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## Mapping modern CO<sub>2</sub> fluxes and mantle carbon content all along the mid-ocean ridge system



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#### ABSTRACT

Ouality criteria have been used to select ~400 vesicularity measurements on zero-age mid-ocean ridge glasses from  $\sim$ 600 data available in the literature published over the past  $\sim$ 30 years. At face value, observations show that for a given depth of sampling, enriched basalts from slow spreading ridge segments are more vesicular than those from depleted and intermediate or fast spreading ridges. A shallower depth of eruption enhances these effects because lower hydrostatic pressure favours bubble expansion. In order to get an insight into these complex and intermingled processes, we used empirical and semi-quantitative approaches based on a limited number of inputs (segment depth D, spreading rate  $\tau$  and K<sub>2</sub>O/TiO<sub>2</sub> ratios). Both models give equivalent results and predict vesicularities within  $\pm 50\%$ . From these calculations, we compute the equivalent CO2 concentration at the depth of eruption all along the oceanic ridge system. The total calculated  $CO_2$  fluxes are low ranging from  $6.5\pm1.8$  to  $8.7 \pm 2.8 \times 10^{11}$  mol/yr and the CO<sub>2</sub> mantle content displays large variabilities from  $66^{+27}_{-19}$  to  $78^{+82}_{-40}$  ppm. In order to test these results, the mantle <sup>3</sup>He fluxes have been evaluated using the calculated CO<sub>2</sub> fluxes and a  $CO_2/^3$ He ratio of  $2.2 \times 10^9$ . These fluxes range from  $295 \pm 82$  to  $395 \pm 127$  mol/yr and are close to the values reported by Jean-Baptiste (1992) (267-534 mol/yr) and the most recent estimate (Bianchi et al., 2010,  $\sim 527 \pm 102$  mol/yr) using box-model of the three main ocean basins constrained by measurements of <sup>3</sup>He and radiocarbon data. As these independent methods give similar helium fluxes at regional and global scales, it provides strong support to a low and heterogeneous mantle carbon concentration and distribution. Finally, the calculated volcanic CO<sub>2</sub> emissions at oceanic ridges correspond to  $\sim$ 30 seconds of anthropogenic emissions, at current rates.

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#### 1. Introduction

The variation in mid-ocean ridge basalt (MORB) vesicularity can be regarded as a measure of CO<sub>2</sub> content of the rock in excess of saturation as CO<sub>2</sub> is the predominant gas in vesicles at the eruption depth. However, the control of the vesicularity is complex because of the transient nature of gas bubbles in melt and the many processes which can operate to enrich, remove or disperse vesicles. It is generally believed that the primary control on the vesicularity of subaqueously erupted basalts is the depth of eruption (Moore, 1979). Moreover, basalt composition also affects the vesicularity of submarine basalts. In addition to these vesicularity controls, variations have been noted between tholeiitic basalts from different mid-ocean ridges which seem to be independent

*E-mail addresses*: Deborah.chavrit@manchester.ac.uk (D. Chavrit), Eric.humler@univ-nantes.fr (E. Humler), Olivier.Grasset@univ-nantes.fr (O. Grasset). of depth or major and trace element chemistry. The solubility of carbon in basaltic melts is very low (Stolper and Holloway, 1988; Dixon et al., 1995). Consequently, most erupted basalts have lost almost all their CO<sub>2</sub> content by degassing prior to the eruption (e.g. Sarda and Graham, 1990; Burnard, 1999; Burnard et al., 2002; Aubaud et al., 2004), which the amount lost is still debated. For this reason, it is not possible to estimate the carbon flux from ridges using the dissolved carbon content of the recovered basalts.

The higher vesicularity of Atlantic spreading ridge basalt suites compared with those from the Pacific (Moore, 1979; Chavrit et al., 2012) suggests that some features of the shallow storage chamber feeding eruption, which have been recorded along the Pacific ridges, may control the vesicularity of erupted lavas (Moore, 1979). However, the presence of a crustal magma chamber beneath the slow spreading Lucky Strike segment of the Mid-Atlantic Ridge (MAR) has been reported (Singh et al., 2006). Chavrit et al. (2012) have shown that samples from the Lucky Strike segment are much more vesicular (~2.70%) than Pacific samples (~0.09%) and thus, the simple rule relating the presence of magma chambers to low vesicularity basalts is questionable. Other mechanisms

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**Table 1**Compilation of the mid-ocean ridge (MOR) CO<sub>2</sub> fluxes.

	Mantle CO <sub>2</sub> (ppm)	$CO_2$ flux $\times 10^{12}$ (mol/yr)
Undegassed MORB		
Javoy and Pineau (1991)	1000	13.4
Saal et al. (2002)	72	1.0
Shaw et al. (2010)	134	1.8
Helo et al. (2011)	1200	16.0
Other methods		
Cartigny et al. (2008)	180	2.3
Marty and Jambon (1987),	164	2.2
Marty and Tolstikhin (1998)		
Hirschmann and Dasgupta (2009)	60	0.8

The  $CO_2$  fluxes have been recalculated using an annual oceanic crust production of 21 km<sup>3</sup>/yr (Crisp, 1984) and a partial melting rate of 10%.

need to be considered, such as the chemical composition of the samples and/or the magma ascent rate (Chavrit et al., 2012).

In order to circumvent the problem of CO<sub>2</sub> degassing from the magma, a variety of methods have been employed to estimate the CO2 flux at mid-ocean ridges (MOR) (Table 1). Early estimates of the mantle  $CO_2$  content ( $\sim 1000$  ppm of  $CO_2$ ) were based upon  $CO_2$ concentrations measured in the so-called "popping-rock" (Javoy and Pineau, 1991). This highly vesicular MORB glass from 14° N on the MAR, which has unfractionated noble gas ratios compared to the mantle ratio, suggests minimal degassing (Javoy and Pineau, 1991; Moreira et al., 1998). Subsequent studies (Saal et al., 2002; Shaw et al., 2010; Helo et al., 2011) derived mantle CO<sub>2</sub> contents by coupling CO<sub>2</sub>/Nb ratios of undegassed melt inclusions, assuming that CO<sub>2</sub> and Nb have similar partitioning behaviour and using the mean MORB Nb contents. Saal et al. (2002) measured CO<sub>2</sub>/Nb ratios of CO<sub>2</sub> undersaturated olivine-hosted melt inclusions from Siqueiros intra-transform spreading centre, providing an estimate of 72 ppm CO2 in MORB mantle source. The highest CO2/Nb ratio (443) obtained in melt inclusions of the Gakkel ridge is most representative of the undegassed pooled melt composition and predicts a mantle CO<sub>2</sub> content of 134 ppm (Shaw et al., 2010). Recently, Helo et al. (2011) measured CO2 in basaltic melts trapped in plagioclase crystals from volcanic ash deposits erupted at Axial Seamount on the Juan de Fuca Ridge. Adopting a CO2/Nb ratio of ~4000 from the least degassed melt inclusion provides a CO<sub>2</sub> content of 1200 ppm for the mantle source.

Other studies used various methods to reconstruct the initial CO<sub>2</sub> content of the magmas before degassing, using carbon isotopes (e.g. Cartigny et al., 2008), the CO<sub>2</sub>/He ratio (Marty and Jambon, 1987; Marty and Tolstikhin, 1998) or the H/C ratios (Hirschmann and Dasgupta, 2009). Cartigny et al. (2008) reassessed the CO<sub>2</sub>/Nb ratio of the popping-rock and found a significantly higher CO<sub>2</sub>/Nb ratios ( $\sim$ 530) than those reported by Saal et al. (2002) ( $\sim$ 239) which implies a CO<sub>2</sub> content in the MORB mantle source of  $\sim$ 180 ppm. Note that based on the H/C ratios, Hirschmann and Dasgupta (2009) calculated the lowest CO<sub>2</sub> flux. Using the global oceanic production rate of 21 km<sup>3</sup>/yr (Crisp, 1984) and assuming a 10% melting rate of the mantle source, the global CO<sub>2</sub> flux at MOR ranges from 0.8 to 16  $\times$  10<sup>12</sup> mol/yr (Table 1).

In the following, we propose a new method to estimate the  $CO_2$  flux released at MOR. This method is based on the vesicularity of the samples, which reflects the  $CO_2$  degassing in MOR context. Two approaches are used to infer the relationships between basalt vesicularities, the depth of eruption, the spreading rate and the compositional variations ( $K_2O/TiO_2$  ratio). Both approaches are based on 397 samples from the literature. Then, using the MORB  $CO_2$  contents calculated from the obtained vesicularities and the MORB productivity, we determine the  $CO_2$  flux at segment and global scales. It also allows us to infer the carbon content of the mantle all along the mid-ocean ridge system.

#### 2. Selected data

Vesicularity data are sometimes visually estimated or analysed using fast methods on small sample areas. If this information is useful to describe the general characteristics of the dredged samples, it cannot be used confidently to compare samples from different studies. Furthermore, the recent image processing methods (i.e. Chavrit et al., 2012; Soule et al., 2012) and micro-tomography (Colin et al., 2013) are *a priori* much more precise than the point counting (i.e. Moore, 1979) and weighting methods (i.e. Sarda and Graham, 1990). Here, we have screened an initial data set of  $\sim$ 600 measurements (Supplementary material Table A), based on the methods used and their description, the number of counted bubbles, the number of counted points, the uncertainties and the reproducibilities of the measurements. For that, we compared the different methods and data obtained between authors (Supplementary material Note A):

- We first compared vesicularities obtained using point counting and using image processing methods. The results show an excellent agreement between both methods, provided more than 2000 points are counted for vesicularities lower than 0.2%.
- By processing 3 times the image mosaic of sample ED DR08 type 2 (Chavrit et al., 2012), we determined an analytical reproducibility of 12% for the image processing method.
- We also assessed possible inter-laboratory biases: generally, vesicularities can display high variabilities between authors, except when vesicularities are obtained using image processing.

Based on those results, the  $\sim$  600 data were sorted into five quality classes (1 to 5 from high to low quality classes). The highest quality classes (1 to 3) are data obtained using image processing (1 and 2) and point counting. Those high quality data have been selected and reported in a worldwide synthesis of 397 vesicularity measurements on MORB (Supplementary material Table A).

#### 3. Vesicularity variability at different scales

#### 3.1. Variability at sample and dredge scales

Five pillow lavas have been selected to test vesicularity variations within a single sample (Supplementary material Fig. A). Between 2 and 5 pieces of glass were extracted from each pillow corresponding to surface areas ranging from 0.44 to 5.01 cm² and vesicularities from 0.12 to 3.05%. Vesicularity variations expressed as the relative standard deviation ( $\sigma\% = 100 \times \sigma/\text{mean}$ ) range from 13% to 63%. The three samples characterised by  $\sigma\%$  larger than 57% are linked to fragments of glass containing few isolated large bubbles. At the dredge scale, Indian samples SWF DR05-1-2g and SWF DR05-GISt° which have similar chemical composition vary by 37%. Dixon et al. (1988) identified similar variabilities at sample scale from Juan de Fuca in the Pacific Ocean. Soule et al. (2012) measured the vesicularity in MORB coming from the same lava flow at 9–10° N on the East Pacific Rise (EPR). The vesicularity they measured on 19 samples varies by 60%.

#### 3.2. Variability at segment scale

Moore (1979) defined 13 segments based on average data (ranging from 5 to 62 samples per site) in the Pacific and Atlantic oceans (4 and 9 sites respectively). He showed that the volume percent vesicles at segment scale vary widely (from 0.25 to 14.2%). The data presented in cumulative histograms show rough lognormal distributions. To first order the slopes in those histograms give an estimation of the standard deviation for the vesicularity

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