



## Sulfur and strontium isotopic study of epithermal mineralization: A case study from the SE Afar Rift, Djibouti



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### ABSTRACT

Epithermal mineralization was recently described in synrift volcanic rocks from the SE Afar Rift in Djibouti. To infer fluid sources in this system, coupled sulfur and strontium isotopic analyses were performed on mineralized samples from hydrothermal veins and stockwork zones. Mineralization includes gypsum and chalcedony, and/or quartz ± carbonate containing gold and sulfides. Sulfur isotopic compositions of sulfides and sulfates were determined using Multicollector-Inductively Coupled Plasma Mass Spectrometer (MC ICPMS); Laser Ablation MC ICPMS; and Continuous Flow Elemental Analyzer-Isotope Ratio Mass Spectrometry while strontium isotopes were determined by Thermal Ionisation Mass Spectrometry (TIMS) methods. Sulfur isotopic composition of sulfides (mainly pyrite) varies from −9.2 to +6.6‰. These values are classically reported for hydrothermal sulfides resulting from the mixing of variable S sources such as seawater sulfate and magmatic source. In addition, individual pyrite grains within the same sample show both positive and negative  $\delta^{34}\text{S}$  values suggesting multi-phase mineralization processes. Coupled sulfur and strontium isotopic compositions of seven sulfate samples hosted in (i) gypsum mound, (ii) evaporites, (iii) stockwork veins in sediments or in volcanic rocks were also investigated to characterize the sources of Sr and S. The  $\delta^{34}\text{S}$  values of the sulfate ranged from −0.8 to +14.3‰ while the  $^{87}\text{Sr}/^{86}\text{Sr}$  ranged from 0.70389 to 0.70639 consistent with variable mixing ratios of Sr and S derived from volcanic sources and seawater-derived sulfate. This study shows that, although precious-metal deposition is related to low-sulfidation epithermal environment, both acidic fluid of magmatic and saline origins contributed to the hydrothermal system in the SE Afar Rift.

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### 1. Introduction

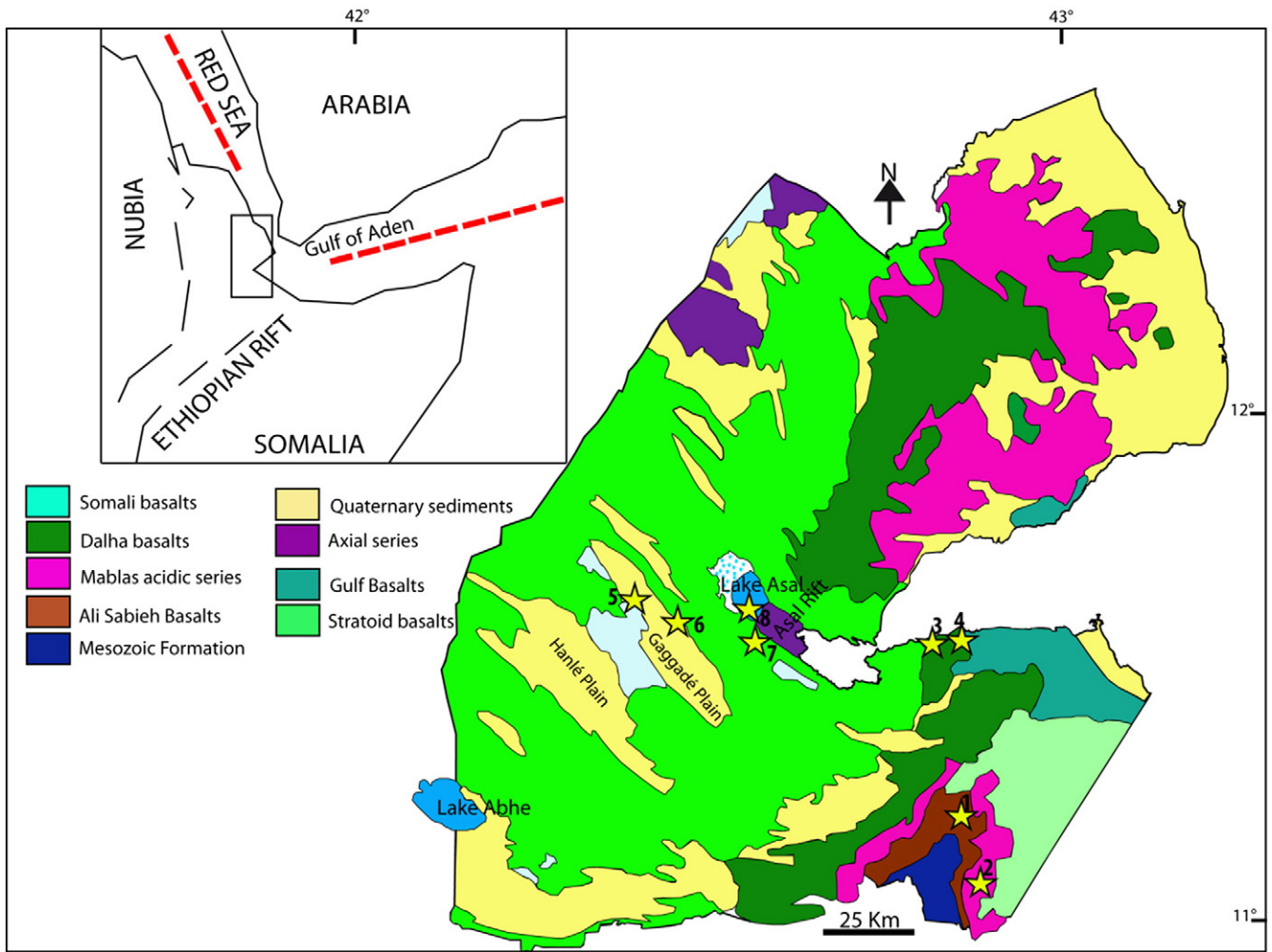
Epithermal deposits are important economic sources of Au and Ag in the world (White and Hedenquist, 1990; Hedenquist et al., 2000; Simmons et al., 2005). Epithermal deposits form in the shallow parts of high-temperature magmatic-hydrothermal systems (Simmons et al., 2005) and are generally grouped into high, intermediate and low sulfidation types based on mineral assemblages and their sulfidation states (e.g., Hedenquist et al., 2000; Sillitoe and Hedenquist, 2003; Einaudi et al., 2003). There has been considerable debate on the origin of hydrothermal fluids leading to the formation of both high and low sulfidation epithermal deposits. Previous studies identified the importance of magmatic fluids while others argued that epithermal deposits form through the circulation of meteoric (sub-surface) fluids, without

a direct relationship to a magmatic source (Henley and Ellis, 1983; Hayba et al., 1985; Simmons et al., 2005). High-sulfidation deposits are considered to form by magmatic fluids (Arribas, 1995) while low sulfidation deposits principally involve meteoric fluids with still significant contribution of magmatic fluids (Simmons, 1995).

Recent field investigations in the SE Afar rift (Djibouti) aimed at addressing the importance of hydrothermal fluids in rift-related volcanic complexes and their relation to the formation of epithermal deposits (Fig. 1) (Moussa et al., 2012). This region has been widely investigated since 1970 as it represents an active geological laboratory to investigate tectono-magmatic rift systems (Stietjes, 1973; Barberi et al., 1975; Le Gall et al., 2010; Pinzutti et al., 2013). The most active area is the Asal Rift which has been recognized as a subaerial analogue of slow-spreading ridges where active tectonics and volcanism can be directly observed. Based on geochemical and field studies, several types of hydrothermal ore deposits were identified in the Djibouti rift system (Moussa et al., 2012). The high abundance of gold in the base metal deposits from Hes Daba and Asa Leyta located nearby the Asal Rift (Fig. 1)

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**Fig. 1.** Schematic geological map of the Republic of Djibouti (SE Afar Triangle) after Vellutini and Piguat (1994). Yellow stars (and corresponding numbers) are the hydrothermal sites studied in this work. 1 = Da'asbiyo 2; 2 = Ali Addé; 3 = Arta Plage; 4 = Arta; 5 = Babba Alou; 6 = Hes Daba; 7 = Asa Leyta; 8 = Asal. In the inset: schematic map of the Afar Depression with the location of Djibouti (black rectangle) and the dashed red lines = Red Sea and Gulf of Aden spreading centers.

provide an ideal analogue, of larger epithermal systems. In this paper, we aim to assess the origin and mixing relationships of the ore fluids in eight different sites from the Asal Rift. We applied coupled sulfur and strontium isotope studies of mineralized veins in the deposits to quantify the sources and interactions of the ore fluids with the source rocks and marine sulfate deposits.

Sulfur isotopic compositions have been widely used to investigate sulfide and sulfate deposition processes, and as tracers of sulfur sources (e.g. volcanic rocks, magmatic volatiles and seawater). Sulfur isotopic compositions of sulfides of epithermal deposits generally yield  $\delta^{34}\text{S}$  values ranging from  $-15$  to  $5\%$  depending on their geological settings: (i)  $-6$  and  $+5\%$  for low-sulfidation with most values close to  $0\%$  (Ohmoto and Rye, 1979; Field and Fifiarek, 1985); (ii)  $-15$  to  $+8\%$  for alkalic epithermal deposits (Richards, 1995) and (iii)  $-10$  to  $+8\%$  for high-sulfidation deposits (Arribas, 1995). Heaviest  $\delta^{34}\text{S}$  values are considered to represent a major contribution of seawater-derived sulfate, while negative  $\delta^{34}\text{S}$  values are best explained by magmatic fluid contribution. In modern seawater, sulfur is found in an oxidized form as sulfate with an homogeneous  $\delta^{34}\text{S}$  composition of  $21 \pm 0.2\%$  (Rees et al., 1978), although this value is known to have varied from  $10$  to  $30\%$  during the Phanerozoic (e.g. Claypool et al., 1980; Paytan et al., 1998). Magmatic rocks derived from the upper mantle have sulfur isotopic composition similar to that of meteorites ( $\delta^{34}\text{S} \approx 0 \pm 2\%$ )

(Thode et al., 1961; Sakai et al., 1984), although recent data have pointed to significant heterogeneity of  $\delta^{34}\text{S}$  values in the mantle (Labidi et al., 2013). Sedimentary sulfides have a wide range of  $\delta^{34}\text{S}$  values ( $-70$  to  $+70\%$ ) (Claypool et al., 1980; Shen et al., 2000; Canfield, 2001) reflecting large S isotope fractionation during microbial sulfate reduction and diagenetic processes. Although sedimentary (i.e. biogenic) contribution may in theory explain the occurrence of isotopically light S isotope signatures in epithermal deposits, the abiotic disproportionation of magmatic  $\text{SO}_2$  is considered to be the main mechanism producing sulfide (i.e.  $\text{H}_2\text{S}$ ) with negative  $\delta^{34}\text{S}$  values.

Sulfur isotopic compositions have been also intensely investigated in seafloor hydrothermal systems located along fast and slow spreading ridges and in back-arc settings (e.g. Arnold and Sheppard, 1981; Alt et al., 1989; Shanks et al., 1995; Rouxel et al., 2004; Ono et al., 2007). These studies demonstrated that along mid-oceanic ridges, seafloor hydrothermal sulfides (active and inactive deposits) commonly yield  $\delta^{34}\text{S}$  values from  $-1$  to  $+11\%$ . These values are explained by a major contribution of sulfur derived from the basalt and a smaller contribution from inorganically reduced seawater sulfate (Arnold and Sheppard, 1981; Shanks et al., 1995). Using multiple S-isotope ratios ( $^{32}\text{S}$ ,  $^{33}\text{S}$  and  $^{34}\text{S}$ ), Ono et al. (2007) have also addressed the importance of partial  $\text{H}_2\text{S}-\text{SO}_4^{2-}$  isotope exchange in hydrothermal chimney environments. The  $\delta^{34}\text{S}$  values for hydrothermal sulfides from sedimented mid-ocean

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