

# Reconstructing the oxygen isotope composition of late Cambrian and Cretaceous hydrothermal vent fluid

Alexandra V. Turchyn<sup>a,\*</sup>, Jeffrey C. Alt<sup>b</sup>, Shaun T. Brown<sup>c,d</sup>, Donald J. DePaolo<sup>c,d</sup>,  
Rosalind M. Coggon<sup>e</sup>, Guoxiang Chi<sup>f</sup>, Jean H. Bédard<sup>g</sup>, Thomas Skulski<sup>h</sup>

<sup>a</sup> University of Cambridge, Department of Earth Sciences, Cambridge, UK

<sup>b</sup> University of Michigan, Department of Earth and Environmental Sciences, United States

<sup>c</sup> University of California, Department of Earth and Planetary Science, Berkeley, United States

<sup>d</sup> Earth Sciences Division E.O. Lawrence Berkeley National Lab, Berkeley, California, USA

<sup>e</sup> Imperial College, Department of Earth Science and Engineering, London, UK

<sup>f</sup> University of Regina, Department of Geology, Canada

<sup>g</sup> Geological Survey of Canada, Québec, Québec City, Canada

<sup>h</sup> Geological Survey of Canada, 601 Booth Street, Ottawa, ON K1A 0E8, Canada

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## Abstract

Oxygen isotope analyses ( $\delta^{18}\text{O}$ ) of 16 quartz–epidote pairs from late Cambrian (Betts Cove and Mings Bight, Newfoundland), Ordovician (Thetford Mines, Québec, Canada) and Cretaceous (Troodos, Cyprus) ophiolites are used to calculate the  $\delta^{18}\text{O}$  of the hydrothermal fluids from which they crystallized. We combine these with 3 quartz–fluid inclusion measurements and 3 quartz–magnetite measurements from the Cambrian ophiolites to explore how the range in the  $\delta^{18}\text{O}$  of submarine hydrothermal vent fluid has varied between the late Cambrian, Cretaceous and today. The range of calculated  $\delta^{18}\text{O}$  values of vent fluid (–4 to +7.4) is larger than that of modern seafloor hydrothermal vent fluid (0 to +4). We employ two numerical models to ascertain whether this range is most consistent with changes in paleo-seawater  $\delta^{18}\text{O}$  or with changes in the reactive flow path in ancient hydrothermal systems. A static calculation of the vent fluid oxygen isotope composition as a function of the water–rock ratio suggests that in an ocean with a lower  $\delta^{18}\text{O}$  than today, the range of vent fluid  $\delta^{18}\text{O}$  should be larger. Our data, however, show little evidence that the  $\delta^{18}\text{O}$  of the ocean was much lower than the global ice-free value of –1.2. A dual porosity model for reactive flow through fractured and porous media is used to model the relative evolution of the  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  of vent fluid in contact with rock. Our  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  for Cretaceous epidotes suggest the strontium concentration of the Cretaceous oceans may have been much higher than at present. The  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$  data from Cambrian epidotes are strikingly different from the younger samples, and are difficult to model unless fluid–rock interaction in the Cambrian hydrothermal systems was substantially different. It is also possible that some of the quartz–epidote veins have been reset by obduction-related metamorphism. Our data suggest that the high calcium-to-sulfate ratio in early (and Cretaceous) seawater may have affected the degree of strontium isotope exchange, causing hydrothermal fluids to have  $^{87}\text{Sr}/^{86}\text{Sr}$  closer to that of seawater than in modern systems.

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## 1. INTRODUCTION

The chemical composition of seawater has changed over Earth history, and this has had a substantial impact on

climate and the evolution of life. Our ability to reconstruct the chemistry of past oceans relies largely on chemical and isotopic measurements made on carbonate and silicate minerals and sedimentary fossils (Epstein et al., 1951; Emiliani, 1955; Shackleton, 1967; Knauth and Epstein, 1976; Knauth and Lowe, 1978; Broecker, 1986; Veizer

\* Corresponding author. Tel.: +44 1223 333479.

E-mail address: [avt25@cam.ac.uk](mailto:avt25@cam.ac.uk) (A.V. Turchyn).

et al., 1999; Zachos et al., 2001). During sedimentation, lithification, compaction and dewatering, however, many minerals undergo a series of chemical and physical reactions collectively called ‘burial diagenesis’ that may alter their original chemical signature, leading to potentially erroneous interpretations of the paleoceanographic record (Berner, 1980; Schrag et al., 1995; Fantle and DePaolo, 2007). This has been a particular problem for measurements of the oxygen isotope ratio ( $\delta^{18}\text{O}$ ) of carbonate minerals and fossils over the Phanerozoic (Jacobsen and Kaufman, 1999; Veizer et al., 1999). The  $\delta^{18}\text{O}$  of carbonate minerals and fossils can be used as a paleothermometer, making this proxy particularly important for the paleoceanographic community. Difficulties arise because carbonate minerals are particularly susceptible to marine burial diagenesis. Reequilibration with pore fluids, and water–rock interaction with the sediment and basement, may have the effect of lowering the  $\delta^{18}\text{O}$  of the carbonate minerals (Schrag et al., 1995).

Circumventing the problem of chemical alteration during burial diagenesis remains a critical issue for paleoceanographic and paleochemical reconstruction (e.g., Pearson et al., 2001). The paradigm of using ‘low-temperature’ minerals for geochemical oceanography allows for high-resolution reconstruction of the temporal evolution of past oceans by sampling well-exposed sections with good age control. Interpretation of these measurements, however, is commonly confounded by the poorly-constrained effects of post burial processes. As an independent approach for fingerprinting past ocean chemistry, several groups have suggested the use of ‘higher temperature minerals’. Circulation of seawater-derived fluids through the cooling upper oceanic crust is driven by the heat of magma chambers beneath ocean ridges (Lister, 1972; Wolery and Sleep, 1976). The chemical signal of paleo-ocean water could, in theory, be recorded in the alteration products of these water–rock interactions. Interpreting the chemical and isotopic signature of these hydrothermal minerals, however, is also challenging.

The majority of studies that have attempted to use the isotopic composition of hydrothermally altered rock to infer the chemical composition of water in past oceans have looked at the whole-rock oxygen or strontium isotope composition (Muelenbachs and Clayton, 1976; Gregory and Taylor, 1981; Muelenbachs, 1986; Holmden and Muelenbachs, 1993; Muelenbachs, 1998; Muelenbachs et al., 2003). As the temperature of water–rock interaction increases with depth in a hydrothermal system, both the type of alteration minerals and their individual oxygen isotope fractionation factors change. Typically low-temperature water–rock interactions in the shallow portion of submarine hydrothermal systems result in altered rock that is enriched in  $^{18}\text{O}$  (relative to unaltered rock), whereas the high-temperature water–rock interactions deeper in hydrothermal systems result in altered rock that is depleted in  $^{18}\text{O}$  (Muelenbachs and Clayton, 1976; Alt et al., 1986a). These profiles of  $\delta^{18}\text{O}$  measurements through exhumed segments of ancient ocean crust (ophiolites) are typically made on the bulk-rock end members of hydrothermal alteration products. Temperature and fluid composition are poorly constrained from

these data (Kasting et al., 2006). Since the flow path of fluids and the prior history of an individual hydrothermal sample are typically not known, these  $\delta^{18}\text{O}$  profiles may yield more valuable information about the nature of fluid flow patterns through ancient ocean crust than they do about the chemical composition of the fluid itself. A few studies have attempted to use both the bulk rock data combined with mineral proportions in the bulk rocks with modal mineralogy to calculate fluid composition (Harper et al., 1988; Alexander et al., 1993; Muehlenbachs et al., 2003).

Analysis of vein-filling minerals in ancient oceanic crust provides an alternative approach to reconstructing paleo-hydrothermal fluid and its relationship to paleo-seawater. Major element ratio analyses of calcite and aragonite vein minerals in ocean crust younger than 170 million years old suggest that hydrothermal alteration minerals do record chemical signals of past oceans (Coggon et al., 2010). Coggon et al. (2010) used the magnesium–calcium ratios and the strontium–calcium ratios of suites of carbonate veins that record the temperature-dependent geochemical evolution of ridge flank hydrothermal fluids, and interpreted the deviation away from their initial seawater compositions as being due to seawater–basalt interactions at moderate temperatures (<60 °C). Although the temporal resolution is not as fine as that of a measured sequence in a fossiliferous carbonate platform, Coggon et al. (2010) demonstrate that the Mg/Ca and Sr/Ca ratios of the Cretaceous and early Cenozoic oceans were lower than today. This had already been suspected from fluid inclusion data (Lowenstein et al., 2001; Horita et al., 2002) and from trace metal concentrations of biogenic calcite (particularly Mg/Ca – Dickson, 2002), but the direct measurement of basalt-hosted carbonate minerals provides independent corroboration of these results.

The chemical evolution of high temperature fluids in hydrothermal cells may be further constrained using mathematical models (DePaolo, 2006; Alt and Bach, 2006; Coumou et al., 2009). Such models can also provide a framework for understanding how high temperature vent fluid records the integrated chemical and physical pathway of a parcel of water through the ocean crust, allowing us to understand the possible chemical link between high temperature vent fluid chemistry and that of seawater. The chemical evolution of a parcel of water in a hydrothermal system depends critically on the residence time of fluid in the ocean crust (how long is it there to react?), the water–rock ratio of the parcel of water with the surrounding basalt (how much rock does the fluid interact with?), and the temperature and nature of the hydrothermal reactions (what chemical reactions are taking place and which minerals are reacting with the water?). Chromatographic effects, which separate differently reactive elements in the circulating fluid, are important in these settings. Modeling work on fluid flow through ocean crust suggests that the residence time of water through the high temperature arm of modern hydrothermal circulation cells is short (~3–5 years) and modeling and observations of ocean crust alteration indicate that axial hydrothermal cells are spatially decoupled from the flow of water through the low-to-moderate temperature

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