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## Geochemical impacts of hydrothermal activity on surface deposits at the Southwest Indian Ridge

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## ABSTRACT

Submarine hydrothermal circulation has attracted much scientific interest since seafloor hydrothermal activity was first observed in the 1970s; an area of particular interest is the impact of exported inorganic and organic materials from hydrothermal vent systems into the open ocean. In 2007, the first active hydrothermal vent field, with vent fluid temperatures up to 379 °C, was discovered at the ultraslow spreading Southwest Indian Ridge (SWIR), where active vents are much less abundant than fast spreading ridges, and the effect of hydrothermal extrusion on surface sediments is not fully understood. To explore how geochemical proxy signatures respond to hydrothermal activity, we investigated the distributions of elements, minerals and lipids in surficial normal marine sediments, metalliferous sediments and low-temperature hydrothermal deposits collected from the SWIR. The results showed different effects of hydrothermal activity on the surface deposits. The normal marine sediments were predominantly calcium carbonate characterized by > 42% CaO and > 90% calcite, with a significant autochthonous marine contribution to organic matter (OM) and a predominance of lower molecular weight alkanols and fatty acids; they were uninfluenced by hydrothermal activity but received some terrigenous input represented by abundant high molecular weight *n*-alkanes with an odd-over-even predominance. The near-field metalliferous sediments and hydrothermal deposits were very different. Some near-field metalliferous sediments were influenced by low-temperature hydrothermal activity, and their distributions of elements and minerals were similar to those of hydrothermal deposits, which were characterized by abundant Fe/Si and opal/natronite. Other near-field metalliferous sediments were evidently influenced by mixing of high-temperature hydrothermal sulfides typically containing abundant Cu/Zn. With respect to the organic matter assemblages, near-field deposits contained little evidence for thermal maturation of organic matter and all were characterized by a strong microbial signature, including hopanoids, isoprenoidal and non-isoprenoidal dialkyl glycerol ether lipids, and low molecular weight *n*-alkanes with an even carbon number predominance. The far-field metalliferous sediments, despite the influence of non-buoyant plumes and slightly higher concentrations of hydrothermal-derived metals (e.g., Fe, Cu and Zn), had the same distribution of organic lipids and major mineral composition (> 90% calcite) as did normal marine sediments. Thus, the influence of non-buoyant plume inputs appears to have been minimal possibly due to the dilution of in situ microorganisms by normal marine organisms in sediment and seawater. Furthermore, these characteristics indicate inorganic indices based on abundant metal elements derived from the hydrothermal systems (such as Fe/Cu/Zn content,  $\Sigma\text{REE}/\text{Fe}$ , the ternary diagram of Fe, Cu  $\times$  100 and Ca) are more sensitive, serving as better proxies than organic matter assemblages to differentiate the effects of diverse hydrothermal activity on surface deposits.

### 1. Introduction

Hydrothermal circulation, a common process along mid-ocean ridges, plays an important role in global ocean cycles via significant inputs of reduced substrates, such as H<sub>2</sub>S, H<sub>2</sub>, CH<sub>4</sub>, NH<sub>3</sub>, Mn<sup>2+</sup> and Fe<sup>2+</sup>, which can fuel chemosynthetic microbial metabolism (e.g., De

Angelis et al., 1993; Elderfield and Schultz, 1996; Mccollom, 2000; Lam et al., 2004; Dick et al., 2009; Petersen et al., 2011; Dick et al., 2013), and even be a significant source of carbon to the deep ocean (e.g., Mccollom, 2000; Lang et al., 2006; Mccarthy et al., 2011). Previous hydrothermal studies have mainly focused on near-field hydrothermal products, such as sulfide structures (e.g., Kato et al., 2010; Peng et al.,

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**Table 1**

The average abundances (and associated indices) for major elements, trace elements and rare earth elements in sediments from the Southwest Indian Ridge.

Type	Al <sub>2</sub> O <sub>3</sub> (%)	CaO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	K <sub>2</sub> O (%)	MgO (%)	MnO (%)	Na <sub>2</sub> O (%)	P <sub>2</sub> O <sub>5</sub> (%)	TiO <sub>2</sub> (%)	LOI (%)
Background sediments	0.86	44	0.70	0.15	0.50	0.062	1.3	0.053	0.058	5.0
M-T1	2.5	39	2.6	0.17	2.1	0.076	1.6	0.076	0.18	6.9
M-T2	0.25	4.6	21	0.46	1.1	3.8	2.9	0.77	0.017	22
M-T3	1.9	5.6	25	0.18	3.8	1.1	2.0	0.56	0.15	11
Hydrothermal deposits	0.041	0.45	8.9	0.33	0.66	0.95	2.7	0.20	0.003	11
Type	V (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)	Cu (ppm)	Zn (ppm)	Sr (ppm)	Mo (ppm)	Ba (ppm)	Pb (ppm)
Background sediments	15	11	14	17	30	25	1400	0.62	290	7.8
M-T1	51	68	18	56	280	86	1200	0.70	290	18
M-T2	210	6.0	33	30	2200	740	390	140	360	94
M-T3	340	160	350	90	11,000	2300	260	120	160	44
Hydrothermal deposits	66	2.2	3.2	19	45	86	190	150	890	4.8
Type	Li (ppm)	Be (ppm)	Sc (ppm)	Rb (ppm)	Y (ppm)	Zr (ppm)	Nb (ppm)	Cs (ppm)	Hf (ppm)	Th (ppm)
Background sediments	10	0.13	1.8	4.0	8.2	8.9	0.90	0.22	0.31	0.74
M-T1	23	0.16	5.4	4.3	9.0	16	0.79	0.21	0.59	0.62
M-T2	38	0.69	0.63	4.9	9.6	5.6	0.29	0.12	0.13	0.17
M-T3	7.2	0.26	4.9	3.2	11	22	0.80	0.16	0.66	0.58
Hydrothermal deposits	57	0.36	0.76	3.0	1.9	1.0	0.064	0.14	0.023	0.037
Type	U (ppm)	Al/(Al + Fe + Mn)	Fe/Ti	ΣREE (ppm)	ΣREE/Fe (10 <sup>-4</sup> )	δCe	δEu			
Background sediments	0.31	0.47	14	24	63	0.65	1.2			
M-T1	0.32	0.41	18	23	13	0.64	1.3			
M-T2	5.3	0.0082	4300	18	1.3	0.56	3.9			
M-T3	12	0.054	570	28	1.6	0.63	1.6			
Hydrothermal deposits	4.2	0.0041	3900	3.6	0.54	0.66	18			

Note: LOI = Loss on ignition,  $\delta Ce = Ce_N / \sqrt{La_N \times Pr_N}$ ,  $\delta Eu = Eu_N / \sqrt{Sm_N \times Gd_N}$ , Ce<sub>N</sub>, La<sub>N</sub>, Pr<sub>N</sub>, Eu<sub>N</sub>, Sm<sub>N</sub>, Gd<sub>N</sub> represent North American shale composite-normalized data.

2011b; Jaeschke et al., 2012; Gibson et al., 2013; Reeves et al., 2014), hydrothermally influenced sediments (e.g., Schouten et al., 2003; Shulga et al., 2010; Shulga and Peresypkin, 2012), and rising plumes (e.g., Bennett et al., 2011; Sands et al., 2012; Estapa et al., 2015), in relation to the characteristics of inorganic (elements and minerals) and organic (lipids) geochemistry, biogeography and biodiversity. However, a growing number of studies have focused on the microbial ecology and biogeochemical cycles involving the transport of metals and organic carbon in non-buoyant plumes (e.g., Bennett et al., 2008; Bennett et al., 2011; Lesniewski et al., 2012; Sylvan et al., 2012; Li et al., 2015; Sander and Koschinsky, 2016). Of particular interest has been hydrothermally derived dissolved Fe, which can be dispersed over thousands of kilometers away from its source into the open ocean and contribute to the global oceanic Fe budget (e.g., Toner et al., 2012; Fitzsimmons et al., 2014, 2017; Resing et al., 2015; Kleint et al., 2016). However, the geochemical characteristics of the sediments influenced by such non-buoyant plumes remain largely unstudied.

Low-temperature hydrothermal systems with formation temperatures of < 100 °C had previously been largely ignored but have recently become research hotspots. Relatively recent investigations of such settings have focused on biogeochemical cycling mechanisms of Fe, Mn, and S (e.g., Butterfield et al., 2004; Perner et al., 2007; Edwards et al., 2011; Sun et al., 2011, 2013, 2015) and the microbial ecology and biogeochemistry of low-temperature hydrothermal environments (e.g., Edwards et al., 2011; Peng et al., 2011a; Li et al., 2012, 2013). These have confirmed that low-temperature settings have geochemical characteristics and microbial communities distinct from those of high-temperature hydrothermal systems (e.g., Blumenberg et al., 2012; Jaeschke et al., 2012; Gibson et al., 2013; Reeves et al., 2014).

Increasing attention is being paid to hydrothermal fields at the ultraslow spreading Southwest Indian Ridge (SWIR) because more hydrothermal vents (including high-temperature and low-temperature hydrothermal fields) than expected were discovered since 2007 (e.g., Fujimoto et al., 1999; Münch et al., 2001; Bach et al., 2002; German, 2003; Tao et al., 2007, 2012), and there have been some reports on the petrology and element geochemistry (e.g., Tao et al., 2011, 2012; Cao et al., 2012; Gao et al., 2016; Z. Li et al., 2016) and molecular biology (e.g., Peng et al., 2011a; Li et al., 2013; Cao et al., 2014; J. Li et al., 2016). However, research on lipid biomarkers in the SWIR hydrothermal systems remains rare (Huang et al., 2014; Lei et al., 2015). There are also relatively few studies on the effects of hydrothermal activity on the surrounding environment, especially the metalliferous sediments (Pan et al., 2016) formed via a combination of sulfide mass wasting and debris flow, low-temperature fluid flow and mineralization, or plume formation, dispersal and fallout (Dias et al., 2008).

The surface deposits (0–10 cm) studied in this paper, including normal pelagic sediment, far-field and near-field metalliferous sediments, and low-temperature hydrothermal deposits, were collected from the first discovered active hydrothermal vent field, the Dragon Vent Field (49°39' E, 37°47' S), a nearby inactive field (50°28' E, 37°39.50' S) and surrounding areas (Fig. 1 and Supporting Information Table S1) during the DY115-20 and DY115-21 expeditions of the R/V Da Yang Yihao in 2009 and 2010, by using a television-video guided grab. These three distinct surface deposits provide an opportunity to explore the potential effects of hydrothermal activity on the surrounding sediments. Our previous study examining the distribution of glycerol dialkyl (and monoalkyl) glycerol tetraether (GDGT and MGTT) archaeal membrane lipids (Pan et al., 2016) clearly showed that GDGT

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