



Texture-specific Si isotope variations in Barberton Greenstone Belt cherts record low temperature fractionations in early Archean seawater

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Abstract

Sedimentary cherts are unusually abundant in early Archean (pre-3.0 Ga) sequences, suggesting a silica cycle that was profoundly different than the modern system. Previously applied for the purpose of paleothermometry, Si isotopes in ancient cherts can offer broader insight into mass fluxes and mechanisms associated with silica concentration, precipitation, diagenesis, and metamorphism. Early Archean cherts contain a rich suite of sedimentological and petrographic textures that document a history of silica deposition, cementation, silicification, and recrystallization. To add a new layer of insight into the chemistry of early cherts, we have used wavelength-dispersive spectroscopy and then secondary ion mass spectrometry (SIMS) to produce elemental and Si and O isotope ratio data from banded black-and-white cherts from the Onverwacht Group of the Barberton Greenstone Belt, South Africa. This geochemical data is then interpreted in the framework of depositional and diagenetic timing of silica precipitation provided by geological observations. SIMS allows the comparison of Si and O isotope ratios of distinct silica phases, including black carbonaceous chert beds and bands (many including well-defined sedimentary grains), white relatively pure chert bands including primary silica granules, early cavity-filling cements, and later quartz-filled veins. Including all chert types and textures analyzed, the $\delta^{30}\text{Si}$ dataset spans a range from -4.78‰ to $+3.74\text{‰}$, with overall mean 0.20‰ , median 0.51‰ , and standard deviation 1.30‰ ($n = 1087$). Most samples have broadly similar $\delta^{30}\text{Si}$ distributions, but systematic texture-specific $\delta^{30}\text{Si}$ differences are observed between white chert bands (mean $+0.60\text{‰}$, $n = 750$), which contain textures that represent primary and earliest diagenetic silica phases, and later cavity-filling cements (mean -1.41‰ , $n = 198$). We observed variations at a $\sim 100\ \mu\text{m}$ scale indicating a lack of Si isotope homogenization at this scale during diagenesis and metamorphism, although fractionations during diagenetic phase transformations may have affected certain textures. We interpret these systematic variations to reflect fractionation during silica precipitation as well as isotopically distinct fluids from which later phases originated. SIMS $\delta^{18}\text{O}$ values fall in a range from 16.39‰ to 23.39‰ ($n = 381$), similar to previously published data from bulk gas source mass spectrometry of Onverwacht cherts. We observed only limited examples of texture-related variation in $\delta^{18}\text{O}$ and did not observe correlation of $\delta^{18}\text{O}$ with $\delta^{30}\text{Si}$ trends. This is consistent with hypotheses that Si isotope ratios are more resistant to alteration under conditions of rock-buffered diagenesis (Marin-Carbonne et al., 2011). Our results indicate that low temperature processes fractionated silicon isotopes in early Archean marine basins, a behavior that probably precludes the application of chert $\delta^{30}\text{Si}$ as a robust paleothermometer. The values we observe for facies that sedimentological and petrographic observations indicate formed as primary and earliest diagenetic silica precipitates from seawater are more ^{30}Si -rich than that expected for bulk silicate Earth. This is consistent with the hypothesis that the silicon isotope budget is balanced by the coeval deposition of ^{30}Si -enriched cherts and ^{30}Si -depleted iron formation lithologies.

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Precipitation of authigenic clay minerals in both terrestrial and marine settings may have also comprised a large ^{30}Si -depleted sink, with the corollary of an important non-carbonate alkalinity sink consuming cations released by silicate weathering.
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1. INTRODUCTION

The apparent overabundance of sedimentary chert is a signal of the early Archean geological record that is thought to reflect the absence of biological controls on the early silica cycle (Siever, 1957, 1992; Maliva, 2001; Maliva et al., 1989, 2005; Perry and Lefticariu, 2003). The Phanerozoic silica cycle was dominated by biologically-mediated precipitation (Maliva et al., 1989; Siever, 1991), so it can be understood as a variation on the modern cycle (Treguer et al., 1995), wherein the evolution and ecology of silicifying organisms played a major role in the distribution of sedimentary chert. The Precambrian silica cycle was dominated by processes of chemical precipitation (Siever, 1957, 1992; Perry and Lefticariu, 2003); in the absence of biological precipitation, early diagenetic replacement of shallow tidal flat carbonates was a common mode of silica precipitation in late Archean and Proterozoic carbonate sequences, likely driven by evaporative processes (Maliva, 2001; Maliva et al., 1989, 2005). However, early Archean sedimentary successions, such as the Barberton Greenstone Belt (3.5–3.2 Ga), South Africa, contain abundant sedimentary cherts that capture different modes of silica precipitation; these cherts occur throughout a variety of paleoenvironments and include some forms that were likely primary precipitates in addition to diagenetically silicified siliclastic or organic-rich sedimentary rocks, volcanoclastic units, and silicified volcanic flow tops (Knauth, 1994; Lowe, 1999). These cherts also present a major mass balance problem in early Archean time. The silica must have been ultimately sourced from silicate weathering, but what of the corresponding alkalinity sink for cations also released during weathering and concentrated in seawater? Was it in salts precipitated from seawater or did the precipitation of authigenic clay minerals also play an important role? In addition to constraining the early silica cycle, a better understanding of Si sinks from Archean seawater also provides insight into the fundamental mass balance between weathering and the production of sedimentary rocks (Milliken et al., 2009), and the possible operation of a silicate-weathering feedback on the early Earth (Walker et al., 1981).

Previous studies have found that trace and rare earth element (REE) geochemistry of early Archean sedimentary cherts are consistent with a strong hydrothermal source (Paris et al., 1985; Kato and Nakamura, 2003; van den Boorn et al., 2010). This can be interpreted in two ways: (1) that cherts are exhalative deposits formed adjacent to hydrothermal vents, or (2) that silica was precipitated from seawater that carried a “hydrothermal” signature. In other words, did hydrothermal fluids interact with ambient seawater before precipitating silica? Answering this question is only a first step toward understanding the Archean silica cycle. If all Archean cherts formed directly from

hydrothermal fluids that had not reacted with seawater, we could stop there. But if fluid interactions did occur, we must also ask how silica is partitioned during these interactions: were there other important silica inputs, what reactions occurred in seawater, and what were the principal silica outputs? These questions have been difficult to resolve with traditional trace and REE geochemical analyses.

Silicon isotopes offer a useful tool for studying the fluxes and processes of the Archean silica cycle. Though the mechanics of the fractionating processes are still not well understood, it is generally observed that silica precipitates are depleted in the heavy isotope of silicon (^{30}Si) relative to the residual fluid by 1–2‰ (Ding et al., 1996; Basile-Doelsch et al., 2005; Fripiat et al., 2007) and kinetic Si isotope fractionations dominate in low temperature Earth surface environments (Ziegler et al., 2005a,b; Georg et al., 2007b, 2009; Delstanche et al., 2009); it is reasonable to suspect that similar kinetic fractionations occurred in Archean marine basins.

Previous studies have used chert $\delta^{30}\text{Si}$ to estimate seawater $\delta^{30}\text{Si}$ composition (Robert and Chaussidon, 2006; van den Boorn et al., 2007, 2010; Abraham et al., 2011), but this required assuming a simple isotope mass balance model (cf. Fig. S2 in Robert and Chaussidon, 2006) in which silica inputs from crustal weathering are balanced by the outputs of hydrothermal silicification and amorphous silica precipitation from seawater. Only two significant isotope fractionations were considered in this simplified framework: the preferential incorporation of light silica into precipitates during (1) hydrothermal and (2) seawater silica precipitation. Robert and Chaussidon (2006) hypothesized that chert $\delta^{30}\text{Si}$, considered to be a seawater silica precipitate and a measure of seawater $\delta^{30}\text{Si}$, was primarily controlled by the temperature gradient between hydrothermal fluids and ambient seawater, with cooler seawater, for example, resulting in a larger fraction of silica output as isotopically light “hydrothermal” silica, leaving the “seawater” silica fraction relatively heavier. Several authors (van den Boorn et al., 2007, 2010; Abraham et al., 2011; Marin-Carbonne et al., 2012) have interpreted the range of $\delta^{30}\text{Si}$ in their chert samples to reflect mixing of isotopically distinct seawater and hydrothermal fluids incorporated into cherts via primary precipitation and/or silicification. The implicit assumption in these interpretations is that seawater Si was not isotopically fractionated by other processes and does not have any additional fractionating sinks. If, as observed in the modern cycle, additional fractionations occurred, then the framework becomes more complex, with the corollary that chert $\delta^{30}\text{Si}$ need not reflect the balance of hydrothermal and seawater precipitates and cannot be uniquely inverted for temperature. In support of this possibility, Chakrabarti et al. (2012) observed an isotopic contrast between peritidal chert and basinal banded iron

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