



Calibration and application of silica-water triple oxygen isotope thermometry to geothermal systems in Iceland and Chile

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Abstract

Triple oxygen isotope analyses were made on geothermal fluids and precipitates from Chile and Iceland to calibrate the silica-water isotopic fractionation for abiogenic silica formation at elevated temperatures and were used to evaluate potential fractionation effects of biogenic vs. abiogenic samples and polymorphism. Coexisting water and amorphous silica precipitated inside the heat exchanger of the Hellisheiði power plant at 60 and 118 °C have triple oxygen isotope fractionations in excellent agreement with previous results from analyses of biogenic silica precipitated in cold waters.

In contrast to samples from the geothermal plant, natural amorphous silica precipitates and waters formed in active hot springs ($T = 63\text{--}84\text{ °C}$) in the Puchuldiza geothermal area of northern Chile gave temperature estimates from the silica-water thermometer far lower ($37\text{--}46\text{ °C}$) than the measured water temperatures. Active silica precipitation was found to only occur at and above the air-water interface on glass slides placed in the hot spring waters for 9 days. The calculated temperatures and visual inspection suggest that precipitation occurred along channel edges when saturation was overstepped by a factor of two. In contrast to the surficial neofomed amorphous silica, subsurface silica samples ($>10\text{ cm}$) have recrystallized to opal-CT and quartz within a sinter mound and these samples preserve isotope temperatures of 82 °C and 89 °C, in good agreement with the ambient temperatures of the thermal spring conduit system. The $\delta^{18}\text{O}$ values of abiogenic, low temperature silica formed in spring water far from the thermal waters with a measured temperature of 19 °C correspond to a silica-water temperature estimate of 20 °C. All samples preserved isotope data corresponding to their expected formation temperatures and appear to be in equilibrium in the triple oxygen isotope system. A best-fit $\theta\text{--}T$ relationship for silica-water using our inorganic silica-water samples is $\theta = 0.5305 - \frac{1.82(\pm 0.02)}{T(K)}$, $R^2 = 0.998$ (where $\theta_{a-b} = \frac{\ln z^{17} O_{a-b}}{\ln z^{18} O_{a-b}}$). This new equation is indistinguishable from a previous empirical fit by Sharp et al. (2016) based primarily on biogenic silica samples, suggesting that the biogenic and abiogenic samples secreted silica with the same fractionation. Our results show that triple oxygen isotope measurements are robust and can be used

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to estimate the temperature of formation, the isotopic composition of the formation water, and discern between equilibrium and non-equilibrium processes.

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1. INTRODUCTION

1.1. Geothermal Systems

Siliceous geothermal systems precipitate amorphous silica at the surface. The presence of siliceous springs indicates extensive water–rock interaction at depth. The maximum amount of dissolved silica in a fluid is controlled by quartz solubility when temperature is greater than 180 °C and chalcedony solubility for fluid temperatures below 110 °C (Arnórrsson, 1975). As the geothermal spring water cools and becomes supersaturated, deposition of siliceous sinter (hereafter referred to as “sinter”) occurs. Temperature is thought to be the main factor governing silica precipitation, while evaporation has an indirect effect by changing silica concentration in the fluid (e.g. Jones et al., 2000; Guidry and Chafetz, 2002; Mountain et al., 2003; Tobler et al., 2008; Nicolau et al., 2014). Sinter deposits can be composed of multiple silica phases including amorphous silica, opal cristobalite and/or cristobalite with tridymite (opal C/CT), and quartz (Herdianita et al., 2000; Campbell et al., 2001; Lynne and Campbell, 2003, 2004; Rodgers et al., 2004; Lynne et al., 2005, 2006, 2007, 2008; García-Valles et al., 2008; Nicolau et al., 2014). Initial deposition is always as amorphous silica while diagenesis to the crystalline, more stable, phases is thought to occur post-deposition with high-temperature fluid and/or steam (Lynne and Campbell, 2004). Sinter normally forms in near-neutral pH waters, with a high silica content, and variable anion and cation concentrations (Nicolau, 2013 and references therein) or as a residue of re-precipitated silica present in an acidic fumarole (White et al., 1956, Rodgers et al., 2004). Macroscopic silica textures are informally called lily pads, mounds (geyserite), rimming, terraces, and discharge aprons that are loosely related to temperature (Cady and Farmer, 1996; Lynne, 2012). Many of these textures are maintained even when the spring is no longer active and they can survive diagenesis (Lynne, 2012; Lynne and Campbell, 2004), making fossil sinter deposits (hereafter referred to as “paleosinter”) an excellent resource for tracking hydrological conditions of the geothermal system. Similar to hot springs, precipitation of silica occurs inside geothermal power plants upon cooling of the water. These deposits are called silica scales and have been widely investigated due to their detrimental effect on the efficiency of geothermal energy production (e.g. Rothbaum et al., 1979; Harrar et al., 1982; Inagaki et al., 2003; Gunnarsson and Arnórrsson, 2003, 2005; Padilla et al., 2005; Meier et al., 2014; Mroczek et al., 2017).

Both siliceous springs and geothermal power plants offer a unique site for the study of silica-water precipitation in terms of oxygen isotope fractionation. Geothermal power

plants provide a controlled environment where the water flow and temperature are nearly constant, reducing the number of unconstrained variables when looking to calibrate or validate silica-water fractionation factors (Kita et al., 1985). Siliceous hot springs are natural systems that can be used to test the conditions at which silica precipitates in a natural setting. Many studies have looked at the $^{18}\text{O}/^{16}\text{O}$ ratio of geothermal water (e.g., Arnórrsson, 1975; Giggenschbach, 1978; Guidry and Chafetz, 2002; Geilert et al., 2015; Pope et al., 2010, 2016) or silica formed in geothermal water (e.g., Murata et al., 1977; Hayashi, 2013), but only two studies, to our knowledge, have measured both the water and the silica in either a geothermal power plant (Kita et al., 1985) or siliceous hot springs (Sharp et al., 2016). Geothermal systems have the potential to provide equilibrium isotope partitioning information over a temperature range that is difficult to duplicate in the laboratory (34–250 °C).

1.2. Triple oxygen isotope system

The $\delta^{17}\text{O}$ value of most terrestrial materials are about half the $\delta^{18}\text{O}$ value. A plot of $\delta^{17}\text{O}$ vs. $\delta^{18}\text{O}$ for rock samples plot with a slope of ~ 0.524 (Miller et al., 1999), whereas global meteoric waters plot with a slope of ~ 0.528 (Luz and Barkan, 2010). Waters with a $\delta^{18}\text{O}$ value greater than -10‰ plot with a slope of ~ 0.5275 (Luz and Barkan, 2010; Li et al., 2015; Sharp et al., 2016). The standard δ -notation (McKinney et al., 1950) to describe the isotopic abundance ratio of a sample is defined as

$$\delta^x\text{O} = \left(\frac{\left(\frac{x\text{O}}{^{16}\text{O}} \right)_{\text{sample}}}{\left(\frac{x\text{O}}{^{16}\text{O}} \right)_{\text{VSMOW}}} - 1 \right) \times 1000, \quad (1)$$

where x is either mass 17 or 18. A plot of $\delta^{18}\text{O}$ vs. $\delta^{17}\text{O}$ of terrestrial samples has a near perfect relationship with an R^2 value of 0.999 (Clayton, 1993; Rumble et al., 2007) and is called the Terrestrial Fractionation Line (TFL). Although deviations from the TFL can be tens of per mil in extraterrestrial samples (Clayton et al., 1973) or stratospheric samples resulting from mass-independent fractionation driven by photochemical reactions (Thiemens and Heidenreich, 1983), most natural materials follow the simple linear $\delta^{17}\text{O}$ - $\delta^{18}\text{O}$ relationship with $\pm 0.1\text{‰}$. For most terrestrial geologic samples, the information provided by $\delta^{17}\text{O}$ values appears to add nothing to that obtained by $\delta^{18}\text{O}$ values alone. However, with improved analytical techniques, it is now recognized that subtle departures from the TFL exist in most terrestrial materials, and these departures can have geological significance such as formation temperatures and the extent of water–rock interaction (Pack and Herwartz, 2014; Herwartz et al., 2015).

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