



# Reconstructing mantle carbon and noble gas contents from degassed mid-ocean ridge basalts



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## ABSTRACT

The fluxes of volatile elements from the mantle have long been used to understand mantle structure and evolution, and are critical controls on Earth's climate stability. Because of the ubiquity of magmatic degassing, inferring pre-degassing volatile concentrations from measured basalts requires the application of a degassing model. Such models, including the commonly-applied equilibrium Rayleigh distillation, typically assume equilibrium or solubility-based partitioning between melt and vapor. Here, we demonstrate that ratios of radiogenic isotopes of He, Ne, Ar and, especially, Xe measured in global mid-ocean ridge basalts (MORBs) are inconsistent with equilibrium degassing models, even when not considering He. We conclude that kinetic disequilibrium is a crucial process affecting volatile abundances during degassing. We present a simple disequilibrium Rayleigh distillation model to reconstruct pre-degassing MORB noble gas and carbon concentrations, which predicts that He and Ne achieve nearly equilibrium partitioning between melt and vesicles, but the slower-diffusing heavier noble gases are strongly affected by disequilibrium, resulting in non-equilibrium fractionation of noble gas elemental ratios, and other volatile element ratios like CO<sub>2</sub>/He.

We apply our model to a large set of MORB data, and find average pre-degassing <sup>3</sup>He, <sup>22</sup>Ne, and <sup>36</sup>Ar concentrations of  $4.4 \pm 0.9 \times 10^{-10}$ ,  $6.6 \pm 1.4 \times 10^{-11}$ , and  $6.8 \pm 4.5 \times 10^{-10}$  ccSTP/g ( $2\sigma$ ), with variations of approximately 2 orders of magnitude, similar to other highly incompatible elements. Pre-degassing noble gas concentrations imply a mid-ocean ridge <sup>3</sup>He flux of  $800 \pm 170$  mol/yr and upper mantle <sup>3</sup>He/<sup>22</sup>Ne and <sup>3</sup>He/<sup>36</sup>Ar ratios of  $6.6 \pm 2.0$  and  $0.64 \pm 0.44$ , but substantial variability in these ratios between samples. Applying our model to CO<sub>2</sub>, we calculate an average mantle CO<sub>2</sub>/<sup>3</sup>He molar ratio of  $1.67 \pm 0.21 \times 10^9$ , which, when combined with our estimate of <sup>3</sup>He flux, implies an upper mantle CO<sub>2</sub> flux of  $5.9 \pm 1.0 \times 10^{13}$  g/yr and a CO<sub>2</sub> concentration of approximately 110 ppm.

Our estimate of the mantle <sup>3</sup>He flux is the first determined independently of oceanographic <sup>3</sup>He measurements, and consequently represents a time-integrated flux substantially longer than ~1000 years. And although at the high end of the range of previous estimates, our estimate does not resolve the long-standing heat-helium paradox. Additionally, our requirement for heterogeneous pre-degassing <sup>3</sup>He/<sup>22</sup>Ne and <sup>3</sup>He/<sup>36</sup>Ar ratios between samples is contrary to conclusions of previous applications of disequilibrium degassing models, which advocated uniform ratios. Furthermore, we find that CO<sub>2</sub>/Ba ratios are highly variable in MORB samples, but still consistent with an average mantle mass ratio of ~100. However, estimating pre-degassing CO<sub>2</sub> concentrations and the mantle CO<sub>2</sub> flux depend strongly on the poorly-constrained carbon diffusivity. Consequently, our demonstration of the prevalence of disequilibrium during mid-ocean ridge degassing, and the potential for disequilibrium in other volcanic settings, highlights the need for better characterization of the physical parameters associated with volcanic degassing.

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## 1. Introduction

The abundances of volatile elements such as carbon, nitrogen, hydrogen, and noble gases place important constraints on the

provenance and timing of delivery of Earth's volatiles (Bergin et al., 2015; Marty, 2012; Morbidelli et al., 2000; Wänke, 1981). The flux of these volatiles between the mantle and surface affects climate stability and planetary habitability (Sleep and Zahnle, 2001; Walker et al., 1981). The mantle flux of He has long been used as a critical constraint on geodynamic modeling and the mantle structure (Gonnermann and Mukhopadhyay, 2009; Kellogg and Wasserburg, 1990; O'Nions and Oxburgh, 1983; Porcelli and Wasserburg, 1995; Tackley, 2015), and a means to establish fluxes and concentrations of other noble gases and elements such as C, N, S (Kagoshima et al., 2015; Marty, 1995, 2012; Marty and Dauphas, 2003; Marty and Tolstikhin, 1998; Trieloff and Kunz, 2005). The potential variability of nonradiogenic mantle noble gas elemental ratios is of interest to mantle dynamics, as uniform ratios are a requirement of “steady-state” mantle models (e.g., Porcelli and Ballentine, 2002), whereas heterogeneity may reflect a mantle incompletely mixed since Earth's accretion and/or different amounts of recycled atmospheric noble gases in mantle reservoirs (Caracausi et al., 2016; Kendrick et al., 2011, 2013; Mukhopadhyay, 2012; Mundl et al., 2017; Parai and Mukhopadhyay, 2015; Tucker and Mukhopadhyay, 2014).

While the budgets of volatiles in surface reservoirs (atmosphere, oceans, and crust) are relatively well quantified, concentrations in the mantle, and fluxes between the mantle and surface remain highly uncertain. Given the large size of the mantle, uncertainties in the mantle volatile concentrations render volatiles the least constrained aspect of the bulk silicate Earth composition.

Mid-ocean ridge basalts (MORBs) can serve as windows into the upper mantle composition. However, mantle volatile concentrations and fluxes are difficult to ascertain from measurements of oceanic basalts because many volatile species degas extensively prior to or during eruption. Consequently, MORB samples interpreted to have retained their pre-eruptive volatiles in vesicles, such as “popping rocks”, are extremely rare (Hekinian et al., 2000; Javoy and Pineau, 1991). Similarly rare are MORBs that have not nucleated vesicles (Le Voyer et al., 2017; Michael and Graham, 2015; Saal et al., 2002; Shimizu et al., 2016), and the undegassed nature of these samples has been questioned (Matthews et al., 2017).

Noble gases are ideal tracers of magmatic degassing processes because of their inert nature and the systematic variation of physical properties such as solubility and diffusivity with mass. More importantly, the radiogenic noble gases  $^4\text{He}^*$ ,  $^{21}\text{Ne}^*$ ,  $^{40}\text{Ar}^*$ , and  $^{136}\text{Xe}^*_\text{U}$  are produced from the radioactive decay of U, Th, and K at known rates (“\*” denotes the radiogenic abundance, i.e. excluding atoms not produced by radioactive decay, and the subscript “U” denotes  $^{136}\text{Xe}^*$  produced by spontaneous  $^{238}\text{U}$  fission; Section 2; Supplementary Information §S1). Consequently, mantle ratios of radiogenic noble gases—referred to as mantle “production ratios”—are fixed, and deviations from these production ratios measured in MORBs can be attributed to elemental fractionation during degassing. In contrast, elemental ratios involving species like  $\text{CO}_2$ ,  $\text{N}_2$ , or nonradiogenic noble gases can be affected by both degassing and heterogeneity in pre-degassing compositions.

Indeed,  $^4\text{He}^*/^{40}\text{Ar}^*$  ratios measured in MORBs often differ substantially from the mantle production ratio, interpreted to result from solubility-controlled He/Ar fractionation experienced during degassing (Jambon et al., 1985). Furthermore, within the context of a solubility-based fractional degassing model, the extent of  $^4\text{He}^*/^{40}\text{Ar}^*$  fractionation from the mantle production ratio is used to quantify the extent of degassing and predict pre-degassing concentrations of other volatiles, including  $\text{CO}_2$  (e.g. Burnard, 2001; Burnard et al., 2014; Colin et al., 2013; Marty and Tolstikhin, 1998; Marty and Zimmermann, 1999; Section 5.3).

Considering multiple radiogenic noble gas ratios can provide stringent constraints on the nature of the degassing, as all noble gases are affected by the same processes. For example, a particular

extent of equilibrium  $^4\text{He}^*/^{40}\text{Ar}^*$  fractionation will be accompanied by a predictable extent of  $^4\text{He}^*/^{21}\text{Ne}^*$  and  $^4\text{He}^*/^{136}\text{Xe}^*_\text{U}$  fractionation. Here, we will show that this prediction is not met in the vast majority of MORBs, demonstrating that MORB degassing cannot be described by any solubility-based degassing model, including the widely-used equilibrium Rayleigh distillation model. We will argue that kinetic fractionation plays a critical role in controlling volatile abundances in MORBs and present a parameterized model for degassing by disequilibrium Rayleigh distillation. While our model is not meant to capture all physical processes associated with magmatism such as storage, transport, and eruption, it demonstrates the manner in which disequilibrium degassing can affect volatiles. We will use it to reconstruct pre-degassing noble gas and  $\text{CO}_2$  concentrations in MORB magmas and the fluxes of these elements from the MORB mantle.

## 2. Equilibrium degassing models

We begin our discussion of mid-ocean ridge degassing by describing the most commonly applied equilibrium degassing models, and then testing them with a compilation of global MORB data.

### 2.1. Model description

The concentrations of volatiles in MORBs are controlled by diffusion of dissolved volatiles from the melt into  $\text{CO}_2$ -rich bubbles (Gerlach, 1989; Jambon et al., 1985; Sarda and Graham, 1990). Because noble gases are highly undersaturated in magmas, they do not nucleate bubbles, but rather partition into existing  $\text{CO}_2$  bubbles. During closed-system degassing, bubbles remain with the melt from which they were derived. Given sufficient time for equilibration, a noble gas  $i$  partitions between melt and bubbles according to its solubility, quantified by Henry's constant  $k_i$ , the ratio of dissolved concentration in the melt  $C_i$  to partial pressure in the vapor  $P_i$ :  $k_i = C_i/P_i$ . Noble gas solubilities decrease with size (Jambon et al., 1986; Lux, 1987); consequently, during equilibrium closed-system degassing, He/Ne, He/Ar, and He/Xe ratios increase in residual melt, where the maximum amount of fractionation is the ratio of solubilities (Jambon et al., 1986).

If buoyant bubbles rise independently of melts, for example if melts stall or ascend slowly, open-system degassing may occur and noble gas fractionations in residual melts larger than solubility ratios is possible. The possibility of open-system degassing via multiple stages of bubble formation and loss presents a challenge to uniquely constrain the degassing history of a sample. To simplify the problem, a particular degassing model is often assumed wherein exsolved volatiles are continuously lost from the magma. This process, described mathematically by Rayleigh distillation, has been commonly used to model the evolution of volatile element ratios and C isotopes during MORB degassing (Burnard, 1999, 2001; Burnard et al., 2002, 2004, 2014; Cartigny et al., 2001, 2008; Colin et al., 2013; Javoy et al., 1982; Marty, 1995; Marty and Tolstikhin, 1998; Marty and Zimmermann, 1999; Parai et al., 2012; Pineau and Javoy, 1983; Pineau et al., 1976; Sarda and Graham, 1990).

Rayleigh distillation can be expressed in closed-form as an equation for the ratio of two volatile species during degassing

$$C_i/C_j = C_{i,0}/C_{j,0} \times f_i^{(1-\alpha_{ij})} \quad (1)$$

Here  $f_i = C_i/C_{i,0}$  is the concentration ratio of a dissolved volatile  $i$  in a melt to its pre-degassing concentration, and the fractionation factor  $\alpha_{ij} = k_i/k_j$  is the ratio of solubilities. A full derivation of the equations describing the evolution of magmatic compositions during Rayleigh distillation is provided in the Supplementary Information (§S2).

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