



New boron isotopic evidence for sedimentary and magmatic fluid influence in the shallow hydrothermal vent system of Milos Island (Aegean Sea, Greece)



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ABSTRACT

Magmatic sources may contribute a significant amount of volatiles in geothermal springs; however, their role is poorly understood in submarine hydrothermal systems worldwide. In this study, new results of B and $\delta^{11}\text{B}$ in 41 hydrothermal vent waters collected from the shallow hydrothermal system of Milos island in the Aegean Sea were combined with previously published data from other tectonic settings and laboratory experiments to quantify the effects of phase separation, fluid/sediment interaction and magmatic contribution. Two Cl-extreme solutions were identified, high-Cl waters (Cl as high as 2000 mM) and low-Cl waters (Cl < 80 mM). Both sets of waters were characterized by high B/Cl ($\sim 1.2\text{--}5.3 \times 10^{-3}$ mol/mol) and extremely low $\delta^{11}\text{B}$ (1.4–6.3‰), except for the waters with Mg content of near the seawater value and $\delta^{11}\text{B} = 10.3\text{--}17.4\%$. These high-Cl waters with high B/Cl and low $\delta^{11}\text{B}$ plot close to the vent waters in sediment-hosted hydrothermal system (i.e., Okinawa Trough) or fumarole condensates from on-land volcanoes, implying B addition from sediment or magmatic fluids plays an important role. This is in agreement with fluid/sediment interactions resulting in the observed B and $\delta^{11}\text{B}$, as well as previously reported Br/I/Cl ratios, supporting a scenario of slab-derived fluid addition with elevated B, ^{11}B -rich, and low Br/Cl and I/Cl, which is derived from the dehydration of subducted-sediments. The slab fluid becomes subsequently mixed with the parent magma of Milos. The deep brine reservoir is partially affected by injections of magmatic fluid/gases during degassing. The results presented here are crucial for deciphering the evolution of the brine reservoirs involved in phase separation, fluid/sediment interaction and magmatic contribution in the deep reaction zone of the Milos hydrothermal system; they also have implications in the understanding of the formation of metallic vein mineralization.

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1. Introduction

Many hydrothermal systems have been discovered in tectonic settings ranging from sediment-starved mid-ocean ridge (MOR) and back-arc basin (BAB), to sediment-hosted ridges (SR) (Lonsdale, 1977; Von Damm et al., 1985a,b; Von Damm and Bischoff, 1987; Campbell et al., 1988; Campbell and Edmond, 1989; Butterfield et al., 1990; Fouquet et al., 1991). The fluids discharging from the hydrothermal vents found in such systems can play critical role in the chemical mass

balance of ocean. Moreover, the fluids influence profoundly the chemistry of the surrounding rocks in the discharge zones, with significant physicochemical transformations occurring in shallow hydrothermal vents compared with the deep-sea environments (Botz et al., 1996; Fitzsimons et al., 1997; Pichler et al., 1999; Prol-Ledesma et al., 2004; McCarthy et al., 2005; Ishibashi et al., 2008; Tassi et al., 2009). However, a systematic comparison of fluid chemistry in terms of the degree of fluid–rock/sediment interaction and the effects of phase separation among MOR, BAB, SR, and shallow-water systems has not been carried out. Magmatic heating provides common driving forces for hydrothermal circulation at mid-ocean ridges and on-land volcanoes. Occurrences of noble and volatile gases, as well as elemental and isotopic changes in chemistry, have been documented in detail, after magmatic intrusion or volcanic eruptions. These results emphasize the importance of magmatic inputs in hydrothermal fluids (Urabe et al., 1995; Von Damm, 2000; Seyfried et al., 2003; Von Damm et al., 2003; Somoza et al., 2004; Ruzié et al., 2012). Several studies have suggested variable pathways

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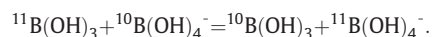
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of magmatic gas/water evolution to cause widespread chemical compositional variation in shallow-water systems, making them good analogs for systems occurring in mid-ocean ridges or on land (Sedwick and Stüben, 1996; Capaccioni et al., 2005, 2007; Tassi et al., 2009). However, the role of magmatic inputs at shallow-sea hydrothermal system remains poorly constrained.

Boron has two stable isotopes, ^{10}B and ^{11}B , with natural abundances of 19.9 and 80.1%, respectively. In seawater, B is present as boric acid ($\text{B}(\text{OH})_3$, a trigonal species) and borate ions ($\text{B}(\text{OH})_4^-$, a tetrahedral species). The ion exchange reaction of ^{10}B and ^{11}B between the two species is described as:



Previously experimental data indicated that $\text{B}(\text{OH})_3$ is enriched in ^{11}B relative to $\text{B}(\text{OH})_4^-$ (Kakihana et al., 1977; Klochko et al., 2006). Their relative proportions are generally a function of pH where the predominant isotopic fractionation occurs via equilibrium exchange between the two aqueous species, $\text{B}(\text{OH})_3$ dominates at low pH and $\text{B}(\text{OH})_4^-$ at high pH (Kakihana et al., 1977; Hershey et al., 1986; Spivack and Edmond, 1987). Boron isotopes provide useful information for deciphering the origins and evolution mechanisms of vent fluids in various geological settings due to the large relative mass difference between the two isotopes and their high geochemical reactivity, which caused significant B isotopic fractionations (e.g., Spivack and Edmond, 1987; Spivack et al., 1987; Palmer and Sturchio, 1990; Palmer, 1991; You et al., 1993, 1994). Almost linear co-variation of B/Cl with 1/Cl is evident in laboratory liquid–vapor phase separation experiments. The observed small difference in B/Cl and 1/Cl trends between experiments and field data indicates that phase separation predominantly controls the Cl concentration in both phases in MOR system (Bischoff and Rosenbauer, 1987; Berndt and Seyfried, 1990; You et al., 1994). B isotopic fractionation is associated with boiling and phase separation in thermal waters, whereby ^{11}B partitions selectively into the vapor phase (Spivack et al., 1990; Leeman et al., 1992; Liebscher et al., 2005). However, empirical and experimental data indicate that B isotopic difference between liquid and vapor is rather small (–3 to –1‰ at ~140–300 °C) and decreases with increasing temperature, with negligible isotopic fractionation above 400 °C (Palmer and Sturchio, 1990; Spivack et al., 1990; Liebscher et al., 2005). The B enrichment in oceanic rocks induced by seafloor alteration, mainly relates to low-temperature mineralogical changes, such as substitution of primary minerals by smectite and palagonitization of basaltic glass during seawater percolation (Seyfried et al., 1984; Spivack and Edmond, 1987). The B and $\delta^{11}\text{B}$ in different geochemical reservoirs showed extremely wide distribution, ranging from <0.1 to >100 ppm and –30 to +60‰ (relative to the NBS SRM-951), respectively (Barth, 1993; Bebout et al., 1993). The dissolved B in these reservoir fluids often represents mixtures of different origins: natural hydrothermal fluids and altered rocks/sediments (Spivack and Edmond, 1987; Berndt and Seyfried, 1990; Palmer, 1991; You et al., 1994). There are several previous studies of B and B isotopic composition in fumarolic condensates and geothermal fluids from volcanic regions (e.g., Vulcano Island, Italy; Taupo volcanic zone, New Zealand); in those studies, the distinct end-member fluid composition for reconstruction of temporal evolution of magmatic sources was identified (Leeman et al., 2005; Millot et al., 2012; Reyes and Trompeter, 2012). In addition, high B/Cl ratio in discharging fluids was ascribed to input of B-enriched component and was used as a proxy for CO_2/Cl to trace the magmatic addition (Giggenbach, 1995).

Based on stable isotope data, specifically S isotope and noble gases data, previous efforts have emphasized the importance of seawater and magmatic water components at the island of Milos, as well as the islands of Santorini and Nisyros, and perhaps in the whole Aegean volcanic arc, although the magmatic contribution is still ambiguously argued (Botz et al., 1996; Marini et al., 2002; Brombach et al., 2003; Naden et al., 2005; Shimizu et al., 2005; Dotsika et al., 2009; Price

et al., 2013). A time-series study of fluid chemistry could provide a more clear view that would enable a better understanding of the processes on Milos. In this study, 41 new samples of vent fluids from a 2003 expedition on Milos were analyzed for B and $\delta^{11}\text{B}$, bringing a new perspective in the understanding of candidates of source fluids in the hydrothermal system. The data are complemented by a compilation of other published geochemical results of fluids or fumarolic condensates from Milos and other locations in the Aegean geothermal system, as well as various other submarine hydrothermal systems (e.g., MOR, BAB, SR) and subaerial volcanoes (e.g., Taupo volcano, New Zealand; Vulcano Island, Italy). We aim to utilize new and published results of B, $\delta^{11}\text{B}$, B/Cl and literature data of halogen (Cl, Br, I) to examine the sources and evolutionary history of the deep saline reservoir, as well as to elucidate the role of vapor–brine phase separation, magmatic fluids, and fluid/sediment interaction in the Milos system.

2. Samples and experiments

2.1. Geological settings

The subduction of the African plate beneath the Eurasian plate has resulted in the formation of the Mediterranean Ridge and the Hellenic Trench, as well as volcanism in the Aegean volcanic arc and the back-arc extension in the Aegean Sea (McKenzie, 1970, 1972; Agostini et al., 2010; Ring et al., 2010). The Aegean volcanic arc is constituted from west to east by the islands of Methana, Milos, Santorini, Kos and Nisyros (Fig. 1) (e.g., Shimizu et al., 2005; Varnavas and Cronan, 2005). The islands of the central-eastern part of the arc (Milos, Santorini, and Nisyros) are presently active geothermal systems, while Methana and Kos display declining hydrothermal activity (Shimizu et al., 2005; Dotsika et al., 2009). The volcanic rocks compositions of arc islands are associated with calc-alkaline and high-K calc-alkaline exception of basalts presented in Santorini island group (Francalanci et al., 2005). The age determinations of erupted lava suggested that the islands lying on the south of volcanic arc (e.g., Methana, Milos, Santorini and Nisyros) show younger ages relative to those from the islands on the north side of arc (e.g., Aegina, Kos), implying the southward migration of volcanic front (Matsuda et al., 1999; Francalanci et al., 2005). Papazachos et al. (1995) reported that the continental crust is relatively thinner in the central sector of the arc than the western and eastern sectors, constraining the influence of shallow crustal contamination effect for magma genesis. The volcanism on Milos Island started from 3.5 to 0.08 Ma, subsequently the volcanic sequence was overlain by recent alluvial deposits (Fytikas et al., 1986; Fytikas, 1989). Several main cycles of volcanic activity have been distinguished, including the initial submarine eruptions and the followed pyroclastic material deposition, which resulted from subaerial eruptions that composed mainly of lava domes and flows (Fytikas et al., 1986). This island is mostly composed of calc-alkaline series volcanic rocks, which range from basaltic andesites to dacites, and rhyolites (Fytikas et al., 1986; Fytikas, 1989). The bulk sediment inshore shows a dominant detrital fraction and is highly influenced by hydrothermal activity (Varnavas and Cronan, 2005).

2.2. Sample location, collection and analytical methods

There are abundant discharges of hot water and steam distributed either on land or offshore within shallow depth at Milos. CO_2 (as high of ~90% v/v) is the dominant component of gas venting (Dando et al., 1995; Botz et al., 1996). Various types of fluid discharge are surrounding the coast of Milos, including intense degassing with steams, emanating of saline fluid as brine pool and diffusive warm fluid (Valsami-Jones et al., 2005; Price et al., 2013). Wells drilled in past years in the Zephyria area as part of the geothermal exploration of the island, reached a maximum depth of 1101–1381 m and found evidence for a saline reservoir distributed at depth 1–2 km with chlorinity of >2.5 times seawater value and temperature of 300–325 °C (Fig. 1) (Liakopoulos et al.,

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