

# CO<sub>2</sub> content beneath northern Iceland and the variability of mantle carbon

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## ABSTRACT

**Primitive basalt melt inclusions from Borgarhraun, northern Iceland, display large correlated variations in CO<sub>2</sub> and nonvolatile incompatible trace elements (ITEs) such as Nb, Th, Rb, and Ba. The average CO<sub>2</sub>/ITE ratios of the Borgarhraun melt inclusion population are precisely determined (e.g., CO<sub>2</sub>/Nb = 391 ± 16; 2σM [two standard errors of the mean], n = 161). These data, along with published data on five other populations of undegassed mid-oceanic ridge basalt (MORB) glasses and melt inclusions, demonstrate that upper mantle CO<sub>2</sub>/Ba and CO<sub>2</sub>/Rb are nearly homogeneous, while CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th are broadly correlated with long-term indices of mantle heterogeneity reflected in Nd isotopes (<sup>143</sup>Nd/<sup>144</sup>Nd) in five of the six regions of the upper mantle examined thus far. Our results suggest that heterogeneous carbon contents of the upper mantle are long-lived features, and that average carbon abundances of the mantle sources of Atlantic MORB are higher by a factor of two than those of Pacific MORB. This observation is correlated with a similar distinction in water contents and trace elements characteristic of subduction fluids (Ba, Rb). We suggest that the upper mantle beneath the younger Atlantic Ocean basin contains components of hydrated and carbonated subduction-modified mantle from prior episodes of Iapetus subduction that were entrained and mixed into the upper mantle during opening of the Atlantic Ocean basin.**

## INTRODUCTION

Magmas deliver carbon from Earth's upper mantle and release it to the atmosphere and oceans as CO<sub>2</sub>. The presence of carbon in the mantle can influence the depth of melting within the mantle (Dasgupta et al., 2013), and CO<sub>2</sub> bubbles in magmas can influence the style of explosive eruptions (Hekinian et al., 2000; Clague et al., 2009). The release of CO<sub>2</sub> into the atmosphere also affects long-term global climate and may provide a positive feedback mechanism to volcanism (Huybers and Langmuir, 2009) that may also influence the response of mid-ocean ridge magmatism to glaciation (Maclennan, 2002) and possibly sea-level changes (Burley and Katz, 2015; Tolstoy, 2015). However, the solubility of carbon in silicate melt decreases strongly with decreasing pressure (Dixon et al., 1995), and so most magmas arrive at Earth's surface having lost most of their carbon via degassing. To circumvent the effects of the degassing process, in this study we examine silicate melt inclusions, which are tiny samples of quenched magma (typically <200 μm diameter) trapped in crystals that grow in the magma prior to eruption. By virtue of being enclosed within their crystal hosts, melt inclusions are prevented from degassing their volatiles during volcanic eruptions, and can be used to study the carbon content of magmas at the time and depth at which the inclusions were trapped. Despite this advantage, nearly all melt inclusion studies that have determined carbon contents, with two exceptions (Saal et al., 2002; Le Voyer et al., 2016), find that the magmas represented by melt

inclusions have still lost carbon by degassing prior to entrapment (e.g., Moore et al., 2015). As a result, it has been very difficult to determine the original carbon content of magmas prior to degassing, and thus to use mantle-derived magmas to estimate the carbon content of Earth's interior.

## SAMPLING AND METHODS

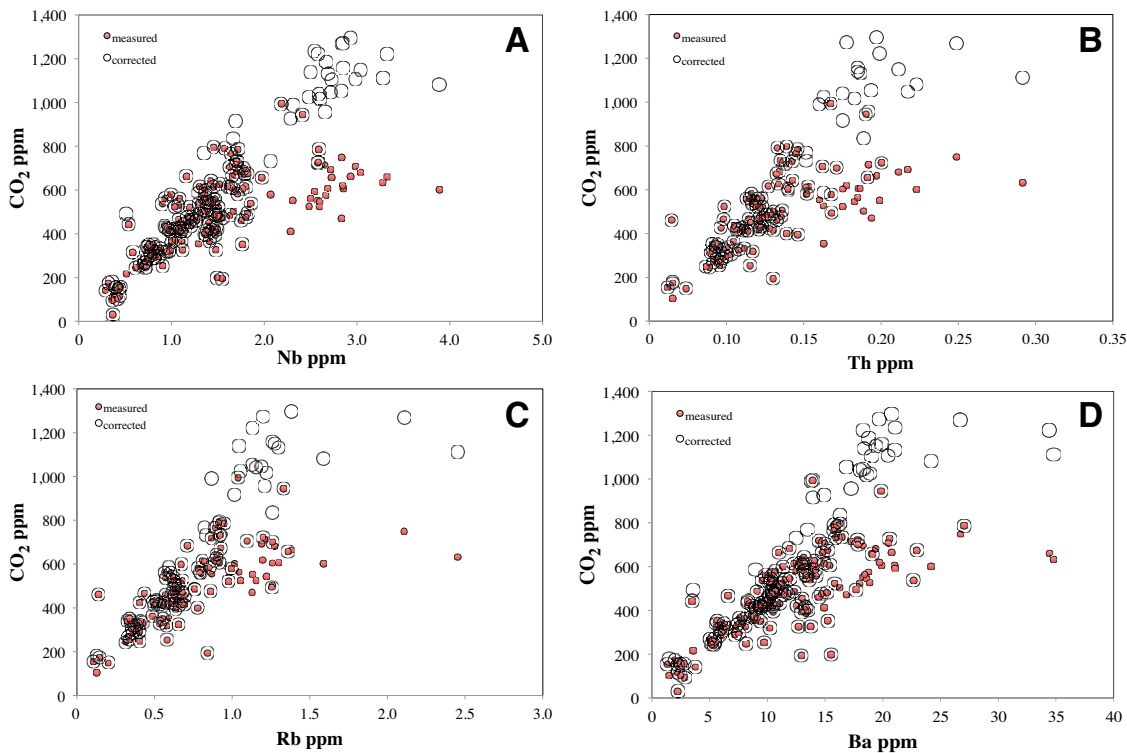
In this study, we report on the volatile content of silicate melt inclusions from Borgarhraun, a monogenetic volcano from northern Iceland that erupted during the last Northern Hemisphere deglaciation (Maclennan et al., 2001). Sample NAL709 is a tephra collected from the site of the eruption vent (lat 65.8234°N, long 16.8665°W), and contains phenocrysts of olivine, Cr-diopside, Cr-spinel, and calcic plagioclase. Detailed studies of Borgarhraun have revealed that the erupted magma was of primitive composition, produced from the local mid-oceanic ridge basalt (MORB) source mantle surrounding Iceland, and was chemically and isotopically heterogeneous (Stracke et al., 2003). Clinopyroxene barometry suggests that the melt began to crystallize at depths as great as 25 km (Maclennan et al., 2001). Melt inclusions in olivine and diopside are numerous and large, with some inclusions >300 μm in diameter (Fig. 1); melt inclusions in Cr-spinel are smaller (<60 μm diameter). We have determined the major element, trace element, and volatile element contents (H<sub>2</sub>O, CO<sub>2</sub>, F, S, Cl) of 205 melt inclusions from this sample (see analysis methods and uncertainties, and Table DR1, in the GSA Data Repository<sup>1</sup>).

## RESULTS

The average major element compositions of the melt inclusion populations from each of the three phenocrysts phases are within error of each other and the Borgarhraun whole-rock compositions, indicating minimal postentrapment modification. Unlike most other melt inclusion studies, we found that most of the melt inclusions (84%) did not display shrinkage bubbles, which can form within melt inclusions due to differential shrinkage of melt and crystal during cooling and result in exsolution of a vapor phase within the melt inclusion while it is still molten (e.g., Moore et al., 2015; Maclennan, 2017). Most of the NAL709 inclusions that contain shrinkage bubbles were found to have trace element compositions with >2 ppm Nb.

The correlation of CO<sub>2</sub> concentrations with the incompatible trace elements Nb, Th, Rb, and Ba is observed in melt inclusions from all three crystal phases and is independent of the presence or absence of a shrinkage bubble. The correlation of CO<sub>2</sub> with nonvolatile trace elements demonstrates that these melt inclusions were trapped before the magma became saturated in a CO<sub>2</sub>-rich vapor phase. The maximum CO<sub>2</sub> concentrations indicate minimum trapping depths of 8–10 km for the

<sup>1</sup>GSA Data Repository item 2018012, methods, data comparisons, mantle CO<sub>2</sub> abundance and flux estimates, and full data tables, is available online at <http://www.geosociety.org/datarepository/2018/> or on request from [editing@geosociety.org](mailto:editing@geosociety.org).



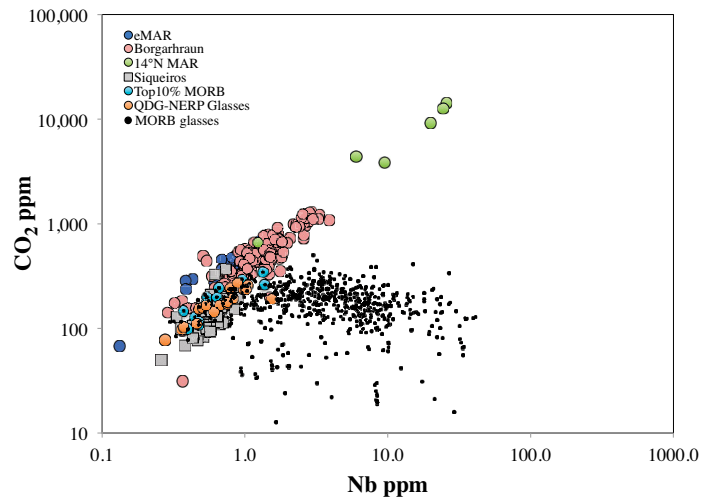
**Figure 1. A: CO<sub>2</sub> versus Nb in Borgarhraun (Iceland) melt inclusions. Solid symbols represent measured CO<sub>2</sub> concentrations; open symbols include CO<sub>2</sub> added from vapor bubbles where present. B: CO<sub>2</sub> versus Th. C: CO<sub>2</sub> versus Rb. D: CO<sub>2</sub> versus Ba.**

initiation of magma crystallization, using the H<sub>2</sub>O-CO<sub>2</sub> solubility model of Dixon et al. (1995). This is consistent with CO<sub>2</sub>-trace element correlations indicating that the melts had not degassed significantly at the time the inclusions were trapped.

#### SYSTEMATICS OF CO<sub>2</sub> AND NONVOLATILE TRACE ELEMENTS

The undegassed nature of the Borgarhraun melt inclusions is an exceedingly rare occurrence, previously observed at only two other locales, the Siqueiros transform on the East Pacific Rise (Saal et al., 2002) and the equatorial Mid-Atlantic Ridge (MAR) (Le Voyer et al., 2016). The well-correlated abundances of CO<sub>2</sub> and incompatible trace elements (ITEs) result in tightly constrained CO<sub>2</sub>/ITE ratios for this population of melt inclusions (e.g., CO<sub>2</sub>/Nb = 391 ± 16, 2σM [two standard errors of the mean], n = 161; Fig. 2). Separate averages of data for melt inclusions trapped in olivine, diopside, and Cr-spinel all overlap each other within errors, and agree with the CO<sub>2</sub>/Nb ratio of the entire melt inclusion population within errors. Similarly precise ratios are obtained for CO<sub>2</sub>/Th (7300 ± 540), CO<sub>2</sub>/Rb (787 ± 36), and CO<sub>2</sub>/Ba (48.3 ± 2.7).

When comparing the Borgarhraun CO<sub>2</sub>/ITE ratios with other sample suites, a limiting factor is the generally large scatter in the estimated pre-eruptive abundances of CO<sub>2</sub>. Thus any comparison of our data to published data must be limited to specific subsets of the global data that report correlated CO<sub>2</sub> and ITE abundances and thus well-constrained CO<sub>2</sub>/ITE ratios. Among the published data, only 4 sample populations meet the criteria for determination of CO<sub>2</sub>/Nb with a precision better than 20% (relative); 2 are from the eastern Pacific (Saal et al., 2002; Shimizu et al., 2016) and the other 2 are from the Atlantic Ocean (Cartigny et al., 2008; Le Voyer et al., 2016). In addition to these four populations, we compare our data with vapor-undersaturated submarine MORB glasses identified from published volatile data using the PetDB petrologic database (<http://www.earthchem.org/petdb>), using only samples with CO<sub>2</sub> data obtained by Fourier-transform infrared spectroscopy or ion microprobe with corresponding data for Rb, Ba, Nb, or Th on the same sample (n = 161). This compilation includes samples from the global data set of Michael and Graham (2015) whose global average CO<sub>2</sub>/Nb is also precisely determined



**Figure 2. CO<sub>2</sub> versus Nb for Borgarhraun (Iceland) melt inclusions (this study) compared with five other high-precision data sets on vapor-undersaturated samples: melt inclusions from the equatorial Mid-Atlantic Ridge (MAR; Le Voyer et al., 2016), glasses from 14°N MAR (Cartigny et al., 2008), melt inclusions from the Siqueiros Fracture Zone (Saal et al., 2002), glasses from the Quebrada-Discovery-Gofar transform and northern East Pacific Rise (QDG-NEPR; Shimizu et al., 2016), and glasses from the Pacific Ocean basin that record the highest 10% of CO<sub>2</sub>/Nb ratios among the literature data. MORB—mid-oceanic ridge basalt.**

(±9% 2σM). The ratios CO<sub>2</sub>/Ba and CO<sub>2</sub>/Rb show a limited range of values; average CO<sub>2</sub>/Rb ratios are nearly the same among the six populations of MORB (±26% 2σ), while CO<sub>2</sub>/Ba varies by ±56% (Table 1; Fig. 3). These results are consistent with the previously observed limited variation of Rb/Ba ratios in global MORB (Hofmann and White, 1983; Jenner and O'Neill, 2012; Kelley et al., 2013).

The situation is different when comparing CO<sub>2</sub> abundances with Nb and Th in these sample suites. The CO<sub>2</sub>/Nb ratio of 391 ± 16 for Borgarhraun is significantly higher than the CO<sub>2</sub>/Nb ratio determined for MORB

TABLE 1. CO<sub>2</sub>-ITE RATIOS AND MANTLE SOURCE CO<sub>2</sub> ABUNDANCES FOR SIX MID-OCEAN RIDGE SAMPLE GROUPS, ALL-MORB, AND AVERAGES FOR ATLANTIC AND PACIFIC OCEAN BASINS

	Borgarhraun <sup>1</sup>	Equatorial MAR <sup>2</sup>	14°N MAR <sup>3</sup>	Siqueiros <sup>4</sup>	QDG-NEPR <sup>5</sup>	Top 10% MORB <sup>6</sup>	ALL-MORB*	Atlantic	Pacific	
CO <sub>2</sub> /Nb	391	557	534	230	277	285	435	414	243	
CO <sub>2</sub> /Th	7304	8253	9765	2999	4039	4438	7383	7553	3306	
CO <sub>2</sub> /Rb	787	1105	1153	955	971	974	1075	852	972	
CO <sub>2</sub> /Ba	48.3	96.6	105.6	74.2	99.8	63.6	101	55.5	76.9	
ε <sub>Nd</sub>	8.78	13.11	7.19	10.36	10.31	9.74	8.51			
H <sub>2</sub> O/Ce <sup>†</sup>	477	423	187	170	227	178	—	368	179	
Mantle CO <sub>2</sub> ppm (Nb)	129	48	393	23	30	51	166	190	35	
Mantle CO <sub>2</sub> ppm (Th)	146	33	459	15	22	44	174	213	27	
Mantle CO <sub>2</sub> ppm (Rb)	118	22	422	27	30	67	191	187	41	
Mantle CO <sub>2</sub> ppm (Ba)	80	22	427	24	35	49	198	176	36	
Average mantle CO <sub>2</sub> ppm	118 ± 64	31 ± 20	425 ± 291	22 ± 17	29 ± 18	53 ± 38	182 ± 59	111 ± 40	67 ± 13	
Total MOR CO <sub>2</sub> flux mol/yr								2.38(±0.92) × 10 <sup>12</sup>		

Note: ITE—incompatible trace element; MORB—mid-oceanic ridge basalt; MAR—Mid Atlantic Ridge; QDG-NEPR—Quebrada-Discovery-Gofar transform and northern East Pacific Rise; MOR—mid-oceanic ridge. Data sources: 1—this study; 2—Le Voyer et al. (2017); 3—Cartigny et al. (2008); 4—Saal et al. (2002); 5—Shimizu et al. (2016); 6—PetDB (Petrological Database, <http://www.earthchem.org/petdb>; this study).

\*ALL-MORB Nd isotopes from Gale et al. (2103); CO<sub>2</sub>-ITE (incompatible trace element) ratios estimated from correlations with Nd isotopes (CO<sub>2</sub>/Nb, CO<sub>2</sub>/Th) or population averages (CO<sub>2</sub>/Rb, CO<sub>2</sub>/Ba) of the sample populations. Mantle source ITE concentrations estimated using Nd isotopes from the MORB source mixing model of Shimizu et al. (2016) as for other groups (see text). Dash (—) represents “not determined.”

<sup>†</sup>H<sub>2</sub>O/Ce ratios for Atlantic and Pacific are the averages of the sample populations discussed in this study.

from the eastern Pacific; melt inclusions from the Siqueiros Fracture Zone on the East Pacific Rise (Saal et al., 2002) have CO<sub>2</sub>/Nb = 230 ± 12 (n = 100), while undegassed submarine glasses from the eastern Pacific (Shimizu et al., 2016) also demonstrate a correlation of CO<sub>2</sub> with Nb yielding a CO<sub>2</sub>/Nb ratio of 277 ± 14 (n = 19). Similar differences are seen in CO<sub>2</sub>/Th.

The Borgarhraun CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th ratios are significantly higher than the Pacific suites, and closer to the high-precision measurements

of MORB melt inclusions from the equatorial MAR (Table 1; Fig. 3), which yield CO<sub>2</sub>/Nb = 557 ± 34 and CO<sub>2</sub>/Th = 8250 ± 760 (n = 21) (Le Voyer et al., 2016). Submarine glass samples from the 14°N segment of the MAR, that include the popping rock 2piD43, have CO<sub>2</sub>/Nb = 534 ± 90 and CO<sub>2</sub>/Th = 9770 ± 1560 (n = 6) (Cartigny et al., 2008). It thus appears that the Atlantic locales (Borgarhraun included) are uniformly higher than the Pacific locales; a simple averaging by ocean basin yields a CO<sub>2</sub>/Nb for the Atlantic (414 ± 17) that is 70% higher than CO<sub>2</sub>/Nb for the Pacific (243 ± 11); the Atlantic CO<sub>2</sub>/Th ratio (7550 ± 460) is more than twice the Pacific CO<sub>2</sub>/Th (3310 ± 220).

### SUBDUCTION ORIGIN FOR MANTLE CO<sub>2</sub> VARIATIONS

The CO<sub>2</sub>/Rb and CO<sub>2</sub>/Ba ratios are clearly more homogeneous than CO<sub>2</sub>/Th and CO<sub>2</sub>/Nb ratios. Five of the six undegassed MORB populations show correlations between <sup>143</sup>Nd/<sup>144</sup>Nd, CO<sub>2</sub>/Nb, and CO<sub>2</sub>/Th (Fig. 3) that suggest that these variations are long-term characteristics of the upper mantle. The more homogeneous CO<sub>2</sub>/Ba and CO<sub>2</sub>/Rb ratios do not show correlations with Nd isotopes, indicating that these ratios are similar in isotopically depleted and enriched components in the upper mantle sources of MORB. It is thus likely that the variable mantle CO<sub>2</sub> contents originate as a result of variable mixing of depleted mantle sources (high <sup>143</sup>Nd/<sup>144</sup>Nd, low CO<sub>2</sub>/Nb, and CO<sub>2</sub>/Th) with small amounts of a subduction component containing elevated abundances of CO<sub>2</sub>, Rb, and Ba (Fig. 3), a signature characteristic of subduction zone fluids (Elliott et al., 1997; Kessel et al., 2005; Kelemen and Manning, 2015). In addition, the regional differences in mantle CO<sub>2</sub> documented here between the Atlantic (high CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th) and Pacific (low CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th) ocean basins correspond to a similar difference in mantle H<sub>2</sub>O abundance, as Atlantic MORB has higher H<sub>2</sub>O/Ce than Pacific MORB (Michael, 1995). There are similar distinctions in Ba/Nb and Ba/La ratios, with Atlantic MORB being systematically higher than Pacific MORB (e.g., Arevalo and McDonough, 2010; Jenner and O’Neill, 2012; Kelley et al., 2013). Among the four nonvolatile trace elements we have considered together with CO<sub>2</sub>, both Nb and Th have low solubility in hydrous fluids from subduction zones (Kessel et al., 2005) that are expected to be enriched in H<sub>2</sub>O, Rb, and Ba as well as carbonate (Kelemen and Manning, 2015). Thus CO<sub>2</sub>/Ba and CO<sub>2</sub>/Rb would be expected to behave similarly to each other in fluid-dominated subduction enrichment processes, and distinct from CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th. The high H<sub>2</sub>O/Ce, Ba/Nb, and Ba/La, and low <sup>143</sup>Nd/<sup>144</sup>Nd are all signatures of mantle enrichment that are characteristic of subduction zone magmas (Elliott et al., 1997; Ruscitto et al., 2012), and by inference high CO<sub>2</sub>/ITE ratios are also characteristic of this signature.

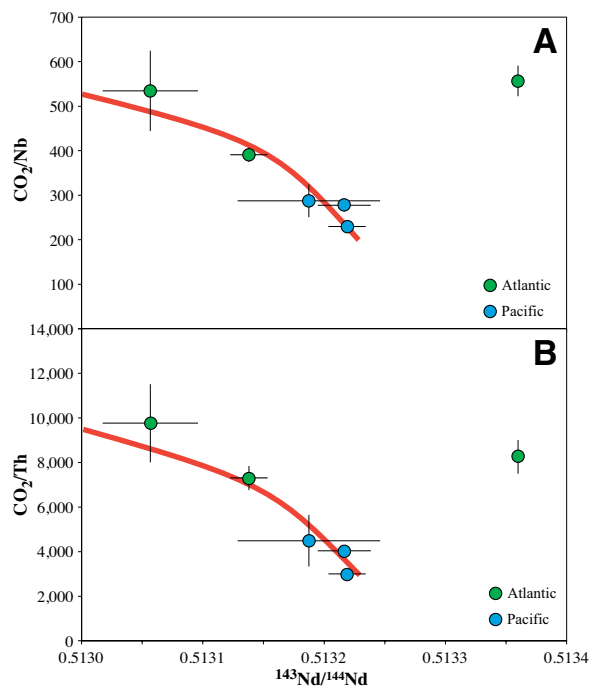


Figure 3. A: Average CO<sub>2</sub>/Nb versus Nd isotopes for the six mid-ocean ridge regions that exhibit CO<sub>2</sub>-incompatible trace element correlations. B: CO<sub>2</sub>/Th versus Nd. Five of the six regions define a correlation suggesting that CO<sub>2</sub>/Nb and CO<sub>2</sub>/Th heterogeneities are long-lived features of mantle sources in the convecting upper mantle. The red curve is a model mixing line between enriched and depleted mid-ocean ridge basalt sources from the model of Shimizu et al. (2016) with CO<sub>2</sub>, Rb, and Th concentrations in the depleted end member adjusted to best fit the data (see text). Atlantic locales have lower <sup>143</sup>Nd/<sup>144</sup>Nd, higher CO<sub>2</sub>/Nb, and higher CO<sub>2</sub>/Th, as well as higher H<sub>2</sub>O/Ce (Michael, 1995) and higher Ba/Nb (Arevalo and McDonough, 2010), characteristic of subduction-modified mantle.

We therefore suggest that the Atlantic Ocean basin has been polluted by small amounts of mantle wedge material that was hydrated and carbonated during the subduction episodes that characterized the convergent margins surrounding the ancient Iapetus Ocean. Opening of the Atlantic Ocean basin along the preexisting Iapetus suture provided opportunity for Iapetus subarc mantle to become entrained and mixed into the asthenospheric flow that produced the mantle that is sampled today along the MAR. The Pacific Ocean basin, having initially formed during the breakup of the Rodinia supercontinent ~750 m.y. ago (Evans, 2009), is nearly 4 times older than the Atlantic Ocean basin (~200 m.y.; McHone and Butler, 1984) and has thus had a more extended history of upper mantle convection to flush out continent-proximal suprasubduction components. As a result, the eastern Pacific mantle is less affected by mixing with subcontinental subduction-modified mantle, resulting in lower H<sub>2</sub>O/Ce, Ba/Nb, Ba/La, CO<sub>2</sub>/Nb, and CO<sub>2</sub>/Th. This conceptual model thus argues for the importance of prior tectonic episodes of subduction in determining the spectrum of volatile, trace element, and isotopic compositions present in the upper mantle beneath ocean basins of different age.

The melt inclusions from Borgarhraun, equatorial MAR, and Siqueiros are the only three documented examples of vapor-undersaturated melt inclusions in existence. However, given that there are regions of the mid-ocean ridge system where MORB magmas erupt with little or no degassing, as documented here and in Shimizu et al. (2016) and Michael and Graham (2015), more vapor-undersaturated melt inclusions are certain to be discovered, and future studies will be very important for constraining the regional distribution of carbon in the upper mantle, the variability of the carbon flux from mid-ocean ridges, and ultimately the origin of carbon throughout the upper mantle.

#### ACKNOWLEDGMENTS

We thank Jianhua Wang for expert assistance with the Carnegie ion microprobes. MacLennan is supported by Natural Environment Research Council grant NE/M000427/1. This research was supported by the Carnegie Institution of Washington and is a contribution to the Deep Carbon Observatory.

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Manuscript received 14 June 2017

Revised manuscript received 18 October 2017

Manuscript accepted 18 October 2017

Printed in USA