



# Contrasting hydrological processes of meteoric water incursion during magmatic–hydrothermal ore deposition: An oxygen isotope study by ion microprobe



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## ABSTRACT

Meteoric water convection has long been recognized as an efficient means to cool magmatic intrusions in the Earth's upper crust. This interplay between magmatic and hydrothermal activity thus exerts a primary control on the structure and evolution of volcanic, geothermal and ore-forming systems. Incursion of meteoric water into magmatic–hydrothermal systems has been linked to tin ore deposition in granitic plutons. In contrast, evidence from porphyry copper ore deposits suggests that crystallizing subvolcanic magma bodies are only affected by meteoric water incursion in peripheral zones and during late post-ore stages. We apply high-resolution secondary ion mass spectrometry (SIMS) to analyze oxygen isotope ratios of individual growth zones in vein quartz crystals, imaged by cathodo-luminescence microscopy (SEM-CL). Existing microthermometric information from fluid inclusions enables calculation of the oxygen isotope composition of the fluid from which the quartz precipitated, constraining the relative timing of meteoric water input into these two different settings. Our results confirm that incursion of meteoric water directly contributes to cooling of shallow granitic plutons and plays a key role in concurrent tin mineralization. By contrast, data from two porphyry copper deposits suggest that downward circulating meteoric water is counteracted by up-flowing hot magmatic fluids. Our data show that porphyry copper ore deposition occurs close to a magmatic–meteoric water interface, rather than in a purely magmatic fluid plume, confirming recent hydrological modeling. On a larger scale, the expulsion of magmatic fluids against the meteoric water interface can shield plutons from rapid convective cooling, which may aid the build-up of large magma chambers required for porphyry copper ore formation.

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## 1. Introduction

The depth ranges and timescales of the intrusion of magmatic bodies into the Earth's crust prohibit direct monitoring and quantification. Geological studies of extinct and exposed fossil intrusions indicate that differences in pluton assembly (Annen et al., 2015), magma injection rates and volumes (Caricchi et al., 2014) and duration of magmatic activity (Michel et al., 2008; von Quadt et al., 2011; Bachmann et al., 2012) can result in different magmatic and associated hydrothermal systems, such as the formation of upper crustal plutons or large volcanic eruptions (Caricchi et al.,

2014). The formation of giant magmatic–hydrothermal ore deposits, including porphyry copper deposits, requires large magma reservoirs (Dilles, 1987; Cline and Bodnar, 1991; Steinberger et al., 2013), but it remains unclear whether pluton-sized volumes of magma are emplaced rapidly (Dilles, 1987; Glazner et al., 2004), or whether plutons are amalgamated from several smaller magma bodies that intruded episodically into the crust (Halter et al., 2005; Michel et al., 2008). Rate and mode of magma injection determine how long a pluton takes to crystallize, together with the rate of heat loss from the magma reservoir. In the permeable upper crust, meteoric water convection contributes significantly to pluton cooling (Cathles, 1977; Hayba and Ingebritsen, 1997; Gerdes et al., 1998) and is therefore expected to influence the lifetime, physical make-up and petrological evolution of upper-crustal

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magma reservoirs (Spera and Bohrsen, 2001; Miller et al., 2007; Schaltegger et al., 2009). The role of meteoric water in hydrothermal ore formation has been variably interpreted as a main agent for metal transport and ore deposition (Sheppard et al., 1971; Taylor, 1971, 1974; Cathles, 1977), as a diluent for metal-rich magmatic saline liquids triggering chemical precipitation of ore minerals (Eastoe, 1978; Dilles, 1987; Sun and Eadington, 1987), or as a dominantly physical agent for cooling magmatic ore fluids (Sillitoe, 1973; Henley and McNabb, 1978; Burnham and Ohmoto, 1980; Hedenquist and Lowenstern, 1994).

Quartz veins in mineral deposits record the compositions of fluids during the thermal history of a magmatic–hydrothermal system. At the Yankee Lode tin deposit (Mole Granite, Australia; Audétat et al., 1998), fluid pressures determined from co-existing vapor and saline liquid inclusions constrain the paleo-depth to  $2.5 \pm 1$  km and document a transition from an early magmatic regime at lithostatic fluid pressure to a cooler hydrostatic environment. This change led to incursion of meteoric water and cassiterite precipitation as the result of mixing between the meteoric water and ascending hot, saline metal-bearing magmatic liquid, as indicated by fluid inclusion LA-ICP MS trace element analyses (Audétat et al., 1998, 2008). Regional-scale oxygen isotope measurements of hydrothermal quartz from deposits distributed around the Mole Granite, using conventional analyses, show an evolution from early magmatic fluids in pegmatitic veins within the roof of the granite ( $\delta^{18}\text{O} \cong 9\text{‰}$ ), through mixing with meteoric water in the tin stage, to purely meteoric waters ( $\delta^{18}\text{O} \cong -15\text{‰}$ ) during the late stage base metal mineralization in distal veins hosted by sedimentary wall rocks (Sun and Eadington, 1987).

Porphyry Cu systems often form at similar depths to Sn deposits (~2–3 km) but their roots are voluminous plutons located at depths of 5 km or more (Sillitoe, 2010). Due to the complexity of copper ore deposition and superimposed and overprinting alteration events, the mechanism of metal precipitation from the mineralizing fluids remains a matter of discussion (Sheppard et al., 1971; Gustafson and Hunt, 1975; Eastoe, 1978; Hemley et al., 1992; Hedenquist and Lowenstern, 1994; Redmond et al., 2004; Landtwing et al., 2005; Sillitoe, 2010; Heinrich and Candela, 2014; Blundy et al., 2015). The currently prevailing theory favors precipitation of ore minerals from magmatic fluids expelled from cooling upper crustal magma chambers (Richards, 2011). In this model, meteoric water incursion only affects peripheral and post-ore alterations, whereas main-stage mineralization and associated potassic alteration is thought to be dominated by magmatic fluids (Gustafson and Hunt, 1975; Bowman et al., 1987; Sillitoe, 2010). Based on stable oxygen and hydrogen isotope analyses of alteration minerals from porphyry copper deposits, meteoric water incursion affects sericitic, argillic and propylitic alterations (Sheppard et al., 1971; Taylor, 1974, 1997; Bowman et al., 1987; Hedenquist and Lowenstern, 1994; Sillitoe, 2010). A significant meteoric water component in the early quartz–K-feldspar–biotite–chalcopyrite precipitating fluid has been inferred for the Butte porphyry copper deposit and a few Tertiary deposits intruded into flat-lying, permeable volcanic host rocks (Sheppard et al., 1971; Sheppard and Taylor, 1974; Taylor, 1974). Also, petrographic evidence from the Far Southeast porphyry copper deposit (Philippines) shows that Cu sulfide mineralization can be associated with the transition from potassic to sericitic alteration (Hedenquist et al., 1998). Oxygen and hydrogen isotope analyses of white mica and biotite from the Far Southeast and the El Salvador (Chile) porphyry copper deposits indicate admixing of 10–20% of meteoric water to a dominantly magmatic–hydrothermal fluid associated with copper precipitation (Watanabe and Hedenquist, 2001).

Fluid inclusion studies (Landtwing et al., 2010; Stefanova et al., 2014) following textural mapping of quartz vein samples by Scanning Electron Microscopy Cathodo-Luminescence (SEM-CL) indicate

that copper precipitated after deposition of most vein quartz at temperatures of 450–350 °C (Rusk and Reed, 2002; Redmond et al., 2004; Landtwing et al., 2005). Such low temperatures cannot be attained by adiabatic cooling of magmatic fluids exsolved from a magma chamber and require an external cooling agent (Henley and McNabb, 1978; Fournier, 1999). Numerical simulations suggest that cooling is controlled by heat loss of a magmatic fluid plume to convecting meteoric water. Copper sulfides precipitate at this interface, which is characterized by a sharp drop in fluid pressure and temperature (Weis et al., 2012). The simulation results are in line with previous interpretations of the evolution from a central potassic to the outer and partly later sericitic alteration zone, corresponding to the transition from a lithostatically pressured magmatic fluid regime to a hydrostatically pressured fluid mixture dominated by meteoric water (Gustafson and Hunt, 1975).

Microtextural and fluid inclusion studies at porphyry copper deposits show that sulfide precipitation during the main ore-stage is commonly characterized by a lack of quartz precipitation and quartz re-dissolution (Rusk and Reed, 2002; Redmond et al., 2004; Landtwing et al., 2005; Stefanova et al., 2014). Retrograde quartz solubility occurs in a region of low pressure just above the critical temperature of pure water (Kennedy, 1950), whereas at higher pressure or in single-phase fluids of higher salinity quartz solubility increases monotonously with temperature (Fournier, 1983). An empirical fit of experimental quartz solubility in the  $\text{H}_2\text{O}$ – $\text{NaCl}$ – $\text{SiO}_2$  system over a wide range of density, salinity and pressure (Akinfiyev and Diamond, 2009) provides a basis for evaluating bulk quartz solubility in vapor and liquid mixtures, to interpret micro-textural observation of quartz precipitation and dissolution. In the present study, we map texturally-controlled variations of oxygen isotope compositions of hydrothermal vein quartz using secondary ion mass spectrometry (SIMS), to interpret these characteristic quartz textures and quantify the variations of meteoric and magmatic fluid contribution during the evolution of magmatic–hydrothermal ore veins.

## 2. Methods

We obtained a total of 298 in-situ  $\delta^{18}\text{O}$  measurements of vein quartz from individual zones identified on the basis of variations in CL intensity. The  $\delta^{18}\text{O}$  values of the associated fluid was calculated using fluid inclusion temperatures. We used doubly polished sections of vein quartz from previous studies (Audétat et al., 1998; Landtwing et al., 2005; Stefanova et al., 2014) in which detailed and texturally controlled fluid inclusion data had been obtained that allow reconstructing the precipitation temperature of individual quartz growth stages. Textural control was ensured by SEM-CL imaging. High-resolution, in-situ  $\delta^{18}\text{O}$  analyses of individual growth stages were then obtained by secondary ion microprobe.

Sample preparation of the polished chips required a special mounting technique by pressing the chips into indium in the sample holder. In this way we could ensure a perfectly horizontal mounting and avoid re-polishing, which might have compromised the precise correlation between previously established microthermometric data of thin, individual growth zones and healed fractures and the oxygen isotope spot analysis targeting these zones.

$^{18}\text{O}/^{16}\text{O}$  ratios were measured using the SwissSIMS Cameca IMS 1280HR at the Institute of Earth Sciences of the University of Lausanne (Switzerland). We used a 10 kV  $\text{Cs}^+$  primary ion beam, a ~2 nA current, resulting in a ~10  $\mu\text{m}$  beam size. The electron flood gun was used to compensate charges.  $^{16}\text{O}$  and  $^{18}\text{O}$  secondary ions were accelerated by 10 kV and analyzed at 3000 in FC multi-collection mode. Faraday cups are calibrated at the beginning of the session. Mass calibration was performed at the beginning of each session and every 12 h. Each analysis took less than 4 min, including pre-sputtering (30 s) and automated centering of sec-

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