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Special Section:
Carbon degassing through volcanoes and active tectonic regions

Carbon Fluxes and Primary Magma CO_2 Contents Along the Global Mid-Ocean Ridge System

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Key Points:
- New analyses (753) and compiled data (2,446) for volatiles CO_2, H_2O, F, S, and Cl are reported in a global suite of mid-ocean ridge basalts
- Estimated primary MORB CO_2 contents vary from 104 ppm to 1.9 wt% and are the result of variations in mantle composition
- High CO_2 fluxes at ridges correlate with high primary CO_2 contents and are due to the presence of subduction components in MORB sources

Supporting Information:
- Supporting Information S1
- Data Set S1

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Abstract The concentration of carbon in primary mid-ocean ridge basalts (MORBs), and the associated fluxes of CO_2 outgassed at ocean ridges, is examined through new data obtained by secondary ion mass spectrometry (SIMS) on 753 globally distributed MORB glasses. MORB glasses are typically 80–90% degassed of CO_2. We thus use the limited range in CO_2/Ba (81.3 ± 23) and CO_2/Rb (991 ± 129), derived from undegassed MORB and MORB melt inclusions, to estimate primary CO_2 concentrations for ridges that have Ba and/or Rb data. When combined with quality-controlled volatile-element data from the literature (n = 2,446), these data constrain a range of primary CO_2 abundances that vary from 104 ppm to 1.90 wt%. Segment-scale data reveal a range in MORB magma flux varying by a factor of 440 (from 6.8×10^5 to 3.0×10^8 m^3/year) and an integrated global MORB magma flux of 16.5 ± 1.6 km^3/year. When combined with CO_2/Ba and CO_2/Rb-derived primary magma CO_2 abundances, the calculated segment-scale CO_2 fluxes vary by more than 3 orders of magnitude (3.3×10^7 to 4.0×10^{10} mol/year) and sum to an integrated global MORB CO_2 flux of $1.32^{+0.77}_{0.85} \times 10^{12}$ mol/year. Variations in ridge CO_2 fluxes have a muted effect on global climate; however, because the vast majority of CO_2 degassed at ridges is dissolved into seawater and enters the marine bicarbonate cycle. MORB degassing would thus only contribute to long-term variations in climate via degassing directly into the atmosphere in shallow-water areas or where the ridge system is exposed above sea level.

Plain Language Summary Estimated CO_2 contents of primary mid-ocean ridge basalts (MORB), calculated on a segment-by-segment basis, vary from 104 ppm to 1.9 wt%. CO_2-enriched MORB are present in all ocean basins, in particular, in the Atlantic Ocean basin, which is younger and more likely to contain admixed material from recent subduction compared to the much older Pacific Ocean basin. CO_2 fluxes at individual ridge segments vary by 3 orders of magnitude due primarily to large variability in primary CO_2 content. This study provides the most detailed and accurate estimate to date of the integrated total flux of CO_2 from mid-ocean ridges of $1.32^{+0.77}_{0.85} \times 10^{12}$ mol/year.

1. Introduction

The 56,000-km length of the global mid-ocean ridge system represents the dominant locus of magma production and heat loss on Earth. Decades of deep-water oceanographic research have revealed that new seafloor is produced over a limited range of crustal thicknesses (6 ± 1 km, Behn & Grove, 2015; Klein & Langmuir, 1987; Van Avendonk et al., 2017; White et al., 1992) and at variable spreading rates that are well-described globally (e.g., Gale et al., 2013). As a result, magma production rates at mid-ocean ridges are well understood and the fluxes of nonvolatile elements emerging from the mantle at ridges are well constrained. These same research cruises have retrieved tens of thousands of individual submarine mid-ocean ridge basalt (MORB) samples, and the study of these samples has revealed that chemical variations in MORB magmas are produced by polybaric melting of mantle sources at variable temperatures, whereas local deviations from global trends are primarily due to variations in mantle composition (Gale et al., 2014; Kelley et al., 2013; Kinzler & Grove, 1992; Klein & Langmuir, 1987; Langmuir et al., 1992).

Carbon is present in MORBs in trace amounts as CO_2-rich vesicles and as carbonate ions dissolved in quenched seafloor glass (Cartigny et al., 2008; Dixon & Stolper, 1995; Javoy & Pineau, 1991; Marty & Tolstikhin, 1998; Michael, 1995). The solubility of CO_2 in basaltic magma is low at hydrostatic pressures corresponding to seafloor water depths (e.g., Dixon et al., 1995; Jendrzejewski et al., 1997; Shishkina et al., 2010), and thus nearly all magmas erupted at mid-ocean ridges have degassed the majority of their carbon. This flux of CO_2 into the oceans contributes to the global surface carbon cycle, and changes in basaltic magma production and associated degassing have been proposed as one of several important forcing mechanisms that have influenced past global climate variations (Burley & Katz, 2015; Robock, 2000; Shindell et al., 2003; Tolstoy, 2015; Zhong et al., 2010). At the same time, it is now widely recognized that most of the Earth's carbon is present in the Earth's deep interior, rather than at Earth's surface (Marty et al., 2013; Marty & Tolstikhin, 1998). Importantly, basaltic magmatism and subduction of oceanic crust represent the two largest outgassing and ingassing fluxes at the interface between the surface and deep-Earth carbon cycles.

These observations provide the motivation of the present study, in which we apply the rapid, precise, and accurate measurement of volatiles by secondary ion mass spectrometry (SIMS; Hauri et al., 2002) to characterize the extensive worldwide sample collection of MORBs to examine in detail the flux of CO_2 from the global ridge system. In this study we report new SIMS data on 753 geographically distributed MORB samples, and combine these data with a quality-controlled database of published data from an additional 2,446 samples (Figure 1). This combined data set permits a description of CO_2 fluxes, and estimated primary magma CO_2 contents, for 381 of the 458 global ridge segments with major element data, in addition to examination of the variability of primary MORB CO_2 contents, the role of carbon in the melting processes that produce MORB, and the role of the oceanic crust in the context of the Earth's deep carbon cycle.

2. Methods

Here we briefly describe the methods used in this study for sample preparation, SIMS analysis of volatile elements, major elements, and trace elements.

2.1. Sample Preparation

The majority of the basalt glasses analyzed in this study were obtained from the Rock and Ore collection of the Smithsonian Institution's National Museum of Natural History. The equatorial mid-Atlantic Ridge glasses were obtained from the Schilling Volcanic Glass collection at the University of Rhode Island (University of Rhode Island Graduate School of Oceanography Marine Geological Samples Laboratory, 2018). Additional glass samples were provided from the Pacific-Antarctic Ridge (PACANTARCTIC 1 and 2 cruises, Clog et al., 2013; Hamelin et al., 2010; Labidi et al., 2014; Moreira et al., 2008; Vlastelic et al., 1999, 2000), the Central Indian Ridge (GIMNAUT, CD127, and KNOX11RR cruises, Füri et al., 2011; Murton et al., 2005; Barry, 2012), the Southwest Indian Ridge (PROTEA-5, MD34, AG22, and AG53 cruises, Arevalo & McDonough, 2010; B. Hamelin & Allègre, 1985; Janney et al., 2005; Le Roex et al., 1989; Mahoney et al., 1992), and the East Pacific Rise (CHEPR and PANORAMA-1 cruises, Langmuir, 2014; Salters et al., 2011). The major element, trace element, and isotopic ratio data for the glasses measured in this study were compiled from the literature (Clog et al., 2013; Cottrell & Kelley, 2011; C. Hamelin et al., 2010; Jenner & O'Neill, 2012; Labidi et al., 2014; Le Voyer et al., 2015; Moreira et al., 2008; Vlastelic et al., 2000, 1999; Nauret et al., 2006). We selected 1- to 3-mm glass chips that did not show any sign of alteration, crystallization, or devitrification. The chips were pressed into indium mounts together with secondary standards (ALV519-4-1 and VG2), and polished using silicon carbide papers, diamond paste, and finally 0.3-μm alumina paste. We washed the mounts using successive baths of distilled water, ethanol, and acetone, then stored them for >48 hr in a vacuum oven at 70 °C, before applying a gold coat.

2.2. Volatile Elements

CO_2 and other volatile elements, along with phosphorus, were measured by SIMS at the Department of Terrestrial Magnetism (DTM), Carnegie Institution of Washington, following the method of Hauri et al. (2002) but with the modification of measuring ^{16}OH (rather than ^{1}H) for determination of H_2O contents. These new data are reported in supporting information Table S1. The analyses were performed during 13 analytical sessions from March 2012 to October 2016, for a combined analytical time of 59 days. For 12 of the 13 sessions, we used a Cs^+, 15 ± 2 nA primary beam accelerated to 10 kV, with charge compensation

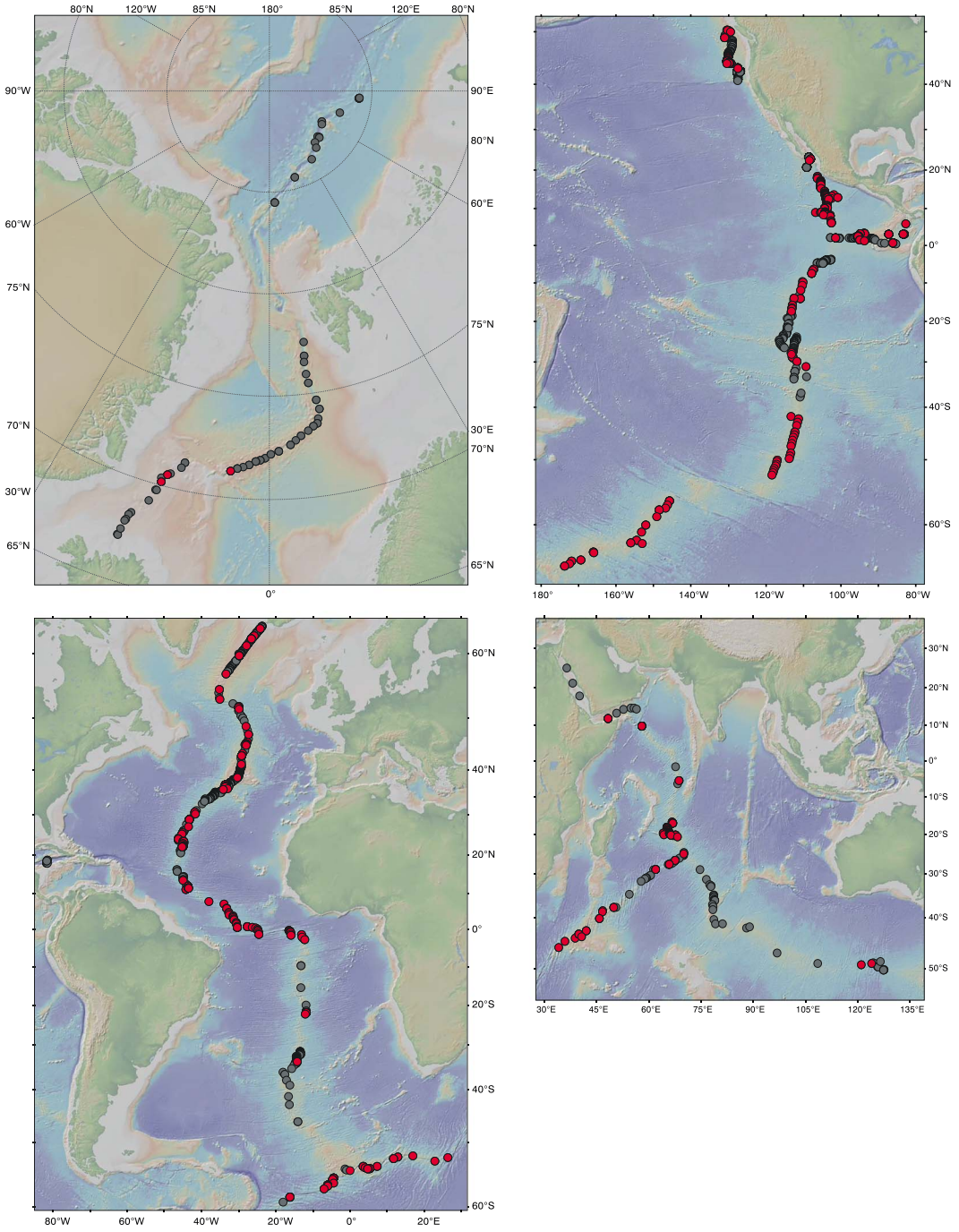

Figure 1. Locations of newly measured mid-ocean ridge basalt samples (n = 753) are shown as red circles, with locations of previously published samples (n = 2,446) shown as gray circles.

provided by an electron flood gun. For one session we used a 10 ± 1 nA primary beam instead, which did not create any differences in the results. For 11 of the 13 sessions we applied a 25-μm^2 raster on the beam, which resulted in a crater size of ~35 μm^2. For two sessions we used a 20-μm^2 raster size, and no differences in the results were observed. The vacuum was in the 10^{-9} to 10^{-10} torr range during each analysis. We used the smallest field aperture (#4) in order to mask the edge of the crater and extract data from a central 10-μm-diameter area in the center of the sputter crater. The mass spectrometer was set to a mass resolution sufficient to separate ^{17}O from ^{16}OH, ^{18}OH from ^{19}F, and $^{16}O_2$ from ^{32}S (Mass Resolving Power ~ 6,000).

Table 1
Concentrations and Reproducibility of Volatile and Phosphorus Contents of Smithsonian Basalt Glass Standard VG2 Measured During the Course of This Study

	CO_2	H_2O	F	P	S	Cl
Unit	ppm	wt%	ppm	ppm	ppm	ppm
Number of analyses (*n*)	76	74	76	77	75	76
Average over *n* analyses	187	0.28	211	970	1,486	259
2σ	16	0.02	15	43	53	17
2σ (%)	8	7	7	5	4	7

After 300 s of presputtering, we successively collected signal for 10 s on mass ^{12}C, 5 s on masses ^{16}OH, ^{19}F, ^{30}Si (reference mass), ^{31}P, ^{32}S, and ^{35}Cl. The waiting time before signal collection was 2 s before ^{12}C and 1 s before all other masses. This cycle was repeated 5 times. A set of basaltic standards and blanks (see Hauri et al., 2002) were used to perform the calibration (linear regression forced through the origin) and to assess detection levels (average detection levels over the 13 sessions: 5 ppm for H_2O, 2 ppm for CO_2, 0.2 ppm for S, Cl, and P, and 0.1 ppm for F). The analytical error (2σ over the five analytical cycles) was typically 4% for CO_2, 2% for others. Each sample was measured three times, and the reproducibility over the three analyses was typically better than 10% for CO_2 and 5% for other elements. Standard ALV519-4-1 was mounted together with samples in each indium mount, and measured every nine analyses, in order to correct for potential instrumental drift within each session, potential offsets between sessions, and to assess long-term reproducibility (better than 5%, 2RSD, for all volatiles, over 2 years). Standard VG-2 was also mounted together with samples in each indium mount to assess the reliability of this correction, and results are reported in Table 1. The reproducibility on VG2 was better than 8% for all volatiles and phosphorus (2σ over 2 years).

3. Published Data Quality Control

Data from PetDB (www.earthchem.org/petdb) for MORB glasses and melt inclusions, consisting of samples containing data for at least one volatile (CO_2, H_2O, F, S, Cl) were downloaded in late 2017; the data are compiled, along with the newly reported data from this study, in supporting information Table S1, and the references for the original data are included in the supporting information Table S5. All corresponding data (major elements, trace elements, isotopes) were included in this download. Concentrations were recalculated as weight percent for H_2O, and parts per million by weight for CO_2, F, S, and Cl for data reported in other units or as oxides (e.g., C data were converted to CO_2 ppm; SO_2 and SO_3 data were converted to S ppm), and the database was checked for obvious typographic errors by comparison with the original references. Samples with CO_2 and/or H_2O were retained in the database if the samples were determined by SIMS, Fourier-transform infrared spectroscopy, or post-1995 vacuum extraction of the volatile fraction dissolved in the glass; vesicle-only analyses were discarded due to the inability to use the data to calculate a glass concentration. Loss on ignition data for H_2O were also discarded, as were four analyses of melt inclusions from Helo et al. (2011) with $>$3,900 ppm CO_2 that were judged to have been contaminated by carbon coat during SIMS analysis. Samples with F data were retained only if they were determined by SIMS or ion-selective electrode methods (Bach et al., 1994; Schilling et al., 1983); electron probe microanalysis (EPMA) for F is subject to interference from overlapping Fe peaks in basaltic glasses, and these data were thus rejected from the database. Samples with S or Cl data were retained if they were determined by SIMS or EPMA.

We also compiled previously published $Fe^{3+}/\Sigma Fe$ ratios of MORB glasses for this study. For this compilation, we report Fe-speciation data collected using micro X-ray absorption near-edge structure (μXANES) spectroscopy, with all data collected at beamline X26A of the National Synchrotron Light Source, Brookhaven National Laboratory, following methods described by Cottrell et al. (2009) and Zhang et al. (2018). The Zhang et al. (2018) study assessed the effects of recoil-free fraction on the room temperature Mössbauer spectra, against which the XANES centroid energy was calibrated by Cottrell et al. (2009). Room temperature Mössbauer spectra overestimate the $Fe^{3+}/\Sigma Fe$ ratios of basaltic glasses, and the updated calibration of Zhang et al. (2018) accounts for this effect. The $Fe^{3+}/\Sigma Fe$ ratios compiled here include those reported by Cottrell and Kelley (2011) and Le Voyer et al. (2015), both updated using the new calibration of Zhang et al. (2018).

Interlaboratory bias between the DTM SIMS method and published data sets is essentially nonexistent, for several reasons. SIMS calibrations for H_2O and CO_2 rely on the use of an extensive suite of MORB and back-arc basin glasses previously analyzed by vacuum manometry and/or Fourier-transform infrared spectroscopy (references in Hauri et al., 2002) originating from laboratories that provided the vast majority of previously published data archived in PetDB. The SIMS method for S and Cl is more precise and has lower detection limits than EPMA. The precision of bias factors between data sets is limited by the reproducibility of the EPMA data and is thus indeterminate. For F the SIMS calibration, which relies on only four glasses (Hauri

et al., 2002), was checked against the data sets of Rowe and Schilling (1979), Schilling et al. (1983), and Bach et al. (1994) by SIMS analysis of samples in common with these studies, which used a halogen-selective electrode wet chemical method. We found excellent agreement (±8% 2σ) with the data set of Bach et al. (1994) but much poorer agreement with the other, significantly older, data sets that used the same method for F analysis. Thus, the comparison for F found no interlaboratory bias but does show clear examples of variable extents of reproducibility of other methods compared with SIMS, which likely permeate much of the published data for fluorine and, to a lesser extent, sulfur and chlorine. In general, interlaboratory comparisons agree to better than 10% for CO_2, H_2O, and F, and by a somewhat worse and variable degree for S and Cl.

The supporting information contains the data used in this paper and is available through the EarthChem Library (https://doi.org/10.1594/IEDA/111195). Table S1 contains the results of new analyses of MORB glasses, together with associated data on major and trace elements, and isotopic compositions. In Table S2, these new data are compiled along with the quality-controlled published data in a single table. Table S3 is a compilation of data (volatiles, major elements, trace elements, isotope compositions of host glass) for MORB melt inclusions, for comparison to the submarine glass data. Table S4 is a modification of the segment-averaged tables from Gale et al. (2013, 2014) that includes data for segment metrics (length, spreading rate, depth, etc.) as well as averaged data for major elements, trace elements, volatiles, and isotopic compositions; this table also includes estimated primary magma CO_2 concentrations, and calculated CO_2 fluxes on a per-segment basis, with estimated uncertainties shown in yellow highlight. Table S5 contains an alphabetic list of all the publications that contained the original data that were used in the data compilations of all of the data tables.

4. Results

The samples in this compiled volatile data set were assigned to individual ridge segments based on a comparison of their sampling coordinates with the global spreading center segment catalog of Gale et al. (2013; Figure 1). In constructing geochemical averages of global spreading segments, Gale et al. (2013) included in their compilation samples within 10 km of the ridge axis. At the slowest spreading rates, a distance of 10 km off-axis corresponds to an age of approximately 1.5 million years, and so in our study individual samples were categorized as *on-axis* if their apparent age was less than or equal to 1.5 million years. Samples older or farther than this were categorized as either *off-axis* or as *transform* samples depending on their location relative to the nearest ridge axis or transform fault. Segment average volatile concentrations were then calculated for all segments that contained at least one on-axis sample analyzed for H_2O, CO_2, F, S, and/or Cl. The PetDB data download also contained a small amount of new major element, trace element, and isotope data published after the compilations of Gale et al. (2013) and Class and Lehnert (2012), and this permitted the calculation of several new segment averages for major elements, trace elements, and isotopes to augment the database. For consistency, these updated segment averages were calculated using the same methods described in Gale et al. (2013); however, it should be mentioned that only samples with volatile data were used to calculate these additional new segment averages. Finally, we recalculated the segment-averaged major and trace element data for segment EPRR31 to account for two additional samples from this segment analyzed by Cottrell and Kelley (2011); samples from this segment have a bimodal distribution of depleted ($n = 3$) and enriched ($n = 3$) MORB. By doing this, the segment averages for all data (major elements, trace elements, volatiles) all come from the same six samples.

The MORB glasses measured in this study range from 2 to 420 ppm CO_2, 0.04 to 1.37 wt% H_2O, 41 to 1,131 ppm F, 121 to 2,380 ppm S, and 1.7 to 5,540 ppm Cl. These new volatile data extend the range of MORB H_2O, F, and Cl abundances, while the data for CO_2 and S cover most of the range defined by previously published data for MORB (up to 600 ppm CO_2 and 3,300 ppm S). H_2O and F are broadly correlated in the global data (Figure 2a), with an average H_2O/F ratio of 14.3 but with a large range displayed by individual samples (0.56 to 216). Sulfur is correlated with total Fe (FeO*) (Figure 2b), reflecting the dependence of sulfide solubility on iron content in mafic melts (Keith et al., 2017; O'Neill & Mavrogenes, 2002). Chlorine is generally not well correlated with H_2O, but a lower bound to the global data is apparent with a Cl/H_2O ratio in the range of 0.01 to 0.05. Most of the samples with higher Cl/H_2O ratios have likely undergone assimilation of Cl-rich seawater-derived components (Le Roux et al., 2006; Le Voyer et al., 2015; Michael & Schilling, 1989). There does not appear to be any relationship of the volatiles with the oxidation state of Fe in MORBs examined in this study.

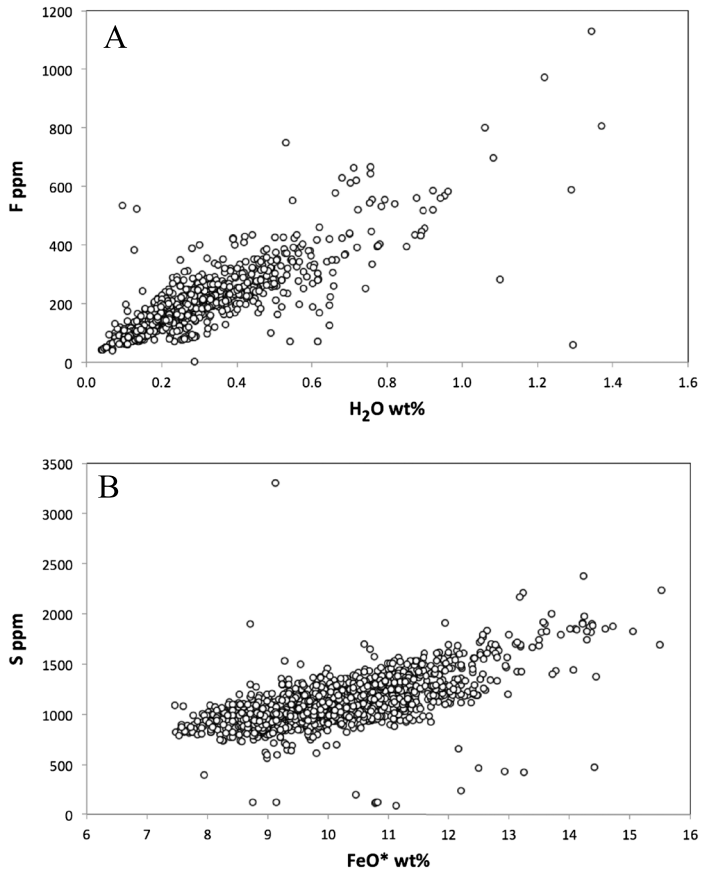

Figure 2. (a) Fluorine versus H_2O and (b) FeO* versus S in mid-ocean ridge basalt glasses from this study. Fluorine and H_2O are not degassed at seafloor pressures typical of mid-ocean ridge basalt eruptions. The correlation of S with FeO* reflects the control of FeO content on the sulfur solubility in basaltic melts, which are typically saturated in a Fe-Ni-Cu monosulfide melt (Jenner et al., 2015).

Segment averages for the volatile data were corrected to MgO = 8 wt% and to compositions in equilibrium with Fo_{90} olivine, in order to compare with the fractionation-normalized database of Gale et al. (2014). This correction for crystal fractionation cannot account for the irreversible loss of CO_2 by degassing; however, we will estimate primary magma CO_2 contents using Ba and Rb as proxies, in a subsequent section (below). The scarcity of available volatile data at any given ridge segment, coupled with potential CO_2, H_2O, or S loss by magmatic outgassing, resulted in poorly defined liquid lines of descent (LLDs) for the volatile data. Thus the 8% MgO corrections for the volatile segment averages were done by calculating a fractionation factor from the ratio of the segment average $P_2O_5(8)$ divided by the segment average P_2O_5 (uncorrected) using the data given in Gale et al. (2013, 2014). P_2O_5 was chosen because it typically forms better-correlated and more linear LLDs than K_2O; nevertheless, for segments lacking P_2O_5 data the $K_2O(8)$ and K_2O data were used. An identical procedure was used to further correct the volatile segment averages to Fo_{90} values, by determining a fractionation factor from the ratio of $K_2O(90)$ to $K_2O(8)$ for each segment reported by Gale et al. (2014).

Uncertainties from numerous sources were quantified throughout the examination of the data. Analytical uncertainties for CO_2 and other elements are typically minor (a few percent relative) compared to the compositional heterogeneity within individual ridge segments; the standard deviations of segment averages were quantified in Gale et al. (2013) while standard deviations for compositions corrected to 8 wt% MgO were quantified in Gale et al. (2014). For the purposes of calculating segment magma fluxes, we have assigned a ±4 km uncertainty on ridge length, ±3 mm/year uncertainty in spreading rates, and a variable uncertainty on crustal thickness (ranging from 3% to 50%) determined by the uncertainty of a regression of estimated errors in crustal thickness versus absolute thickness determined seismically (Behn & Grove, 2015; Van Avendonk et al., 2017).

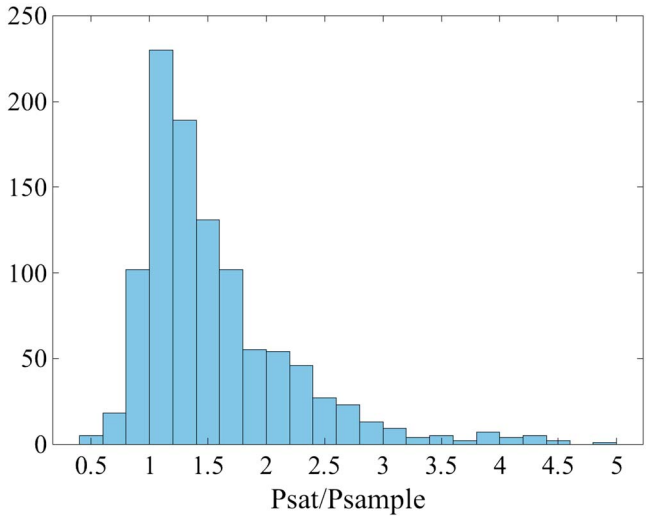

Figure 3. Histogram of the pressure of vapor saturation (P_{sat}) normalized to the pressure of sampling (P_{sample}) for the mid-ocean ridge basalt glasses measured in this study. The distribution is strongly peaked near unity but displays a tail to high ratios (oversaturation in CO_2-H_2O vapor) due to rapid magma ascent, during which vesicle growth is limited by the diffusion of carbon in the melt.

The uncertainty that is most difficult to quantify is our assumption of single average values for the ratios CO_2/Ba (81.3 ± 28%) and CO_2/Rb (991 ± 13%) for MORB; these values are the averages of the population averages of six groups of CO_2-undersaturated MORB glasses and melt inclusions (Hauri et al., 2017). To the extent that normal MORB (N-MORB) can be characterized as a mixture of depleted MORB (D-MORB) and enriched MORB (E-MORB; e.g., Donnelly et al., 2004), this assumption implies that D-MORB and E-MORB sources have the same CO_2/Ba and CO_2/Rb ratios. Two studies support this assumption. Shimizu et al. (2016) estimated CO_2 contents in Pacific E-MORB from their Cl contents and the constant CO_2/Cl ratio in Siqueiros Fracture Zone D-MORB melt inclusions, yielding CO_2/Ba ratios of 105 ± 61 for Pacific D-MORB and 134 ± 23 for Pacific EMORB. Similarly, in the Atlantic the D-MORB melt inclusion suite of Le Voyer et al. (2017) has CO_2/Ba of 97 ± 10, very close to the CO_2/Ba of 106 ± 34 of the highly enriched E-MORB popping rocks from 14°N on the Mid-Atlantic ridge (MAR; Cartigny et al., 2008; Hauri et al., 2017). Despite the possibility that this assumption may eventually be contradicted, it presently provides a data-supported basis from which to estimate the primitive, pre-eruptive CO_2 content for degassed MORB. We emphasize here that a key underpinning of our estimations of undegassed global MORB CO_2 contents is the assumed constancy of CO_2/Ba and/or CO_2/Rb across the full compositional spectrum of MORBs; uncertainties on the CO_2 concentrations estimated here will increase if these ratios are later shown to vary. We calculate undegassed MORB CO_2 concentrations by multiplying these ratios by the Ba and/or Rb concentrations measured in the basalts. In our calculations of primary CO_2 content, segment-scale CO_2 fluxes, and overall ridge flux we have combined all of these sources of error in quadrature, resulting in the stated uncertainties listed with the data in the yellow-highlighted columns of Table S4.

5. Degassing of CO_2 From MORB

MORBs are erupted on the seafloor under the hydrostatic pressure of the overlying water column. The solubility of CO_2 in MORB is strongly dependent on pressure (Dixon & Stolper, 1995), which would predict that MORB CO_2 concentrations should correlate with the pressure of eruption on the seafloor. Yet MORBs often have dissolved CO_2 contents that exceed the equilibrium solubility predicted at the eruption pressure. In most cases, this oversaturation is due to rapid melt ascent during eruption, during which diffusion of carbon in the melt is too slow to maintain equilibrium between the dissolved CO_3^{2-} in the melt and the CO_2 vapor within the vesicles (Chavrit et al., 2012; Javoy & Pineau, 1991).

The vapor solubility model of Dixon et al. (1995) has long been used to constrain vapor saturation pressure (P_{sat}) in MORBs because these authors determined CO_2 and H_2O solubility specifically on MORB melt compositions. When we use measured CO_2 and H_2O contents in MORBs to calculate P_{sat} using the Dixon et al. (1995) model, and compare these with the pressures of MORB sample collection (P_{sample}), the distribution of the P_{sat}/P_{sample} ratio is strongly peaked near unity (median = 1.37) but is clearly skewed toward high P_{sat}/P_{sample} ratios (Figure 3). This skew is likely due to vapor oversaturation in MORBs driven by high magma ascent rates, as discussed above. At the same time, there is a sharp cut-off below unity, with few samples displaying P_{sat}/P_{sample} significantly <1.

A similar comparison of P_{sample} with P_{sat} predicted by other experimental solubility studies provides a rough approximation of the accuracy of these solubility models. The model of Papale et al. (2006) produces P_{sat}/P_{sample} ratios that are 60% lower on average compared with Dixon et al. (1995), and suggests that most MORB are vapor undersaturated. Vesicle abundances observed in MORB, however, contradict this prediction (Chavrit et al., 2012), indicating that the Papale et al. (2006) model likely underestimates vapor saturation pressures in MORB. The Papale et al. (2006) model was calibrated over a very wide range in melt compositions, whereas the Dixon et al. (1995) model was calibrated specifically on MORB

compositions at pressures <1 kbar, so disagreement between these models is perhaps not surprising. The solubility model of Iacono-Marziano et al. (2012) provides values of P_{sat} for MORB that are within 2% of those calculated from the Dixon et al. (1995) model. More recently, the CO_2-H_2O solubility model of Ghiorso and Gualda (2015) results in P_{sat}/P_{sample} ratios that are 20% lower on average compared with Dixon et al. (1995), with a peak of the P_{sat}/P_{sample} ratio distribution closer to unity (median 1.10) and a lower limit of the ratio of ~0.6. The Ghiorso and Gualda (2015) model thus also predicts more samples than Dixon et al. (1995) to be vapor undersaturated. Both models provide reasonable estimates of vapor saturation pressures, but we prefer the model of Dixon et al. (1995) because it is specific to MORB and it predicts that fewer MORBs are vapor undersaturated, which is consistent with the presence of vesicles in the vast majority of MORBs.

The ratios of volatile to nonvolatile elements have been useful to examine processes of loss or gain of volatile elements, such as degassing, dehydration, and shallow-level assimilation that do not affect nonvolatile elements that otherwise share similar mineral-melt partitioning behavior during partial melting and fractional crystallization. Further constraints on the extent of degassing of individual MORB samples can thus be obtained by examination of the ratios of CO_2/Rb and CO_2/Ba, which exhibit a narrow range in genuine vapor-undersaturated MORB glasses and melt inclusions. Rosenthal et al. (2015), Michael and Graham (2015), and Hauri et al. (2017) recently reviewed the data for vapor-undersaturated MORB; in this compilation we use average ratios CO_2/Rb (991 ± 26%) and CO_2/Ba (81.3 ± 56%) to represent undegassed MORBs, which are calculated as the average of the population averages for six groups of vapor-undersaturated samples given in Table 1 of Hauri et al. (2017). CO_2/Rb and CO_2/Ba ratios are much more uniform than CO_2/Nb or CO_2/Th ratios in undegassed MORBs. The ratio of the measured CO_2/Ba ratio in degassed MORBs to the average CO_2/Ba ratio for undegassed MORB gives a measure of the amount of CO_2 lost by degassing. The extent of degassing of typical MORB samples estimated in this way demonstrates that a majority of samples degassed >70% of their initial inventory of CO_2 (Figure 4). Approximately 3% of MORB samples, however, exhibit CO_2/Rb ratios close to the undegassed ratio, and most of these samples have been identified previously (Hauri et al., 2017; Le Voyer et al., 2017; Michael & Graham, 2015; Saal et al., 2002; Shimizu et al., 2016). Based on this analysis, emplacement of MORB on the seafloor typically results in eruptive degassing into the oceans of ≥70% of the primary carbon carried by MORB magmas.

The CO_2/Ba ratios of melt inclusions from mid-ocean ridges also bear importantly on the carbon budget of MORB magmas. The olivine-hosted melt inclusion data of Saal et al. (2002; Siqueiros FZ, East Pacific Rise), Le Voyer et al. (2017; equatorial mid-Atlantic Ridge), and Hauri et al. (2017; northern Iceland), for example, all display excellent correlations of CO_2 with Ba (and with Rb, where measured), demonstrating the undegassed nature of these melt inclusion populations. An additional 113 melt inclusions have been measured for CO_2 and Ba from the Gakkel Ridge (Shaw et al., 2010; Wanless et al., 2014), the Juan de Fuca Ridge (Helo et al., 2011; Wanless & Shaw, 2012), and from the Lucky Strike segment of the mid-Atlantic Ridge (Wanless et al., 2015). Of these 113 melt inclusions, 22 exhibit CO_2/Ba ratios within 20% of the average undegassed ratio of 81.3 (Figure 4b). The majority have likely experienced CO_2 degassing either prior to melt inclusion entrapment or into secondary vapor bubbles that often form within melt inclusions during post-entrapment cooling (e.g., Moore et al., 2015) but are difficult to measure quantitatively. More data could be readily obtained from MORB melt inclusions through concentrated efforts to quantify the effect of vapor bubbles within melt inclusions (e.g., Aster et al., 2016) and by targeting ridge segments that erupt samples on the seafloor that have experienced little to no degassing (e.g., Le Voyer et al., 2017).

Matthews et al. (2017) present an alternative interpretation of certain suites of MORBs with CO_2 that is well correlated with highly incompatible trace elements. Such suites are often interpreted as undegassed with respect to CO_2, because the degassing process fractionates CO_2 from other highly incompatible elements. Matthews et al. (2017) instead envisages a process by which magmas emerging from the melting regime stall at a depth where enriched magma batches become oversaturated in CO_2, leading to degassing. This degassing is followed by limited mixing at shallower levels, which can produce correlated CO_2/Ba ratios in MORBs that are lower than the original mantle source CO_2/Ba ratio. Yet this process is limited in its ability to explain CO_2/Ba ratios in highly depleted MORB, many of which arrive on the seafloor with low CO_2 contents but also

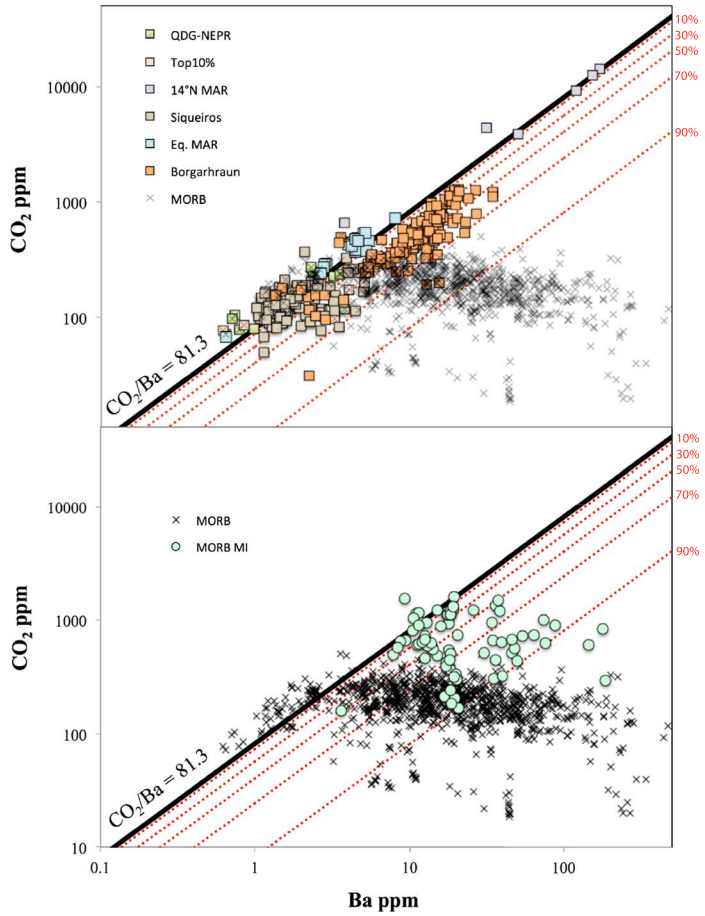

Figure 4. (a) CO_2 versus Ba for suites of vapor-undersaturated melt inclusions and mid-ocean ridge basalt (MORB; squares, summarized in Hauri et al., 2017) together with data from globally distributed MORB (crosses). (b) Same diagram as panel (a) showing only MORB glasses and MORB melt inclusions from Shaw et al. (2010), Wanless & Shaw (2012), Helo et al. (2011), and Wanless et al. (2014, 2015). The heavy black line represents the average CO_2/Ba (81.3) derived from the CO_2/Ba ratio of undersaturated samples. Red dashed lines with labels indicate the extent of CO_2 degassing.

vapor undersaturated. Such magmas would need to degas CO_2 at pressures significantly lower than their seafloor collection pressures (e.g., Siquerios data of Saal et al. (2002); Quebrada-Discovery-Gofar Transform data of Shimizu et al. (2016); the vapor-undersaturated data of Michael and Graham (2015)), or at pressures within the eruptive Layer 2 of the oceanic crust (equatorial MAR data of Le Voyer et al. (2017)). For the more intermediate-enriched data sets (Borgarhraun, Iceland, and Top 10% MORB data sets of Hauri et al., 2017; 14°N MAR, Cartigny et al., 2008) the highest CO_2 contents vary widely, and each sample suite would need to have stalled and degassed at a very specific depth (e.g., not just the Moho depth) in order to produce such similar CO_2/Ba ratios as reported in Hauri et al. (2017). We thus conclude that, although the processes in Matthews et al. (2017) are plausible, they are unlikely to have operated in the samples that currently constrain the CO_2/Ba ratio of MORB sources.

6. Variations in Primary MORB CO_2 Contents

The restricted ranges in CO_2/Rb and CO_2/Ba ratios of undegassed MORB permit an estimation of the initial (undegassed) CO_2 concentration in MORB primary magmas, given an estimate of their primary Rb and/or Ba abundances. The studies of Gale et al. (2013, 2014) provide such estimates based on the compositions of MORB magmas averaged by ridge segment, and corrected for shallow-level crystal fractionation to MgO = 8 wt% and also further corrected to be in equilibrium with Fo_{90} olivine. The data compilations of

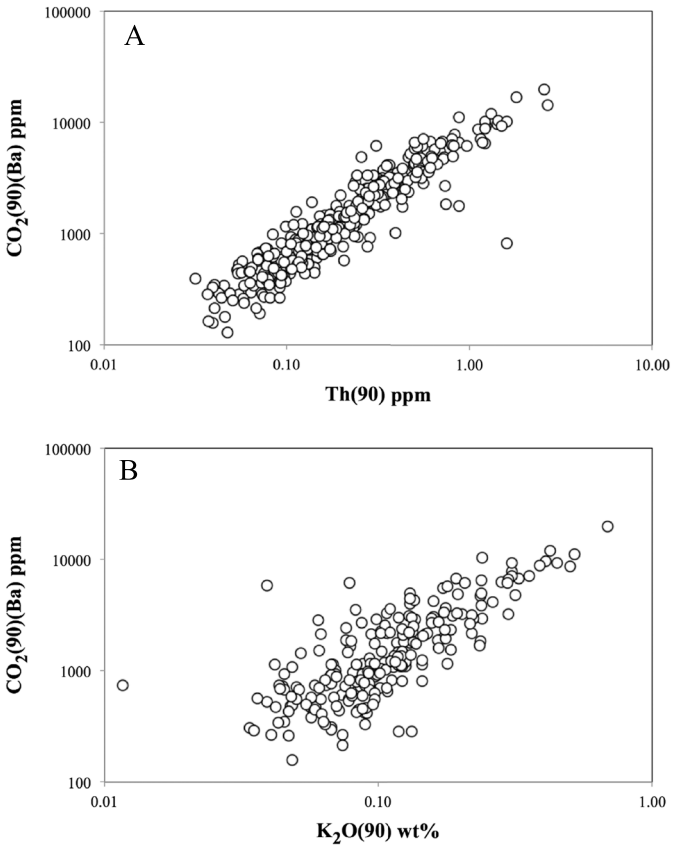

Figure 5. Segment-averaged estimated primary CO_2 versus (a) segment-averaged primary Th and (b) segment-averaged primary K_2O for global mid-ocean ridge basalt; CO_2 is estimated from the segment-averaged Ba content multiplied by a constant CO_2/Ba (81.3).

Gale et al. (2014) and this study provide primary MORB magma estimates for 403 segments that contain data for Ba and/or Rb, out of the 711 global spreading ridge segments identified by Gale et al. (2013). These segment-averaged concentrations were then multiplied by the undegassed-average ratios of CO_2/Rb (991) and CO_2/Ba (81.3) to calculate segment-averaged primary CO_2 concentrations. MORB primary CO_2 thus calculated varies over a factor of 180, from 104 ppm to 1.90 wt%. These calculated primary CO_2 contents further enable calculation of CO_2/Nb and CO_2/Th ratios for all segments that have segment-averaged Nb and Th data. These latter ratios are used to compare with prior studies that used CO_2/Nb ratios (e.g., Cartigny et al., 2008; Saal et al., 2002), while Th is another highly incompatible element that has a mantle melt partition coefficient similar to CO_2 (Rosenthal et al., 2015), and there exists Th data of sufficient quality in MORB and MORB melt inclusions to make a useful comparison (Hauri et al., 2017).

Primary MORB CO_2 abundances can be compared with other indices of depletion and enrichment in MORB such as the K_2O/TiO_2 ratio, which is low in depleted MORB and high in enriched MORB. The lowest calculated primary MORB CO_2 abundance is 104 ppm CO_2 at a depleted segment of the Galapagos Spreading Center (segment GALA16 with K_2O/TiO_2 of 0.05), and the highest is 1.90 wt% CO_2 at a highly enriched segment of the Southwest Indian Ridge (segment SWIR9 with K_2O/TiO_2 = 0.66). The distribution of MORB primary CO_2 contents is log-normal with a median value of 2,100 ppm. In general, the segment-averaged primary CO_2 contents, as derived from reconstructed primary Ba and Rb concentrations, correlate well with segment-averaged primary Th and K_2O contents (Figure 5). Such correlations are most reasonably interpreted as signatures of variation in mantle source composition, rather than variable mantle temperature or spreading rate. In this view, variations in the primary MORB CO_2 contents determined here would also reflect mantle source variations.

Segment-averaged CO_2/Nb ratios calculated above vary by more than a factor of 10, from a low of 152 at the very depleted segment GALA16 on the Galapagos Spreading Center, to a high of 1,760 at enriched segment SWIR22 on the Southwest Indian Ridge. This range in CO_2/Nb contrasts with the much more limited range in CO_2/Nb measured in undegassed MORB glasses or melt inclusion populations (230–730, Cartigny et al., 2008; Hauri et al., 2017; Saal et al., 2002). We emphasize again that the CO_2/Nb ratios constrained here effectively reflect variations in Ba/Nb or Rb/Nb in MORB because of the assumptions adopted in the calculation of CO_2 concentrations. Yet the calculated segment-averaged CO_2/Nb ratios display rough trends with segment-averaged Sr and Nd isotopes (Figure 6) that are consistent with the relationships observed by Hauri et al. (2017) and further support the inference that primary CO_2 contents of MORB, inasmuch as these are inseparable from Ba and Rb, are a function of mantle composition. It remains to be seen if this large range in estimated CO_2/Nb ratios is reflected in actual sample measurements, but the data in this study provide a first-order prediction and help identify specific ridge segments to focus on to test the accuracy of these estimates. The variations are consistent with the dichotomy of CO_2/Nb and CO_2/Th ratios between Atlantic and Pacific Ocean basins, which could be a result of a greater amount of recycled subduction zone components in the Atlantic Ocean basin compared with the relatively older Pacific Ocean basin (Dixon et al., 2017; Hauri et al., 2017).

7. The Flux of CO_2 From the Global Ridge System

Crustal thickness at mid-ocean ridges is known to vary from a few kilometers to over 20 km at northern Iceland (Menke, 1999), controlled by a combination of the local magma production rate and the spreading rate. The study of Behn and Grove (2015) summarized seismically determined measurements of crustal thickness at ridge axes, where it varies from 2 km at the slow-spreading Gakkel Ridge to 30 km at central Iceland; additional data for crust <20 Ma old was taken from Van Avendonk et al. (2017). Seismically

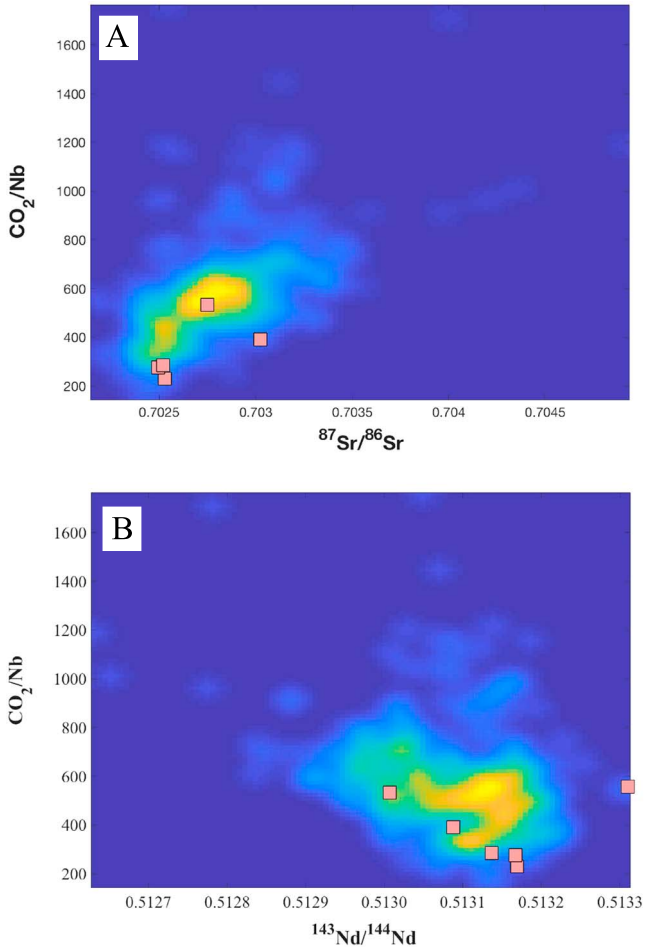

Figure 6. Heat map diagram showing relationship between segment-averaged calculated mid-ocean ridge basalt CO_2/Nb ratios and (a) Sr isotopes and (b) Nd isotopes. CO_2/Nb ratios were calculated using segment-averaged CO_2(8) contents estimated from segment-averaged Ba(8) (Gale et al., 2014) multiplied by CO_2/Ba of 81.3; this estimated primary CO_2 content is divided by segment-averaged Nb(8) to calculate primary CO_2/Nb. Yellow colors indicate increasing density of data points. Isotope ratios are segment averages from Gale et al. (2013). The pink squares are the average values for six populations of vapor-undersaturated glasses and melt inclusions summarized by Hauri et al. (2017).

determined crustal thickness correlates well with mean ridge depth (Figure 7), reflecting isostatic compensation of MORB crust at the ridge. These constraints permit an estimation of the crustal thickness at all of the 711 segments defined by Gale et al. (2013) based on the axial depth. We then calculate a segment-averaged magma production rate by multiplying the crustal thickness by the mean segment spreading rate, which we then use to calculate the fluxes of CO_2 (and other elements) on a segment-by-segment basis. There is one exception to the relationship between ridge depth and crustal thickness at the northernmost three segments of the Alarcon Ridge, where the ridge crest shallows as it begins to intersect the incipient continental rift in the Gulf of California; here we have chosen to assign an average crustal thickness of 6.4 km to these three ridge segments (EPRR1, EPRR2, and EPRR3).

Carbon dioxide fluxes, normalized by ridge length, vary by a factor of >100 because they are the products of local primary magma CO_2 contents and local magma production rates, which are uncorrelated. Fluxes of CO_2 vary from a low of 1.52×10^6 mol/year/km at segment SWIR17 (which is both depleted and slow-spreading) to a high of 4.74×10^8 mol/year/km at segment JUAN1 (which has high primary CO_2 and moderate spreading rate). High CO_2 fluxes are independent of spreading rate, occurring at both slow- and fast-spreading ridge segments. High CO_2 fluxes also occur on segments both near to and far from hotspots, and there is a clear tendency for estimated CO_2 fluxes to increase toward hotspots (Figure 8), in concert with enrichment in highly incompatible elements (i.e., Ba, Rb). Provided that the mechanism of mantle enrichment for hotspots does not fractionate CO_2 from Ba or Rb, then this observation supports our perspective that variations in the MORB primary magma CO_2 content are mostly driven by variations in mantle composition rather than magma production dynamics or mantle temperature.

When summed over the length of the global mid-ocean ridge system, we calculate the global MORB magma production rate to be 16.5 km^3/year, significantly lower than the widely cited 21 km^3/year estimated by Crisp (1984); this difference in magma production rate alone accounts for 27% of our total flux calculation. The total flux of CO_2, summed over the widely varying fluxes at individual ridge segments (Figure 9), is $1.32^{+0.77}_{-0.85} \times 10^{12}$ mol/year. Importantly, for individual ridge segments with no Ba or Rb data from which to constrain CO2, we assume an average magmatic CO2 content calculated from the log-normal mean of those segments with Ba or Rb constraints. This estimate lies within the range of CO_2 fluxes estimated by Resing et al. (2004) from CO_2 and ^{3}He fluxes into the water column above ridges, and by Barry et al. (2014) in their study of Iceland CO_2 fluxes. Our estimate overlaps the lower average flux found by Saal et al. (2002; $0.93 \pm 0.28 \times 10^{12}$ mol/year), who based their estimate on the CO_2 content of the depleted Siqueiros Fracture Zone melt inclusions. Our estimate is lower than the estimates of Marty and Tolstikhin (1998), Cartigny et al. (2008), Michael and Graham (2015), Le Voyer et al. (2017), and Hauri et al. (2017) based on a variety of approaches, but these prior studies utilize much more limited data sets than we have employed here. Our estimate is substantially higher than the study of Chavrit et al. (2014) who considered vesicle abundances in MORB; their underestimation is likely biased by CO_2 contained in vesicles that were lost from samples by degassing. In general there is a good correlation of segment-scale CO_2 flux with the segment-averaged primary CO_2 content (Figure 9), while the correlation of CO_2 flux with magma flux is much weaker.

The MORB flux of CO_2 is nearly quantitatively delivered to the overlying water column, during eruptive degassing and later by hydrothermal circulation, where it becomes part of the marine bicarbonate cycle. Short-term global effects of large variations in the ridge CO_2 flux are, however, muted by the ~100,000-

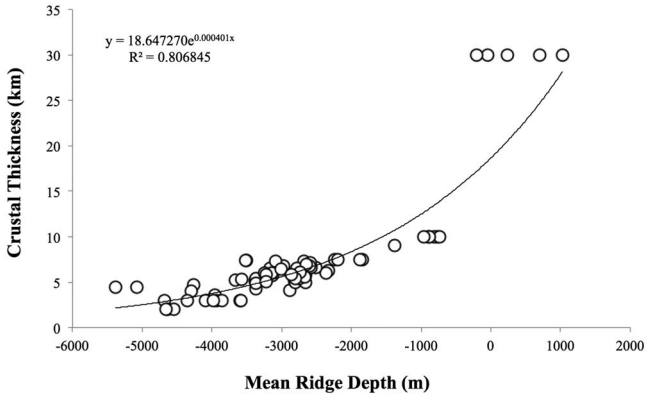

Figure 7. Correlation between seismically determined ridge crest crustal thickness (Behn & Grove, 2015; Van Avendonk et al., 2017) and ridge depth (Gale et al., 2013). This relationship allows for an estimation of crustal thickness at each ridge segment.

year residence time of carbon dissolved in seawater (Broecker & Peng, 1986) and thus variations in the ridge CO_2 flux are likely to influence short-term surface climate conditions only if large fluxes bypass the water column in areas where the ridge is shallow or exposed above sea level. On the longer timescale, the ridge CO_2 flux is dwarfed by the deposition of carbonate sediments and organic carbon on the seafloor and formation of carbonate alteration veins within the crustal and mantle sections of the oceanic lithosphere, making the seafloor a net sink for carbon by a large factor (Alt & Shanks, 2003; Alt & Teagle, 1999; Hayes & Waldbauer, 2006; Kelemen & Manning, 2015). Much (but not all) of this carbon is present in forms, and at locations, where it can be returned to the deep crust and/or mantle in subduction zones.

8. Influence of Carbon on Mantle Melting

Several studies have demonstrated that the presence of carbonate in the mantle can strongly influence the depth of melt initiation beneath ridges (Dasgupta & Hirschmann, 2006, 2007; Dasgupta et al., 2013; Keller et al., 2017; Stagno et al., 2013), as incipient melting of carbonate may represent the first step in a polybaric process that ultimately results in eruption of MORB. To first order, the 2-order-of-magnitude range in primary MORB CO_2 content must reflect a large variation in the carbon content of the MORB source mantle. Le Voyer et al. (2017) hypothesized how changes in the carbon content of the mantle could result in geophysical observations illuminating the depth beneath ridges where low, but possibly interconnected, melt fractions (near 0.05%; Minarik & Watson, 1995) could first be detected at constant potential temperature. Specifically, they showed that changing the CO_2 content of relatively oxidized and anhydrous mantle from 20 ppm to 140 ppm increased the depth where interconnected melt (F~ 0.05%) would exist from 75 km to 275 km. If a combination of seafloor seismic and electromagnetic methods could elucidate clearly such low melt fractions (Eilon & Abers, 2017; Gaillard et al., 2008; Sifré et al., 2014), then a test of this hypothesis might be possible by comparing the geophysical signals at ridge segments characterized by widely different primary magma CO_2 contents as identified here.

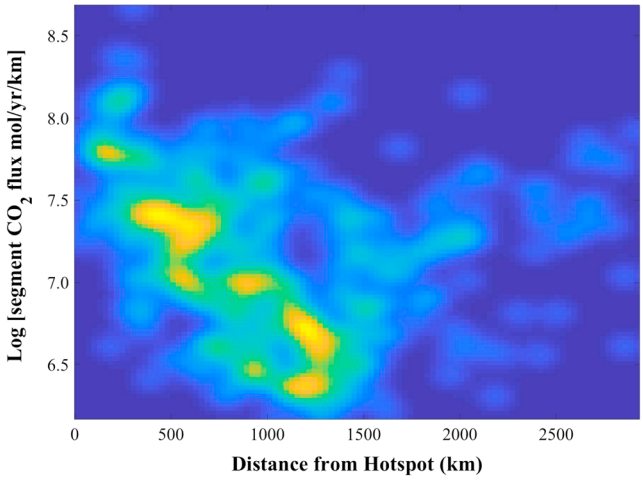

Figure 8. Heat map diagram of segment-averaged CO_2 flux (log scale, mol/year/km) versus distance to nearest hotspot (Gale et al., 2013); brighter color (yellow) scales with increasing density of data points. Individual segments with high primary CO_2 contents are observed both near to and far from hotspots, but the data density suggest an increase of CO_2 approaching hotspots at distances $<$1,400 km from the nearest hotspot.

This test is complicated, however, if carbonate is not the stable phase of carbon in the mantle at the depth of incipient melting. While direct analyses of MORBs and their peridotitic residues suggest that mantle oxygen fugacity (fO_2) is near the quartz-fayalite-magnetite buffer, where carbonate would be the stable phase (Bezos & Humler, 2005; Cottrell & Kelley, 2011; Birner et al., 2018), oxygen fugacity (fO_2) may decrease with depth in the convecting oceanic mantle, as observed in the cratonic xenolith record and supported by theory and experiment (Eggler & Baker, 1982; O'Neill et al., 1993; Ballhaus, 1995; Frost & McCammon, 2008; Rohrbach et al., 2007; Stagno and Frost, 2010; Stagno et al., 2013). It has been suggested that this decrease in mantle fO_2 would reduce carbonate to graphite, diamond, or metal alloy, and this would prevent carbon from fluxing the mantle. In this scenario, the existence of an interconnected melt network would not occur until upwelling mantle became oxidized enough to stabilize carbonate (~ QFM-2) or reached the onset of silicate melting, (e.g., Stagno et al., 2013). However, even in the limit where the mantle is too reduced to stabilize carbonate, carbon would dissolve in silicate melts and become exhausted within the first 1–2% of melting for typical carbon concentrations that characterize the MORB mantle (20–200 ppm C; Le Voyer et al., 2017; Rosenthal et al., 2015; Saal et al., 2002). Thus, whether the mantle is reduced or oxidized, and regardless of whether carbon actively fluxes mantle melting or not, we expect carbon to behave like a highly incompatible element and produce the correlations

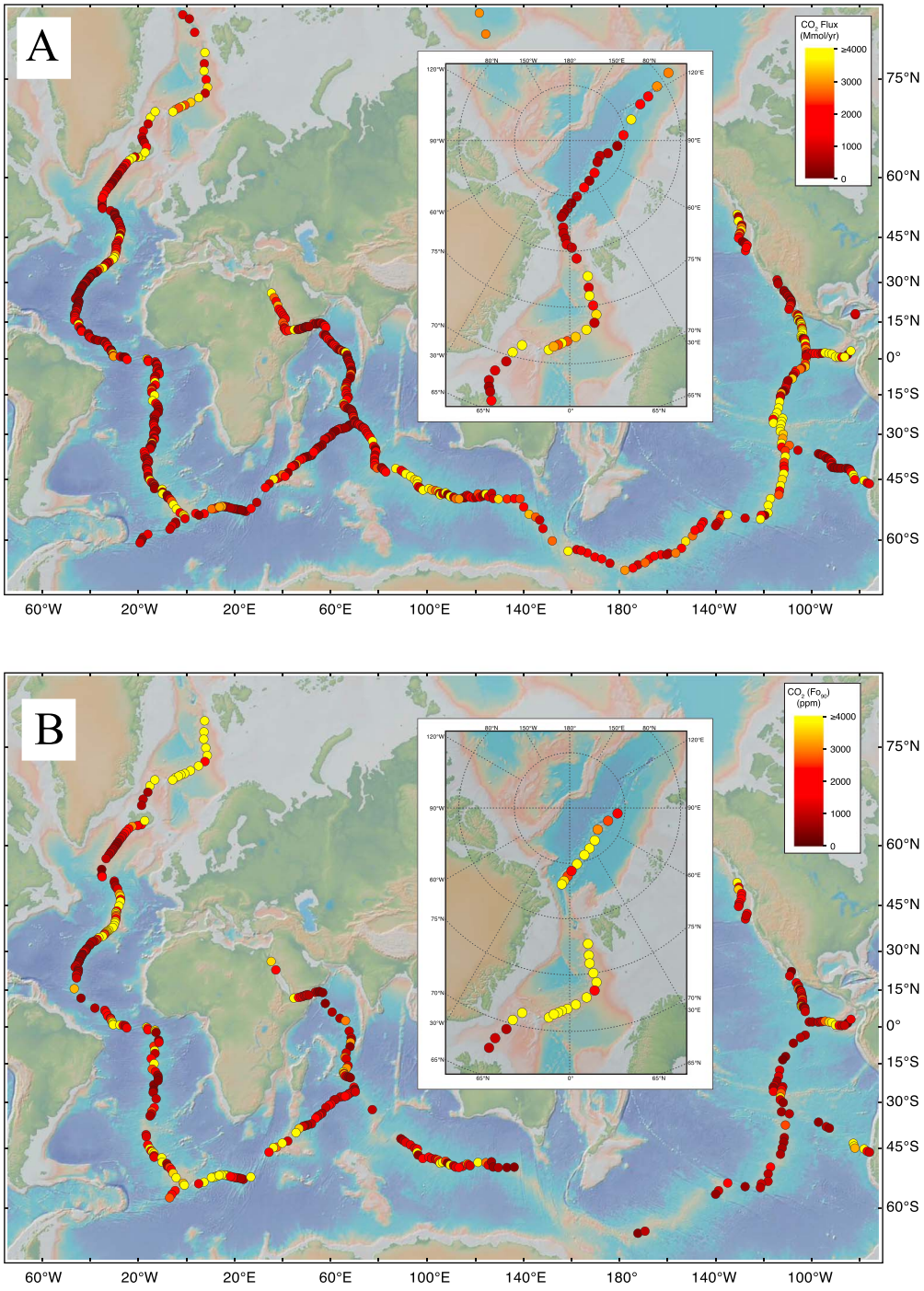

Figure 9. (a) Map of segment-averaged CO_2 fluxes, and (b) estimated primary magma CO_2 concentrations along the global mid-ocean ridge system. Both quantities are derived from segment-averaged Ba and Rb abundances and average CO_2/Ba and CO_2/Rb ratios (see text). For segments without Ba or Rb data, CO_2 fluxes in (a) were calculated from the global average CO_2 flux normalized by ridge length (mol/year/km) derived from segments with Ba or Rb data, multiplied by segment length, spreading rate, and crustal thickness (estimated from the relationship in Figure 7).

of C with Ba, Rb, Th, and Nb that characterize suites of undegassed melt inclusions for typical mantle carbon concentrations (cf. Hauri et al., 2017). That we observe carbon's incompatible behavior up to much higher carbon concentrations supports other inferences, based on MORB and peridotite analyses, that graphite is not stable at shallow depths beneath the ridge.

9. Conclusions

We have determined the concentration of CO_2 and other volatiles in 753 MORB glasses by SIMS from all the major ocean basins and, when combined with data from prior studies, these data provide global coverage of carbon fluxes from mid-ocean ridges. Assuming constant ratios of CO_2/Ba and CO_2/Rb, we estimate primary magma CO_2 concentrations that vary from 104 ppm to 1.90 wt%, MORB CO_2 fluxes that range from 3.3×10^7 to 4.0×10^{10} mol/year at the scale of individual ridge segments, and a global integrated CO_2 flux of $1.32^{+0.77}_{-0.85} \times 10^{12}$ mol/year, overlapping the global CO_2 flux estimated from depleted MORB ($0.93 \pm 0.28 \times 10^{12}$ mol/year, Saal et al., 2002) but considerably lower than the flux estimated from E-MORB popping rock at 14°N on the mid-Atlantic Ridge (15×10^{12} mol/year, Javoy & Pineau, 1991). The accuracy of the CO_2 fluxes determined here is due to consideration of the heterogeneity of trace element abundances in MORB along the global ridge system, with the assumption that our global CO_2/Ba and CO_2/Rb ratios are both representative and homogeneous in the MORB mantle.

The primary magma abundances, and segment-scale fluxes, of mid-ocean ridge CO_2 are heterogeneously distributed both along the ridge system and between major ocean basins. Areas of both high flux and high primary CO_2 are predicted at hotspots (Iceland, Azores, Bouvet, Afar, Reunion, Easter) and regionally along the Gakkel Ridge (Arctic). On the East Pacific Rise, these fluxes are correlated with deep areas of elevated mantle temperatures (Dalton et al., 2014) and high magma production rates. High primary CO_2 contents are, however, mainly predicted for areas of enriched mantle composition, which may be associated with long-term enrichment of the mantle as measured by radiogenic isotopes.

Large variations in primary MORB CO_2 imply similar large variations in mantle carbon content beneath ridges. If the upper mantle is sufficiently oxidized, these variations would result in large changes in the depth at which interconnected melt (F ~ 0.05%) would exist (from 75 to 275 km). Such large depth differences might be resolvable using geophysical methods by comparing ridge segments characterized by vastly different CO_2 contents as we have described here.

Acknowledgments

We would like to dedicate this paper to our colleague and mentor, Erik Hauri. As co-authors, it was both an honor and a challenge to step into his shoes in order to finish this work after his passing. With a passion for scientific achievement, an always positive attitude, and an everlasting care for peers, Erik was a cornerstone of the Deep Carbon Observatory Initiative. His legacy work on carbon in the mantle will inspire the next generation of scientists to press farther. Many thanks to Jianhua Wang for expert assistance with temperamental instrumentation in the Carnegie SIMS lab, and to Tim Gooding and the Smithsonian Department of Mineral Sciences for assistance with sample preparation. We thank the geochemistry group at IPGP, University of Paris for sharing data and samples from the Pacific-Antarctic Ridge. We wish to thank the captain, crew, and scientific parties of the cruises PACANTARCTIC 1 and 2 cruises (PAR), the GIMNAUT, CD127, and KNOX11RR cruises (CIR), the PROTEA-5, MD34, AG22, and AG53 (SWIR), and the CHEPR and PANORAMA-1 cruises (EPR). We also acknowledge support from the U. California Ship Funds, and NSF grant OCE-0726573 (DRH). National Science Foundation grant OCE-1555523 provides support for the curation and distribution of geological samples at the University of Rhode Island. This is a contribution to the Deep Carbon Observatory.

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