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Systematic variations of trace element and sulfur isotope compositions in pyrite with stratigraphic depth in the Skouriotissa volcanic-hosted massive sulfide deposit, Troodos ophiolite, Cyprus

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ABSTRACT

The Troodos ophiolite represents one of the best-preserved fossil analogs of modern oceanic crust and includes numerous volcanic-hosted massive sulfide deposits. The Skouriotissa deposit can be separated into a stockwork ore zone and an overlying massive sulfide lens that is covered by metalliferous sediments representing the former sulfide–seawater interface. Pyrite is the dominant sulfide mineral within these ores. The trace element composition of pyrite varies systematically with stratigraphic depth (down to ~150 mbsf) probably reflecting fluid temperature variations and effects of phase separation (Co, Ni, Se, Te, Bi and Cu). Metal remobilization due to hydrothermal zone refining (Zn, Sb and Pb) and fluid-seawater mixing at the seafloor (Mo) represent further important processes controlling the pyrite chemistry. Massive sulfide-hosted sphalerite and euhedral pyrite probably formed from hot fluids (~400 °C), while the occurrence of colloform pyrite indicates lower precipitation temperatures (<400 °C). Similar $\delta^{18}O$ quartz-fluid equilibration temperatures (~400 °C) in the stockwork zone suggest that the Skouriotissa fluids did not cool significantly during the final 150 m of fluid ascent to the seafloor. The $\delta^{34}S$ composition of deep stockwork pyrite (−1.4‰) suggests that an isotopically light magmatic volatile phase (<0‰) was added to the hydrothermal system of Skouriotissa. During further fluid ascent about 38% of Cretaceous seawater ($\delta^{34}S$ = 18–19‰) was added leading to the precipitation of stockwork pyrite with positive $\delta^{34}S$ values (6.1‰). In addition, the chemical and textural similarities between Skouriotissa and modern seafloor vent systems and massive sulfide deposits suggest that the Skouriotissa hydrothermal system has a modern analog.
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1. Introduction

During the last decades numerous hydrothermal vent sites have been discovered along mid-ocean ridges (e.g., Tivey et al., 1995; Hannington et al., 2011), back-arc spreading centers (e.g., Fouquet et al., 1993; Herzig et al., 1993) and at island arc volcanoes (e.g., de Ronde et al., 2011; Berkenbosch et al., 2012). The global distribution of these vent sites implies that hydrothermal activity is concentrated along convergent and divergent plate margins that represent zones of intensive magmatic and tectonic activity (Fouquet, 1997; Hannington et al., 2011). The magma ascending beneath spreading ridges represents the heat source that drives the hydrothermal fluid convection through the oceanic crust. The fluid mobilizes metals in the source region and transports these metals to the seafloor where they mainly precipitate as metal sulfides due to mixing of the discharging fluids with ambient seawater (e.g., Alt, 1995). Maturation processes in long-living hydrothermal vent systems result in the formation of large massive sulfide mounds at the seafloor (Petersen et al., 2000). The obduction and preservation of such sulfide mounds in ophiolites lead to the exposure of volcanic-hosted massive sulfide (VHMS) deposits on land (Hannington et al., 1998).

The Cyprus-type VHMS deposits in the Troodos ophiolite represent the fossil analogs of sediment-starved modern black smoker systems occurring at the seafloor along intermediate and slow spreading-ridges (Oudin and Constantinou, 1984; Hannington et al., 1998, 2011). Due to the young age of the Troodos ophiolite (90–92 Ma, Turonian; Mukasa and Ludden, 1987) and the absence of post-depositional high-grade metamorphism primary seafloor-precipitation textures are preserved within the VHMS deposits (Lydon, 1984; Cann and Gillis, 2004). The internal structure of these deposits consists of a massive sulfide lens and an underlying stockwork complex where pyrite is the dominant sulfide mineral. The chemical composition and the textural occurrence of these ores are comparable to those observed in modern seafloor deposits (Heaton and Sheppard, 1977; Oudin and Constantinou, 1984; Hannington et al., 1998). However, stockwork ores are rarely exposed at the seafloor (Embley et al., 1988; Fouquet et al., 1988) and can

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only be recovered by deep ocean drilling (Hannington et al., 1990, 1998). In contrast, ophiolite-hosted sulfide deposits represent a unique opportunity to study ore-forming processes as a function of stratigraphic depth. Recent studies of active and fossil black smoker vents have shown that trace elements in pyrite and in other sulfide minerals provide important information on the physicochemical fluid conditions during sulfide precipitation (Maslennikov et al., 2009; Wohlgemuth-Ueberwasser et al., 2015; Keith et al., 2016). The δ^{34}S composition of hydrothermal sulfides can be used to determine the S source and provides information on processes that control the distribution and speciation of S in hydrothermal systems (Peters et al., 2010; McDermott et al., 2015). Hence, physicochemical changes in the ascending hydrothermal fluids can result in trace element and S isotope variations in pyrite reflecting different stratigraphic levels within a VHMS deposit.

Here we present trace element and S isotope data of pyrite from stockwork and massive sulfide samples of the Skouriotissa VHMS deposit from the Troodos ophiolite, Cyprus. Our results indicate that the trace element composition of pyrite is mainly a function of fluid temperature, phase separation and hydrothermal zone refining causing systematic variations with stratigraphic depth and textural appearance. The δ^{34}S composition of pyrite varies due to a variable S contribution of seawater and magmatic origin.

2. Geological setting and sample localities

The Troodos ophiolite is located in the eastern Mediterranean on the island of Cyprus. The oceanic crust of the ophiolite formed during the Cretaceous (90–92Ma, Turonian; Mukasa and Ludden, 1987) in the Tethys ocean at a spreading center that was affected by a subduction component (e.g., Pearce and Robinson, 2010; Regelous et al., 2014). Ultramafic mantle rocks, plutonic rocks, sheeted dikes, lavas and marine sediments comprise a complete sequence of oceanic lithosphere (Fig. 1, Gass, 1968). The Troodos VHMS deposits are situated within the extrusive lava unit (Oudin et al., 1981; Hannington et al., 1998). Most of the large sulfide deposits are located along the northern flank of the ophiolite associated with three north–south striking rift structures expressed by deep-seated detachment faults representing fossil upflow zones of high temperature hydrothermal fluids (Schiffman and Smith, 1988; Bettison-Varga et al., 1992; Booij et al., 2000). Epidosites occur in these upflow zones and represent base metal-depleted epidote- and quartz-rich rocks formed by hydrothermal alteration of sheeted dikes during fluid ascent. It is assumed that the protolithic sheeted dikes represent the metal source for the overlying sulfide deposits (Schiffman and Smith, 1988; Bettison-Varga et al., 1992; Booij et al., 2000).

The Solea graben (Fig. 1) represents the oldest and best-exposed rift structure within the Troodos ophiolite (Varga and Moores, 1985). Skouriotissa (35°5.331′N/32°53.309′E, Fig. 1) is one of the largest and most Cu-rich VHMS deposits (on average 2.3 wt.% Cu; Hannington et al., 1998) on Cyprus and is situated near the rift axis of the Solea graben (Bettison-Varga et al., 1992; Hannington et al., 1998; Schiffman and Smith, 1988). Field observations revealed that the stockwork complex represents the major zone of Cu mineralization as indicated by gray to blue rock colors due to secondary minerals in this region (Fig. 2A). A massive sulfide lens is exposed above the stockwork mineralization zone in the upper part of the sulfide deposit (Fig. 2B), which is covered by metalliferous sediments (Fig. 2C), umber and ocher. The upper part of the deposit, composed of massive sulfides and metalliferous sediments, is interpreted to be the fossil analog of a sulfide mound as exposed on the modern seafloor (cf. Hannington et al., 1998).

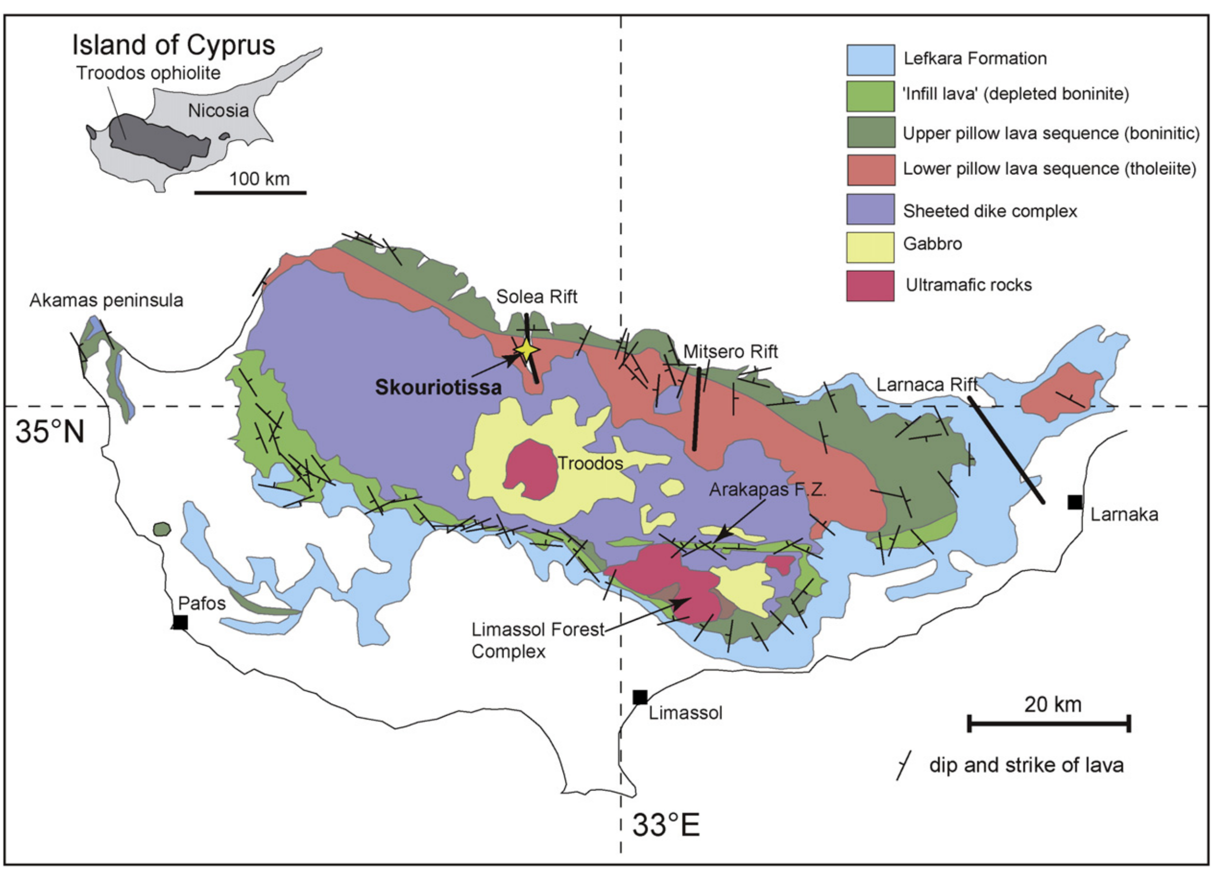

Fig. 1. Simplified map of the Troodos ophiolite, modified after Osozawa et al. (2012). The magmatic rocks are of Late Cretaceous age (90–92 Ma, Turonian; Mukasa and Ludden, 1987), while the carbonate sedimentation represented by the Lefkara formation started during Maastrichtian time (66–72 Ma; Robertson, 1977). Abbreviations: F. Z.=fracture zone.

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