. Spilliaert *et al.*, 2006; Johnson *et al.*, 2008; Blundy *et al.*, 2010), or assimilation of crustal carbonates (e.g. Marianelli *et al.*, 2005).

One way to test for CO₂ fluxing is to consider the expected effects. Modeled vapor isopleths predict the range of H₂O and CO₂ contents of a melt in equilibrium with a vapor of fixed composition. Therefore, the divergence of MI compositions from an equilibrium degassing path towards a vapor isopleth is expected if CO₂ fluxing has occurred. Vapor isopleths in Fig. 7 demonstrate that fluxing of a CO₂-rich vapor (97–99 mol % CO₂) could cause the observed H₂O and CO₂ trends in the Terror and Bird MIs. However, there are several lines of evidence that are inconsistent with a CO₂ fluxing interpretation. First, magma dehydration during CO₂ fluxing results in solidus elevation, which induces crystallization (Johnson *et al.*, 2008). A decrease in melt H₂O content of 1 wt % results in a 38 ± 5°C liquidus elevation of an olivine-saturated melt (Médard & Grove, 2008). This

degassing-induced undercooling has been modeled by simulating the isobaric cooling of a melt (with the composition of primitive MI, 59T-32) using Petrolog3 (Danyushevsky & Plechov, 2011) and applying the olivine–melt model of Danyushevsky (2001). The result for 1 wt % of water loss was ~3.7 wt % of olivine crystallization and a corresponding increase of melt K₂O content by 0.1 wt %. Thus, a negative correlation, albeit minor, would be expected between H₂O and K₂O contents if CO₂ fluxing had occurred. A negative correlation is not observed between melt H₂O content and extent of fractionation in most samples (Fig. 6a). Hut Point MI compositions show a similar trend. However, K₂O abundance increases with decreasing CO₂ content (Fig. 6b), which is not expected for CO₂ fluxing, and diffusive water loss has been identified in these MIs. Second, CO₂ fluxing is often considered to occur isobarically (e.g. Spilliaert *et al.*, 2006; Collins *et al.*, 2009), which is because the fluxing vapor must ascend rapidly relative to the melt to avoid dilution by water that partitions from the melt into the CO₂-rich vapor. If there is a constant supply of CO₂-rich vapor ascending rapidly relative to the magma, the dilution of the vapor phase (to a lower mole fraction of CO₂) will be less important. Isobaric CO₂ fluxing results in declining magmatic water concentration along a vapor saturation isobar and isobaric crystallization. The Ross Island MI data are not consistent with these processes. The composition of one sample (41T) has some attributes that can be explained by isobaric dehydration (Fig. 7), but the data for this sample are inconsistent with isobaric fractionation (Fig. 6a).

Despite the lack of evidence supporting CO₂ fluxing as a primary control on melt H₂O and CO₂ at Terror or Bird, it is still likely that this process has occurred to some extent. We have shown evidence of high magmatic CO₂ concentrations and early CO₂ saturation, and Eschenbacher (1998) demonstrated that the modern CO₂ emissions from Erebus volcano are high relative to predictions based on magma supply rate and the CO₂ contents of Hut Point MIs. It is likely that a deeply derived CO₂-rich vapor could have interacted chemically with hydrous magmas during ascent (Yoshimura & Nakamura, 2010). One way to explain the apparent lack of evidence for CO₂ fluxing is that melt dehydration may have been minor. Variable vapor bubble volume per cent provides evidence that exsolved vapor is likely to have been abundant at the time of MI entrapment. If the separate vapor phase contained H₂O, it would buffer magmatic volatile contents during fluxing, diminishing the dehydration effects (Yoshimura & Nakamura, 2010). Another possible explanation is that CO₂ fluxing may not have been important until after MI entrapment, in which case it would have contributed to diffusive H⁺ loss, particularly in small MIs.

Drain-back mixing

It has been shown that mixing of undegassed and degassed magma (drained back from shallow depths)

can result in a non-equilibrium-degassing trend (Witham, 2011), which is a process that occurs at Kilauea volcano, Hawaii (Dixon *et al.*, 1991; Wallace & Anderson, 1998). Shallow magmas that have cooled and degassed H₂O become negatively buoyant and sink. The descending magma mixes and re-equilibrates with the surrounding magma as it descends. H₂O diffuses more rapidly than CO₂ in the melt, which nudges the melt compositions away from the equilibrium degassing trends. At Ross Island, drain-back mixing is unlikely. MIs have consistently high concentrations of S (Fig. 6c), which is inconsistent with mixing with a shallowly degassed magma (Dixon *et al.*, 1991). Additionally, degassing of H₂O does not occur until depths that are much shallower than MI entrapment depths (Fig. 7). Thus, this process would require magmas to descend for tens of kilometers (Fig. 7).

Mixing of diverse magmas

Mixing of magma batches containing MI-bearing olivine could significantly affect the distribution of the H₂O and CO₂ contents of the MIs within a single eruptive unit. MIs that formed in different magmas and experienced different pre-eruptive histories would not necessarily follow a coherent degassing path. Variability in melt H₂O and CO₂ contents could arise in several different scenarios. Melts parental to the MI could travel along slightly different *P–T–t* paths, undergoing different crystallization histories. MIs may have resided at different depths in the crust for different lengths of time, CO₂ fluxing may have affected some melts but not others, and primary melts could be formed from slightly different extents of partial melting. These processes all have the potential to influence the behavior of H₂O and CO₂.

MI compositions from this study, especially those from Terror and Bird, do not form tight arrays on major, trace, or volatile element plots (Figs 3–6). The K₂O/TiO₂ ratio, which is not affected by PEC or diffusive exchange of Fe–Mg or H⁺, is useful for assessing the mixing of different olivine and MI populations (Fig. 9). Unlike in MI suites from Iceland (e.g. MacLennan, 2008), MI compositional heterogeneity in the Ross Island samples does not decrease with magmatic differentiation, suggesting that the variations are not caused primarily by concurrent mixing and crystallization. Rather, most MI entrapment appears to have occurred prior to mixing. Bird MIs appear to form a mixing array on some plots (e.g. Figs 4c, d and 9), which would be expected if mixing occurred during or before MI entrapment. However, as discussed above, the trend is mostly due to the influence of two MIs (98 T-4 and 98 T-12) with anomalously low Ce contents (e.g. Fig. 8).

The chemical variability observed in Ross Island MIs is common to MI suites from elsewhere (Kent, 2008). For Ross Island, the heterogeneity is at least partly inherited from variability in the parental melts, as indicated by the wide spread in Dy/Yb of the MIs, which cannot be due to fractionation alone (Fig. 5). Dy/Yb is

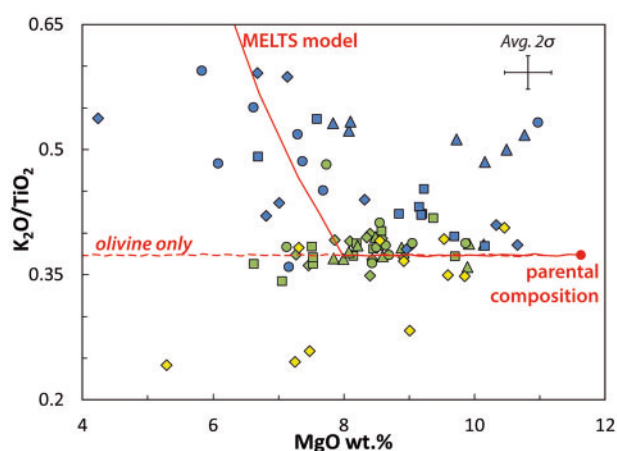


Fig. 9. K₂O/TiO₂ ratios vs MgO content for melt inclusions (MIs) from Ross Island. The trends indicate that MIs formed from melts of different composition. Lines show results of MELTS models, which are the same as in Fig. 4.

particularly sensitive to the presence of garnet in the residue during mantle melting. Variation could be related to the modal abundance of garnet in the source or to the extent of partial melting.

There are two possible scenarios to describe how MIs that formed in different melts could have later become incorporated into the same magma. First, some of the MI-bearing olivines could be cumulates that were re-entrained during the ascent of a later magma batch. This phenomenon has been suggested to occur at Kilauea volcano (Fodor & Moore, 1994). Support for re-entrainment of cumulates at Terror comes from the abundance of olivine-rich xenoliths, which are probably cumulates, in its eruptive products (Wright & Kyle, 1990b). Second, Terror magmas could have formed by batch assembly, meaning that MIs formed in different batches of magma that were later mixed and then erupted together as a single unit. Such a process is likely to happen beneath Ross Island, where small volumes of melt are produced during low degrees of partial melting (3–7%; Sun & Hanson, 1975).

Regardless of the source of the chemical variation, the implication of this finding is that complex histories, potentially involving the overprinting of several different processes that influence the behavior of H₂O and CO₂, can explain the lack of any evidence found for a single primary control on MI H₂O and CO₂. For Hut Point, the non-equilibrium degassing behavior can be attributed primarily to diffusive loss of H⁺.

Open-system processes

Additional evidence for open-system processes in samples from Terror and Bird comes from the observation that bulk-rock compositions are more evolved than those of the MIs (except 59 T), and matrix glass compositions are highly variable, although they typically lie within the bulk-rock compositional arrays (Fig. 3). All samples contain high-Fo olivine (Fo_{88–91}), which occurs with plagioclase, alkali feldspar and kaesutite. Most

kaersutite crystals are reversely zoned, with 30–270 μm thick rims. Kaersutite cores typically have rounded edges (Supplementary Data Fig. EA5.1), indicating that dissolution occurred at some stage followed by new growth episodes. As noted above, MI compositions for the most part do not lie along mixing arrays, an aspect that is interpreted to indicate that compositions have been controlled by processes occurring post-entrapment. It is suggested that the parental magma for the MIs either mixed with a more evolved, kaersutite-bearing magma or interacted with a crystal-rich mush.

Further complicating the pre-eruptive histories of the Terror and Bird samples is the occurrence of xenocrystic orthopyroxene, which is not a crystallizing phase in basanites, and high-Fo, low-Ca olivine. Both phases were likely to have derived from the mantle. If this is the case, the xenocrysts must have survived transportation through >25 km of crust (Finotello *et al.*, 2011), implying a relatively short period of crustal residence.

Magmatic plumbing system

Magma storage

A standard method to investigate crystallization depths (and infer storage depths) using MI compositions is to model vapor saturation pressures (e.g. Metrich & Clocchiatti, 1989), which requires the assumptions that the melts were vapor saturated at the time of trapping and that the internal pressure (or pressure of last equilibration) of the MI was maintained throughout ascent and eruption. For the Ross Island MIs, the first assumption is reasonable. CO_2 is the least soluble of the measured volatile species and does not behave as an incompatible element in melt evolution diagrams (e.g. Fig. 6b), which is robust evidence that the melt parental to the MI was vapor saturated during MI entrapment. The second assumption poses a difficulty. The effects of decompression of the external melt on the internal pressure of a melt inclusion have been shown to be generally minor (Tait, 1992) and the CO_2 contents have already been corrected to account for CO_2 loss to bubbles inside the MIs during cooling and PEC of olivine. However, diffusive loss of H^+ after trapping also causes an internal drop in pressure (Bucholz *et al.*, 2013), and this has not been accounted for in the present study. MIs from Hut Point, and possibly those from Terror and Bird to some extent, have undergone diffusive loss of H^+ . This phenomenon is explored in this section. It is stressed that pressure estimates should be viewed as minimum values. It is also noted that in using the eruptive products of monogenetic vents to make inferences about the plumbing system of each volcano, there is potential for bias in the record of information obtained.

To determine vapor saturation pressures, the model proposed by Iacono-Marziano *et al.* (2012) has been used because it is specifically calibrated for mafic compositions, including alkali basalts. MIs from Hut Point, Terror, and Bird record vapor saturation pressures between 160 and >560 MPa, which correspond to depths

of 6 to >19 km. This upper limit approaches the Moho depth for the region (19–27 km; Finotello *et al.*, 2011).

All four of the Hut Point samples belong to a single unit of hyaloclastite with remarkably minor chemical variation and only tenuous evidence of a single erosive break (Kyle, 1981a), which could indicate that the magmas were sourced from a stable, well-mixed reservoir. If the magmas parental to the Hut Point MIs were stored in a hypothetical reservoir, a locus of melt inclusion vapor saturation pressures would be expected. However, we have already argued that these MIs have undergone a variable degree of diffusive loss of H^+ , which lowered the internal pressure of the MIs. The initial H_2O contents of the MIs can be inferred by assuming an $\text{H}_2\text{O}/\text{Ce}$ ratio of 130, which the MI compositions appear to converge upon (Fig. 8). Using the difference between calculated H_2O and observed H_2O contents (termed $\Delta\text{H}_2\text{O}$), the extent of diffusive loss of H^+ can be approximated, assuming minimal water was lost (or gained) from the initial melts by different processes. Interestingly, $\Delta\text{H}_2\text{O}$ varies approximately linearly with vapor saturation pressure for MIs from Hut Point. This nearly linear drop in internal pressure with diffusive loss of H^+ has been observed elsewhere (Mironov *et al.*, 2015). It has been suggested that the internal pressure of MIs decreases during diffusive loss of H^+ , resulting in the additional transfer of CO_2 into each vapor bubble (Bucholz *et al.*, 2013). CO_2 in the bubble has already been allowed for by adding CO_2 back into the MI compositions, but this approach only accounts for the effects of cooling and crystallization of olivine. Thus, the pressure decrease that correlates with a decline in H_2O content must be due to additional vapor bubble growth caused by diffusive loss of H^+ . The correlation implies that the Hut Point melt inclusions formed at a similar depth. If the trend is extrapolated to $\Delta\text{H}_2\text{O} = 0$, a pressure of about 500 MPa, corresponding to a depth of ~18 km, can be inferred for MI entrapment and magma storage.

Terror and Bird magmas are unlikely to have resided in large, stable magma reservoirs. The compositions of MIs from these volcanoes are inconsistent with isobaric entrapment, which would be expected if the MIs reflected crystallization in a magma chamber. Additionally, there is no systematic relationship between $\Delta\text{H}_2\text{O}$ and entrapment pressure (Fig. 10). Furthermore, eruptive products from these volcanoes are commonly primitive in composition and contain abundant mantle xenoliths (Kyle, 1990b; Wright & Kyle, 1990a, 1990b), despite the fact that the magmas ascended through more than 20 km of crust (Finotello *et al.*, 2011). Interestingly, mantle xenoliths are abundant at Hut Point in surface samples, but they are absent from the unit studied in the DVDP core. It is possible that the DVDP samples track an earlier stage in the evolution of the volcanic plumbing system during which a more established reservoir existed. Later eruptive products may have bypassed the reservoir or it may have become inactive.

Evidence has been presented to show that Terror and Bird magmas mixed with a more evolved magma,

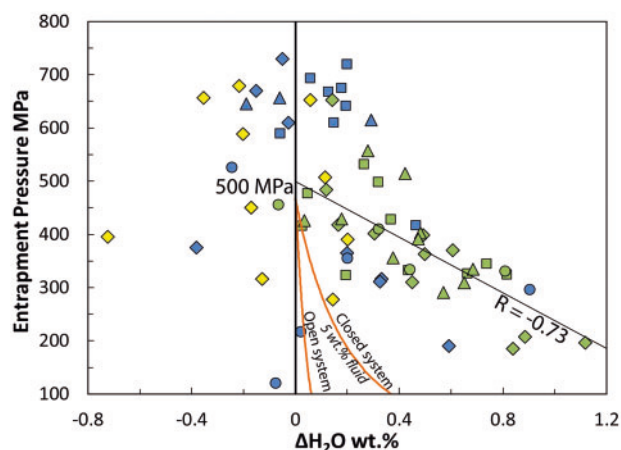


Fig. 10. Relationship between H_2O loss and modeled entrapment pressure for Ross Island melt inclusions (MIs), showing that MIs in which diffusive loss of H^+ has occurred have entrapment pressures that are low relative to MIs that have not lost H^+ . Only the samples with trace element data that have been corrected for vapor bubble formation are shown. $\Delta\text{H}_2\text{O}$ is the difference between calculated H_2O concentration (assuming $\text{H}_2\text{O}/\text{Ce}$ of 130) and observed H_2O concentration. Open- and closed-system degassing paths are the same as in Fig. 7. The linear regression is for all Hut Point samples that have a positive value of $\Delta\text{H}_2\text{O}$.

which is likely to have been stored at shallow depths. The presence of xenoliths in two samples (59T and 98T) and the lack of dissolution textures in orthopyroxene in all samples are evidence for short crustal residence times. Thus, the ascending mafic magmas would have had to interact only briefly with the more evolved magma prior to eruption. Volumetrically minor amounts of evolved basanite to phonolite are found at all three volcanoes (Kyle, 1990b; Wright & Kyle, 1990a, 1990b), indicating the potential existence of relatively small reservoirs where magma can stall and evolve in the cool upper crust.

Magma ascent

The ascent rate of Hut Point magmas can be quantified by considering the extent of diffusive re-equilibration of H^+ in MIs because this process is limited by the rate of diffusion of H^+ and occurs during magma ascent and post-eruptive slow cooling (Lloyd *et al.*, 2013). H^+ loss owing to slow cooling is assumed to be negligible because the Hut Point samples are hyaloclastites. The H^+ diffusion model of Chen *et al.* (2013), which is based on the solution of Qin *et al.* (1992), has been applied. Chen *et al.* (2013) invoked the coupled proton and metal vacancy diffusion mechanism (Demouchy & Mackwell, 2006), the slower of the two H^+ diffusion mechanisms in olivine. The metal vacancy diffusion mechanism is preferred here because it is supported by recent MI dehydration experiments (Chen *et al.*, 2011; Gaetani *et al.*, 2012; Bucholz *et al.*, 2013), and under reducing conditions, the faster of the mechanisms (proton-polaron; Mackwell & Kohlstedt, 1990) is likely to be less important. The Chen *et al.* (2013) model incorporates variable

boundary conditions for temperature and external melt H_2O concentration. For initial temperature, we used our estimates of entrapment temperature, and final temperature was set to the quench temperature (using the methods outlined above). For external water, three ascent scenarios have been modeled, using degassing paths calculated with VolatileCalc (Newman & Lowenstern, 2002), and these are shown in Fig. 7. It is assumed that all MIs were entrapped at 500 MPa or 18 km depth (Fig. 10), and that all MIs were carried in magmas that followed the same degassing path. The initial H_2O concentrations were calculated using an $\text{H}_2\text{O}/\text{Ce}$ ratio of 130, and an average value of Ce , which yielded 1.5 wt % H_2O . It should be noted that this is a simplification and a limitation of the model. However, it is inferred that magma compositions may have been homogenized in a reservoir beneath Hut Point, resetting MI H_2O contents to a single value.

Model results vary significantly between the three degassing scenarios, with the largest offset occurring between results for an open-system degassing model and the two closed-system degassing cases. Ascent rates vary over about three orders of magnitude ($\sim 5 \times 10^{-4}$ to $4 \times 10^{-1} \text{ m s}^{-1}$; Fig. 11a), and the time for magma ascent to occur ranges from ~ 1 day to 1 year (Fig. 11b). These estimates assume constant ascent rates from mid- to lower crustal depths (~ 18 km) to the surface. A more likely scenario is that magma ascends at an increasing rate as volatiles exsolve and add buoyancy to the melt, and at shallower depths, diffusive loss of H^+ becomes more important as the gradient between the MI H_2O contents and the external melt increases as water begins to degas. The most likely scenario is that magma initially ascends more slowly than our calculated rates, and as magmas ascend the velocity increases to rates greater than our predictions. Additionally, CO_2 fluxing may have affected the melt in the host magma after MI entrapment. A larger H^+ concentration gradient would develop at greater depth, requiring faster magma ascent. Thus, ascent rates should be taken as minimum values.

Few studies have constrained magma ascent rates from mid- to lower-crustal depths (e.g. Armienti *et al.*, 2012; Ruprecht & Plank, 2013). Despite the significant variability, the model results presented here put important constraints on the system. The minimum ascent rates of the model overlap with those obtained by Ruprecht & Plank (2013), which are mantle-to-surface ascent rates based on Fe–Mg interdiffusion in olivine. The results modeled here require that magmas ascended from depths of 18 km to the surface in less than a year. The high end of these modeled ascent rates overlaps with estimates for ascent from mid- to upper-crustal reservoirs (Lloyd *et al.*, 2013, 2014). Also within this range are 2–10 m s^{-1} ascent rates, based on xenolith settling velocities, obtained for alkalic magma by Rutherford & Gardner (1999), based on xenolith settling velocities. Our results underscore the importance of understanding the degassing path of the melt in the

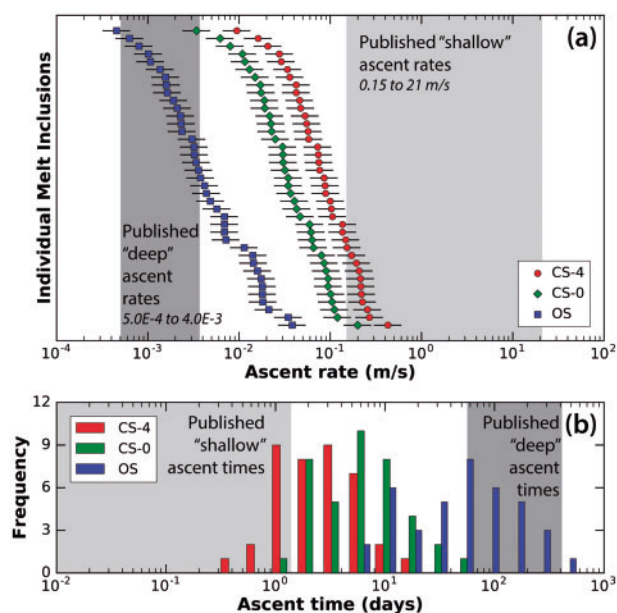


Fig. 11. Magma ascent rates and ascent times for three degassing scenarios (shown in Fig. 7) for Ross Island melt inclusions (MIs) and comparisons with other estimates. CS-4 is closed system, 4 wt % exsolved fluid. CS-0 is closed-system, 0 wt % exsolved fluid. OS is open system. (a) Results of diffusion modeling for individual MIs. Both cases of closed-system degassing yield similar results. Error bars reflect the uncertainty in temperature. Published 'deep' ascent rates give the ascent rate through the entire crust (Ruprecht & Plank, 2013). Published 'shallow' ascent rates are estimates for ascent from a crustal reservoir to the surface (Rutherford & Gardner, 1999; Lloyd *et al.*, 2013, 2014). (b) Histogram of magma ascent time from an 18 km deep reservoir. All but one of the estimates is consistent with magma ascending from a deep crustal reservoir in less than 1 year. Published 'deep' and 'shallow' ascent times are based on ascent rates referenced in (a) applied to ascent from 18 km.

host magma when modeling MI dehydration. A good approach to this problem is to examine large MIs, which are likely have lost less H^+ , and to obtain an empirical degassing path. For Ross Island, that was not possible because the studied MIs were entrapped at great depth and most MIs lost significant H^+ during ascent.

Ascent rates calculated for Hut Point should be taken as minimum values for Terror and Bird samples, which contain MIs with less conspicuous evidence of diffusive loss of H^+ . Greater abundance of microlites in the Hut Point samples (>80%) relative to Terror (<10–35%) and Bird (30–70%) samples corroborates this interpretation (Supplementary Data Fig. EA5.4). Because Hut Point samples are rapidly quenched marine hyaloclastites, microlite crystallization is likely to be pre-eruption in origin. Differences in the characteristics of the volcanic systems at Hut Point, Terror, and Bird could explain the differing ascent rates. Each edifice differs in size. Terror and Bird have large volumes (1700 and 470 km³, respectively) relative to Hut Point (100 km³). The load imposed by an edifice has been shown to influence the development of ascending dikes (Karlstrom *et al.*, 2009). Perhaps Hut Point has a less developed network of dikes, resulting in slower magma ascent, owing to its

relatively small size. An additional factor could be the location of the volcanic centers. Most of the recent extension in the Terror Rift has occurred to the north of Ross Island (Hall *et al.*, 2007). Terror and Bird are closer to the influence of this extension, which could promote dike formation and rapid magma transport through areas of weaker crust (Gudmundsson, 2006).

Comparison with Erebus

In this section, the H_2O and CO_2 contents of olivine-hosted MIs from Erebus previously studied by Eschenbacher (1998) and Oppenheimer *et al.* (2011) are re-examined and comparisons are made with results from the present study. The Erebus samples are from localities SE of the summit and have compositions and mineral assemblages consistent with the eruptive products of the volcano, which has led to the suggestion that they belong to the same differentiation series (Eschenbacher, 1998). Erebus whole-rock compositions define a single liquid line of descent, like those of the surrounding volcanoes (Fig. 3), that ranges from evolved basanite to phonolite. Using a least-squares mass-balance approach, Kyle *et al.* (1992) suggested that Erebus magmas have a similar parent to magmas erupted at Hut Point. However, differences in the mineralogy of the Erebus and Hut Point samples (importantly, the lack of kaersutite in Erebus rocks) indicate that the Erebus magmas undergo hotter, drier differentiation (Kyle *et al.*, 1992), which is supported by a recent phase equilibria study (Iacovino *et al.*, 2016).

H_2O and CO_2 concentrations of Erebus melt inclusions

Relative to other Ross Island volcanoes, Erebus samples have low concentrations of H_2O and CO_2 (~0.1 wt % H_2O and ~0–0.2 wt % CO_2 , except 97009, which has slightly elevated values of 0.5–0.7 wt % and 0.1–0.4 wt %, respectively; Fig. 7), compared with the surrounding volcanoes (0.4–2.0 wt % H_2O and 0.2–1.8 wt % CO_2 ; Fig. 7). The H_2O contents of Erebus MIs cannot be reconciled with a standard model of melt evolution (path of ascent and crystallization in Fig. 7).

Erebus MIs were entrapped at low pressure (most <150 MPa). Low H_2O (~0.1 wt %) and CO_2 (<0.2 wt %) concentrations in most MIs are inconsistent with equilibrium degassing of H_2O in a shallow magma chamber (Fig. 7). High emissions of CO_2 from Erebus led Eschenbacher (1998) to suggest a persistent flux of deeply derived CO_2 through the magmatic system. CO_2 fluxing could potentially result in H_2O and CO_2 trends of Erebus MIs (Fig. 7), which was previously suggested by Oppenheimer *et al.* (2011). To explain the low- H_2O , elevated- CO_2 MIs, the fluxing vapor would have to be extremely CO_2 -rich (>98 mol % CO_2) and vapor–melt equilibrium would need to follow tephriphonolite isopleths (Fig. 7). These isopleths differ in shape from those predicted for the same composition magma by other models (e.g. Papale *et al.*, 2006), which are similar in

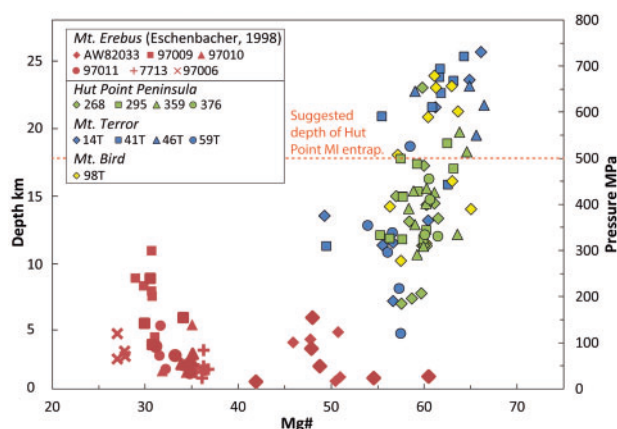


Fig. 12. Pressures (and depths) of formation vs Mg# [molar $\text{Mg}/(\text{Mg} + \text{Fe}^{2+})$] for Ross Island melt inclusions (MIs). Erebus data give consistently lower pressures than data for other volcanoes. All MIs have been corrected for PEC and vapor bubble growth (in the cases where a bubble was present). MIs from Erebus lack vapor bubbles (Eschenbacher, 1998). The dashed orange line indicates the inferred depth of formation of Hut Point MIs (see Fig. 10).

shape to the basanite isopleths (Fig. 7). Diffusive loss of H^+ is a possible explanation for the low H_2O contents of Erebus MIs. However, the MIs lack vapor bubbles, which would require resorption of bubbles formed during diffusive loss of H^+ during heating of the MIs after entrapment. Another possibility is that Erebus magmas are inherently drier than those of the surrounding volcanoes.

Erebus plumbing system

MI entrapment pressures provide evidence for extensive shallow crystallization at Erebus (Fig. 12), perhaps indicating the existence of a shallow storage region. MI compositions range from basanite to phonolite, and most (83%) have been entrapped at pressures of <150 MPa, corresponding to depths of <6 km (Fig. 12). The presence of a shallow reservoir is supported by radioactive equilibrium between $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{227}\text{Ac}/^{231}\text{Pa}$, which indicates long (>100 years) residence times for the Erebus phonolitic magmas (Sims *et al.*, 2013). Shallow storage was also proposed by Oppenheimer *et al.* (2011), who, however, suggested that most differentiation and dehydration occurs in a deep reservoir (>800 MPa). The wide range of chemical compositions observed at shallow depth indicates that either differentiation is shallow or that some melts bypass a hypothetical deep region of differentiation. The later scenario requires that for differentiated melts, the deep crystallization is not recorded in MIs.

CONCLUSIONS

H_2O (0.4–2.0 wt %) and CO_2 (0.2–1.8 wt %) variations in MIs from Hut Point, Terror, and Bird are inconsistent with modeled equilibrium degassing paths. MIs from Hut Point underwent post-entrapment diffusive re-equilibration of H^+ . Diffusion model estimates of magma ascent are interpreted to indicate that Hut Point

magmas ascended from depths of ~ 18 km in less than a year, but the results strongly depend on the degassing path of the melt in the host magma. MIs from Terror and Bird lack a strong indication of diffusive loss of H^+ , indicating that the magmas ascended more rapidly. The inferred differences in ascent rate are reflected in the abundance of microlites in the samples. The H_2O and CO_2 contents in Terror and Bird MIs require more complex pre-eruptive histories, which probably have been influenced by magma mixing. For all samples, there is no direct evidence of CO_2 fluxing.

MIs were entrapped over a wide range of pressures (160 to >560 MPa). Adjusting for the effects of diffusive loss of H^+ and vapor bubble growth, the Hut Point MIs are likely to have been entrapped at ~ 18 km depth. The Terror and Bird MI compositions reflect polybaric entrapment and contain xenocrystic material. The most likely scenario for these volcanoes is that magmas ascend relatively rapidly through a network of sills and dikes.

MIs from Erebus (Eschenbacher, 1998) have low H_2O and CO_2 concentrations (most ~ 0.1 wt % and ~ 0.2 wt %, respectively). Calculated MI vapor saturation pressures indicate significant shallow (<6 km) crystallization at Erebus. This shallow differentiation is not observed at the surrounding volcanoes.

MI studies that assess the H_2O and CO_2 contents of MIs must consider diffusive loss of H^+ . We have demonstrated the importance of evaluating MI morphology and textural indications of ascent rate in host tephra. We also underscore the importance of careful interpretation of MI data and working with a sufficiently large dataset, because the compositions of individual MIs, even within the products of a single eruption, sometimes are influenced by different pre-eruptive histories.

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SUPPLEMENTARY DATA

Supplementary data for this paper are available at *Journal of Petrology* online.

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