



The geochemical characteristics and Fe(II) oxidation kinetics of hydrothermal plumes at the Southwest Indian Ridge

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ABSTRACT

The Southwest Indian Ridge (SWIR) is one of the world's slowest spreading ridges with a full spreading rate of $\sim 14 \text{ mm a}^{-1}$. Due to its low thermal budget, high-temperature hydrothermal activity along the SWIR was once considered to be impossible. The Chinese cruise DY115-19 on board R/V Dayang Yihao successfully discovered the first SWIR active hydrothermal field at $37^{\circ}47'S$ $49^{\circ}39'E$, located at a magmatically robust spreading segment. Here, the geochemical characteristics of hydrothermal plumes from the hydrothermal field are first reported, and water column anomalies of light transmission, Fe, Mn, Al, both dissolved and particulate, are discussed. The total Fe and dissolved Mn concentrations in the plumes varied from 13.7 to 277.4 nM and 0.47 to 10.41 nM, respectively. The composition of Fe-Mn-Al implied that particles in the plumes were mainly hydrothermal in origin, also included small contributions of resuspended sediments or background particles. Dissolved Fe constituted a considerable fraction of the total Fe, more than 80% in plume samples from station CTD 4. High Fe concentrations might be sustained in the dissolved phase because of the existence of organic complexes and nanoparticles. On board incubation experiments verified the Fe(II) oxidation half-lives for plumes of CTD 4 and CTD 13 were 1.8 and 1.6 h, respectively, which are much longer than the calculated value of ~ 0.5 h based on the deep water pH and oxygen concentration.

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1. Introduction

After the discovery of hot springs on the East Pacific Rise (EPR) (Corliss et al., 1979; Edmond et al., 1979, 1982; Spiess et al., 1980), detailed surveys of hydrothermal vents and plumes have been extensively conducted mainly at ridges in the Pacific and Atlantic oceans. It was assumed for a long time that only fast to intermediate spreading ridges might generate such high-temperature vents. Until recently the ultraslow spreading ocean ridges have been little studied, typically along Gakkel Ridge in the Arctic Ocean (Edmonds et al., 2003) and Southwest Indian Ridge (SWIR) (German et al., 1998). The results of the two surveys both indicated a higher frequency of venting than expected, with 9–12 discrete vents sites along 1100 km of Arctic ridge and six locations within two 200 km long sections of the SWIR. According to the study of Baker et al. (2004), ultraslow ridges are several times more efficient than faster-spreading ridges in supporting active vent fields due to the combination of three-dimensional magma focusing at volcanic centers, deep mining of heat from gabbroic intrusions and direct cooling of the upper mantle, and nonmagmatic heat supplied by exothermic serpentinization.

The SWIR is a major plate boundary of the world oceans, separating Africa and Antarctica, linking the Atlantic Ocean with the Indian Ocean and the Pacific Ocean. The underlying basement consists mainly of ultramafic rocks (Dick et al., 2003). The first discovery of hydrothermal vents at SWIR was made in Feb. 2007 using the Autonomous Benthic Explorer of the Woods Hole Oceanographic Institution (WHOI) (Tao et al., 2007). The hydrothermal field is located on SWIR at $49^{\circ}39'E$, $37^{\circ}47'S$, and it consists of three active zones extending ~ 1000 m laterally (Tao et al., 2012). In this study, the geochemical characteristics and light transmission properties of hydrothermal plumes on the SWIR are first detailed, including Fe, Mn, and Al concentration, both dissolved and particulate.

The kinetics of Fe(II) oxidation in plume water was also investigated by on board incubation experiments. Fluid emitting from hydrothermal vents is acidic, contains high Fe(II) concentrations and when it enters into the cold, mildly alkaline and well oxygenated surrounding seawater, Fe(II) is oxidized and forms Fe hydroxide. Fe(II) oxidation in hydrothermal plumes is a key process because it impacts the geochemistry of other elements, for example, within the buoyant plume, the oxyanions, V, P, and As, are co-precipitated with Fe oxyhydroxides, and the scavenging of rare earth elements by these Fe precipitates begins in the buoyant plume and continues in the neutrally buoyant plume (Edmonds and German, 2004; Feely et al., 1992, 1996; German et al., 1990; German et al., 1991a,b). Recent studies focusing on vent-derived Fe indicated that Fe can be stabilized in the dissolved state by organics and nanoparticles in the fraction $<0.4 \mu\text{m}$ (Bennett

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et al., 2008; Tagliabue et al., 2010; Yücel et al., 2011) and exert great influence on the local and global ecological environment.

2. Samples and methods

2.1. Dissolved and particulate trace elements

All samples of CTD 4, CTD 9, CTD 12, and CTD 13 were collected in the hydrothermal field (Fig. 1) during the fifth leg of the Chinese cruise DY 115–21 on board R/V Da Yang Yihao in Jan. 2010. The four stations were conducted as on-line measurements with a seabird 911 CTD probe combined with a MAPR (Miniature Autonomous Plume Recorder) with an optical backscatter sensor (OBS) for turbidity measurements.

Total Fe (TFe) concentrations in the plumes were determined on board ship using the modified method of King et al. (1991). In brief, a 200 ml aliquot of unfiltered seawater was transferred into a pre-cleaned polyethylene bottle and, immediately after sampling, 200 μ l ultrapure HNO₃ and 500 μ l 10% hydroxylamine hydrochloride were added to dissolve the particulate and reduce Fe(III) to Fe(II). After standing for half an hour, the pH was adjusted to 6.5–8.0 using 5.0 M ammonia. Then, the water sample was passed through a C18 Sep-Pak column loaded with the colorimetric reagent ferrozine [3-(2-pyridyl)-5,6-bis(4-phenylsulfonic acid)-1,2,4-triazine] by peristaltic pump. The C18 Sep-Pak column preparation and the colorimetric reagent ferrozine loading were according to King et al. (1991). For removal of the non-acid soluble particulate, an in-line filter holder (Millipore Corporation) with 47 mm filter (0.4 μ m) was used. All solutions were purified by flowing them through a C18 column loaded with ferrozine in order to obtain a low Fe blank. Contamination from the ship and air was minimized because the entire sample stream was closed to the atmosphere. The Fe(FZ)₃ complex was quantitatively retained and was washed with 10 ml methanol and the maximum absorbance was determined at 562 nm. The detection limit with a concentration factor 20 is 1.35 nM, and the standard error is 5%.

Particulate and dissolved phases from plumes (sample locations based on the light scattering anomaly) were collected using rosette-mounted Niskin bottles. Two to four liter seawater samples were filtered onto acid-cleaned Whatman Nuclepore polycarbonate filters (0.4 μ m). Then the particulate samples were rinsed with Milli-Q water in order to remove the sea salts. They were frozen until elemental

analysis. About 1 l of filtered water was acidified with ultrapure HNO₃ to pH < 2.0 for dissolved trace elements analysis.

Dissolved Fe (DFe), Mn (DMn), Al (DAI) concentrations in 1 l aliquots were determined using ICP-MS following preconcentration with Chelex 100. Chelex 100 resin (50–100 mesh; 2.0 g) was added to seawater. The mixture was equilibrated with the resins for 24 h on a shaking plate. The long equilibration time is required when metal species with slow dissociation rates are present. The liquid phase was separated by filtration, and the resin rapidly washed with Milli-Q water and 2.0 M HNO₃ according to Rahmi et al. (2007). For particulate Fe (PFe), Mn (PMn), Al (PAI) measurement, filters were heated to reflux with concentrated ultrapure nitric acid in 7 ml Teflon vials for 3 h and analyzed by ICP-MS. The precision and accuracy of trace element determinations for both sets of analyses is < 4%.

2.2. Iron oxidation kinetics experiments

Iron oxidation incubation experiments were conducted on board R/V Da Yang Yihao. Water samples for incubation were collected from the plumes at stations CTD 4 and CTD 13. The incubation method followed Statham et al. (2005). Before the incubation, an iron II spike (as ferrous ammonium sulfate) was added to increase the iron concentration by ~500 nM without significant change in seawater pH. The incubation bottle was kept in the dark in a 2 °C water bath. The iron concentration over several hours was determined using the colorimetric ferrozine technique of Stookey (1970).

3. Results and discussion

3.1. Hydrothermal plumes indicated from TFe and DMn profiles

After hydrothermal springs at mid-ocean ridges erupt from the seabed, the resultant plumes are enriched in suspended particulate load and dissolved chemical tracers (Baker et al., 1995). Therefore, light transmission and Fe and Mn can be used as tracers of hydrothermal activity (Charlou and Donval, 1993; Klinkhammer et al., 1985; Scheirer et al., 1998). CTD-nephelometer plots of the four stations are shown in Fig. 2. The anomalies generally ranged from 2400 m to 2900 m depth. Larger signals and profiles with multiple peaks were found at the CTD 4 station, with two levels: from 2100 to 2300 m and from 2400 to 2500 m. The strongest plume signals occurred in the lower plume in close vicinity to the seafloor. The weaker

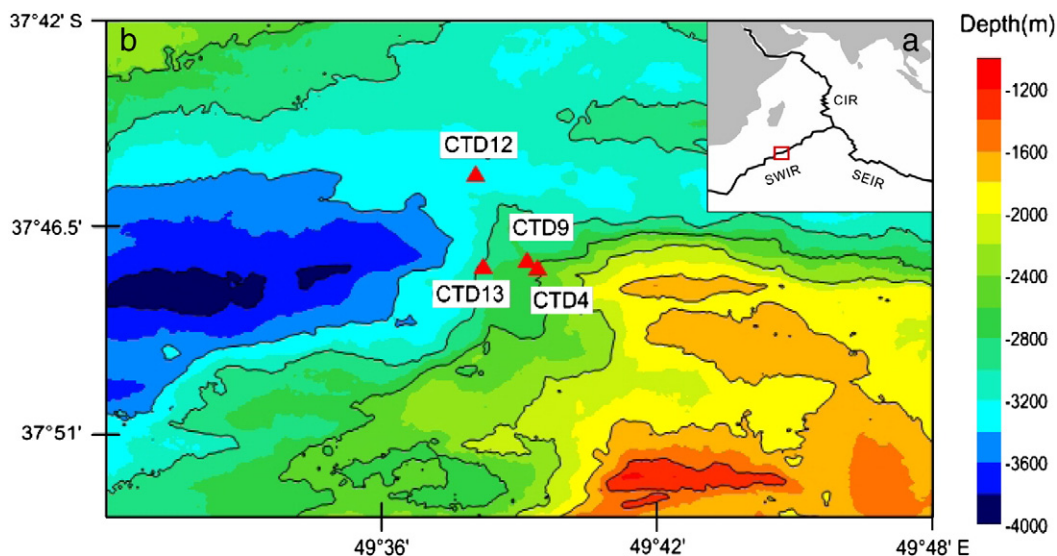


Fig. 1. (a) Sketch of the location of the Southwest Indian Ridge. (b) Bathymetric map of the area within the red box shown in (a), and locations of CTD stations.

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