



Hydrogen and oxygen isotope behaviors during variable degrees of upper mantle melting: Example from the basaltic glasses from Macquarie Island

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ABSTRACT

We present measurements of hydrogen and oxygen isotopes in MORB glasses from Macquarie Island (SW Pacific Ocean) coupled with determination of bulk H₂O content by two independent techniques: total dehydration and FTIR. The incompatible trace elements in these glasses vary by a factor of 12 to 17, with K₂O varying from 0.1 to 1.7 wt.%; these ranges reflect a variable degree of closed-system mantle melting, estimated from 1 to 15%. Water concentrations determined by the two techniques match well, yielding a range from 0.25 to 1.49 wt.% which correlates positively with all of the measured incompatible trace elements, suggesting that water is un-degassed, and behaves conservatively during mantle melting. Also, the agreement between the FTIR-determined and extracted water contents gives us confidence that the measured isotopic values of hydrogen reflect that of the mantle. Comparison of the range of water content with that of other incompatible trace elements allows estimation of the water partition coefficient in lherzolite, 0.0208 (ranging from 0.017 to 0.023), and the water content in the source, 386 ppm (ranging from 370 to 440 ppm). We observe a fairly narrow range in δD and δ¹⁸O values of $-75.5 \pm 4.5\%$ and $5.50 \pm 0.05\%$ respectively, that can be explained by partial melting of normal lherzolitic mantle. The measured δD and δ¹⁸O values of Macquarie Island glasses that range from nepheline- to hypersthene-normative, and from MORB to EMORB in composition, are identical to those in average global MORB. The observed lack of variation of δD and δ¹⁸O with 1 to 15% degree of mantle melting is consistent with a bulk melting model of δD and δ¹⁸O fractionation, in which water is rapidly scavenged into the first partial melt. The narrow ranges of δD and δ¹⁸O in normal mantle are mostly due to the buffering effect of clino- and orthopyroxenes in the residual assemblage; additionally, fast “wet” diffusion of oxygen and hydrogen isotopes through the melting regions may further smooth isotopic differences.

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1. Introduction

Water is the key highly entropic component that influences the degree and depths of mantle melting (e.g., Asimow et al., 2004). While the main reservoir for water appears to be the hydrosphere, there is still discussion on how much water is present in the mantle. The latter depends strongly on the partition coefficient of water between nominally anhydrous peridotitic assemblages and basaltic melts. Experimental work has demonstrated the range of possible DH₂O at variable pressures (e.g. Hirschmann et al., 2009), suggesting that water may be present in the mantle in greater quantities than previously thought (ca more than 500 ppm). Investigation of MORB and OIB glasses for water concentration, and consideration of the

H₂O ratio to similarly incompatible trace elements (Danyushevsky et al., 2000; Dixon et al., 2002) suggest wide variations among mantle derived melts.

The hydrogen isotopic composition of the mantle serves the purpose of understanding the global water cycle. It is important for a variety of topics in modern geosciences: 1) the bulk Earth δD value and its origin, 2) the global balance of water and the amount of water in the mantle (e.g. Kelley et al., 2010), 3) the mechanisms of dehydration of the subducted slabs and evidence for past subduction (e.g. Kingsley et al., 2002), and 4) the explanation of the apparent disequilibrium between the δD of the upper mantle and the δD of sea water (e.g. Taylor, 1974). Based on known isotope fractionation factors, the δD value of -80% for MORB is too low for what would be required for high-T equilibrium with the 0‰ seawater, suggesting that the mantle and the hydrosphere may not be at steady state (Ito et al., 1983; Lecuyer et al., 1998; Shaw et al., 2008), or that there is a deeper mantle reservoir with isotopically-heavier water that has not yet been

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accounted for. Given that MORB represents 80% of the total magma output on earth (e.g. [McBirney, 2006](#)), understanding the D/H fractionation during MORB genesis is an important yet insufficiently explored topic.

The hydrogen isotope geochemistry of mantle rocks as revealed by analysis of submarine basaltic glasses hinges on recognition of primary values despite various degassing and alteration effects (e.g. [Craig and Lupton, 1976](#); [Poreda, 1985](#)). Prior work that relied on conventional extraction methods which required relatively large (0.5–1.0 g) quantities of water-poor glasses have ascribed more than 60% of the variations to the mantle sources parental to MORB (ca. –30 to –90‰). Similar arguments have been made concerning the large (ca. 2‰) isotopic range for oxygen isotopes in MORB measured on whole rocks by conventional methods, but subsequent work on phenocrysts, especially refractory olivine, has found a far narrower range for the mantle and has resulted in the recognition of small, sub-permil effects ([Eiler, 2001](#); [Bindeman, 2008](#)).

The development of continuous flow analytical methods in the past decade has allowed a significant reduction in the amount of material required for hydrogen isotopic measurements (from multi micromolar to a few hundred nanomolar quantities, [Sharp et al., 2001](#); [Eiler and Kitchen, 2001](#); [O'Leary et al., 2007](#)) without losing much precision (typically 1–3‰). The simultaneous development of secondary ionization mass spectrometry (SIMS) techniques ([Hauri et al., 2006](#); [Shaw et al., 2008](#)) has permitted in situ analysis of glasses and melt inclusions, although questions remain concerning the degree to which their H₂O contents reflect that of the original magma. The SIMS methods rely on developing an empirically calibrated set of glass standards to account for matrix effects on glasses of variable SiO₂, H₂O and other oxide content, which is sometimes a daunting task (e.g., [Hauri, 2002](#); [Hauri et al., 2006](#)). In the case of continuous flow measurements it is important to establish that there is complete extraction of water and no isotope fractionation.

Reduction in sample size by 2–3 orders of magnitude (e.g. from 0.5–1 g to 0.5–3 mg) has tremendously simplified the procedure of hydrous mineral and glass separation, permitting selection of the least altered glasses or minerals from more altered specimens based on microscopic and Fourier transform infrared spectroscopy (FTIR) investigations. As post-magmatic alteration of water-poor glasses typically results in water uptake, it is possible to investigate total water, and H₂O/(OH) glass speciations in glass and their relation to the quenched magmatic values (e.g., [Conde et al., 2009](#)) and to distinguish primary vs. post-magmatic alteration processes ([Friedman et al., 1993](#); [Anovitz et al., 2008](#)). The secondary alteration uptake typically results in the increasing concentration of molecular water, which is absorbed below the glass transition temperature, and it is possible to recognize this water as a separate peak on FTIR spectra.

In this work we performed careful investigation of oxygen and hydrogen isotopes and water contents in a set of well-characterized glasses by relying on the FTIR-selected freshest material. The glasses that we selected range in water content from 0.25 to 1.49 wt.% and are from Macquarie Island, S. Pacific Ocean. Their major element composition ranges from strongly Ne-normative to tholeiitic, hypersthene-normative, spanning from MORB to EMORB in trace elemental signatures, and exemplifying different degrees of mantle melting ([Table 1](#)).

2. Geology of Macquarie Island, petrology and geochemistry of its glasses

Macquarie Island ([Fig. 1](#)), located 1200 km southwest of New Zealand, is a tectonically-uplifted portion of the mid-ocean rift, on the boundary between the Pacific, Australian, and Indian plates ([Daczko et al., 2003a, 2003b](#)). Currently, it is part of the active Australian–Pacific oceanic transform plate boundary between the Alpine Fault of New Zealand and the Australian–Pacific–Antarctic triple junction ([Fig. 1a](#)). Geologically it is composed of a tectonized but typical mid-ocean ridge assemblage of rocks of Miocene age, comprised primarily of pillow basalts, with a lesser proportion of massive gabbro, peridotite and basaltic dike complexes

([Fig. 1](#) and [Goscombe and Everard, 1998, 2001](#)). It is important to note, that deep-oceanic red pelagic mudstones are present among the basaltic rocks, and that the oceanic crust of Macquarie Island is interpreted to have formed at 2–3.5 km depth, in a slow-spreading rate environment ([Portner et al., 2010](#)).

Basaltic glasses considered in this work occur as quenched rims on pillow lavas and, more commonly, as fragments in hyaloclastite breccias ([Kamenetsky et al., 2000](#); [Varne et al., 2000](#) and references therein). The latter usually consist of sub-angular to angular or rounded glass lapilli, glassy and crystalline pillow fragments up to 3 cm in size (~40% of breccias), and may carry olivine and/or plagioclase crystals, in a matrix of smaller glassy debris cemented by a greenish-gray chlorite–smectite aggregate ([Fig. 2](#)) possibly after shattered and dispersed glass. In thin section the glasses are transparent and dark to light brown in color, and may contain scattered opaque zones rich in crystallites. Microphenocrysts of olivine (20–300 μm) with trapped spinel and melt inclusions are common ([Fig. 2](#)), but make up less than 3–5 vol.% of the rock. In some glass samples, Cr-spinel and plagioclase crystals are also present, although no clinopyroxene was observed.

Based on Mg numbers and other parameters, two groups of glasses have been recognized: primitive Group-I glasses represent melts related to each other by very low to moderate degrees of melting of a homogeneous spinel lherzolite mantle source, whereas Group-II glasses are related to the first group by small amounts of crystal fractionation ([Kamenetsky and Maas, 2002](#)). Samples that we selected for this investigation are primarily from Group-I, which display the maximum range of H₂O and K₂O contents and are, interpreted to represent variable degrees of fractional mantle melting ([Kamenetsky et al., 2000](#); [Varne et al., 2000](#); [Kamenetsky and Maas, 2002](#); [Kamenetsky and Eggins, 2012](#)).

The isotopic compositions of Sr and Nd of studied glasses plot firmly within normal MORB fields, and sample to sample variability in our dataset is very small ([Fig. 3](#)). It is possible, however, that there is more fertile, pyroxene rich, and/or less fertile, pyroxene-poorer, mantle under Macquarie Island, that has ingrown slightly variable amounts of radiogenic products. Partial melting of these areas may generate the correlation of radiogenic isotopes and La/Sm ratios presented in [Kamenetsky and Maas \(2002\)](#).

At the estimated water depths of 2–3.5 km at the time of basalt eruption, which correspond to 205–360 bar pressure, the basalt may contain 1.5–1.9 wt.% water (estimated using program VolatileCalc, [Newman and Lowenstern, 2002](#)), which is higher than our highest measured water concentration of 1.49 wt.%. Nonetheless, most glasses contain abundant spherical bubbles of low-density CO₂ (<5 to 100 μm). Despite this CO₂ degassing, the water concentrations in these gas bubbles are expected to be minimal along any degassing path on a CO₂–H₂O graph, in accordance with the solubility of these components in basaltic melts ([Newman and Lowenstern, 2002](#)). The undegassed nature of the studied glasses with respect to H₂O is also robustly demonstrated by the correlation of H₂O with all incompatible trace elements ([Fig. 4](#)), suggesting that water behaves conservatively as an incompatible trace element. Thus, these glasses represent rapidly quenched fractional or batch melts derived from the mantle that never achieved water saturation or experienced water loss; hence these glasses provide good insight into the D/H ratios of the mantle. Since these melts erupt unmixed, thereby preserving the distinct trace elemental signatures characteristic of different degrees of mantle melting, we suggest that there was not a single magma chamber.

The Group-I glasses that are considered in this work represent the most primitive and least fractionated melts, which are in Fe–Mg equilibrium with the olivine microphenocrysts that they contain ([Fig. B1](#) in [Appendix A](#)); the degree of olivine fractionation in these primitive glasses does not exceed 5–8% ([Kamenetsky et al., 2000](#)). Since such small degrees of fractionation will increase concentrations of nearly all incompatible trace elements and water indiscriminantly by this small amount, we chose not to adjust measured concentrations for fractionation.

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