



Desilication in Archean weathering processes traced by silicon isotopes and Ge/Si ratios



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ABSTRACT

The influence of continental weathering on oceanic seawater compositions in the Precambrian is poorly understood. Here we combine Si isotopes and Ge/Si ratios of a Mesoarchean paleosol (~2.95 Ga) and overlying shales as proxies for weathering processes and Si mass transfer at the early Earth's surface. Results show that, as with modern soils, neoformation of secondary clay minerals in the paleosol was associated with the fractionation of Si isotopes and Ge/Si ratios in response to chemical weathering and soil desilication. Furthermore, the loss of Fe-bearing minerals, most likely Fe-bearing smectites, produced additional controls on Si and Ge mobility, through the coupled dissolution and re-precipitation of clay minerals under reductive conditions. Opposite fractionation behaviors are observed: neoformed secondary clay minerals acted as a ²⁸Si and Ge sink, whereas the leaching of Fe-bearing minerals released ²⁸Si and Ge into soil solutions. An attempt to estimate $\delta^{30}\text{Si}$ and Ge/Si signatures of released dissolved elements into soil solution provided $\delta^{30}\text{Si}$ and Ge/Si signatures of about +0.1‰ and 1.4 $\mu\text{mol/mol}$, respectively. Furthermore, shales deposited shortly after paleosol formation display $\delta^{30}\text{Si}$ and Ge/Si compositions that may be explained as mixtures of the recognized paleosol components. The shale record suggests that weathering-induced desilication and leaching of Fe-bearing minerals, to a lesser extent, might have been widely effective during the Mesoarchean for the transfer of Si from the continents to the hydrosphere. Comparison of Si isotopes recorded in detrital sediments and chemical precipitates over time might thus provide a useful supplementary tool to decipher the impact of putative increases in continental weathering and desilication, relative to changes in hydrothermal and continent-derived solute inputs to ocean and ocean cooling.

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1. Introduction

It is commonly stated that the chemical composition of the ocean in the Archean, as recorded by chemical sedimentary rocks, was more strongly influenced by high-temperature hydrothermal inputs (e.g., black smoker fluids) relative to continent-derived solutes (Veizer et al., 1982; Derry and Jacobsen, 1990; De Wit and Hart, 1993; Allwood et al., 2010). However, the composition of Mesoarchean epicontinental sea deposits indicates a strong impact of solute inputs derived from continental weathering. Y/Ho ratios and Nd isotopic compositions of banded iron formations (BIFs) from the Pongola Supergroup, South Africa, suggest a continent-derived freshwater contribution of about 10–15% to seawater from which the BIFs precipitated

(Alexander et al., 2008, 2009; Delvigne et al., 2012). Heavy Cr isotope compositions and U enrichment in these BIFs provide additional evidence for large Cr and U pools derived from continental weathering (Crowe et al., 2013).

Recent advances in analytical capabilities have added Si isotopes and Ge/Si ratios to the repertoire of tracers used in the study of soil formation processes and biogeochemical evolution of water runoff to the ocean. Relative to crustal rocks displaying little variations (Ge/Si between 1 and 3 $\mu\text{mol/mol}$ and $\delta^{30}\text{Si}$ about $0 \pm 0.4\%$; Douthitt, 1982; Mortlock and Froelich, 1987; Ding et al., 1996; Savage et al., 2012), neoformed clay minerals incorporate preferentially light Si isotopes and Ge relative to Si, resulting in lighter $\delta^{30}\text{Si}$ values (from -2.59 to -0.53% ; Ziegler et al., 2005a; Opfergelt et al., 2010) and higher Ge/Si ratios (4.8–6.3 $\mu\text{mol/mol}$; Kurtz et al., 2002; Cornelis et al., 2010; Lugolobi et al., 2010). The $\delta^{30}\text{Si}$ and Ge/Si values in modern soils are therefore controlled by the relative proportion of primary minerals and neoformed clays, i.e., the degree of weathering (Ziegler et al., 2005a; Opfergelt et al., 2010). Soil Si depletion (desilication) relative to the parent material accompanying the replacement of primary

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minerals by secondary minerals controls the Si isotope compositions of secondary clay fractions (Opfergelt et al., 2012). There is an apparent smaller Si isotope fractionation factor associated with the neof ormation of poorly desilicated clays (e.g., smectite) compared to more desilicated clays (e.g., kaolinite) (Opfergelt et al., 2012). As a result of this Si isotope fractionation process during clay neof ormation, soil-derived dissolved Si conveyed by continental water runoff to the ocean is isotopically heavier than the parent silicate material of the soil (Cardinal et al., 2010; Hughes et al., 2013).

Si isotopes and Ge/Si ratios have not yet been employed as tracers of chemical weathering and runoff chemistry to investigate paleoweathering processes despite the strong advantage that Si isotopes are insensitive to metamorphic resetting (André et al., 2006). The present study aims to fill this gap, through analysis of a set of well-characterized paleosol and shale samples from the ca. 2.95 Ga Pongola Supergroup, Kaapvaal craton, South Africa.

2. Material and methods

A well preserved 2.95 Ga paleoweathering profile was recently identified in a drill core (TSB07-26, 28°16'6.20"S, 031°13'24.33"E) in the Pongola Supergroup (Denny Dalton area, White Mfolozi Inlier, south eastern border of the Kaapvaal craton, South Africa) directly below the unconformity that separates the Mozaan Group from the Nsuze Group (Fig. 1). Note that the profile studied, herein called Denny Dalton paleosol, is younger than the basal Nsuze Group paleosol described by Grandstaff et al. (1986). It was previously investigated, in a different borehole by Nhleko (2003), and later by Crowe et al. (2013), who concluded that it conforms to all five criteria that should identify a paleosol (Rye and Holland, 1998). The Denny Dalton paleosol developed on amygdaloidal basaltic andesite of the Nsuze Group and was subjected

to greenschist-facies metamorphism. Prior to paleosol formation, the Nsuze Group was affected by tilting, so that the paleosol developed on an angular unconformity, with the stratification in the lava flows being developed at an angle of c. 10° (Hicks and Hofmann, 2012). The uppermost part of the altered volcanic rock, to about 190 cm below the paleosurface, consists mainly of sericite occurring with minor chlorite and rutile (Sericite-Dominated Zone, SDZ). Immediately below the SDZ there is a Chlorite-Dominated Zone (CDZ) with quartz, epidote and rutile. A set of nine samples (seven from the drill core and two from surface exposure 28°13'56.5"S, 031°11'28.5"E) were analyzed through the paleosol from the parent andesite to the most altered andesite, immediately below the unconformable surface at the base of the Mozaan Group. In addition, four shale specimens from the Mozaan Group were collected from the same drill core, at a depth ranging from 175 to 170 m (Fig. 1).

Major element concentrations in paleosol and shale samples were analyzed by XRF spectrometry (Philips X'Unique XRFs) at University of KwaZulu-Natal. The accuracy of the major element determinations was checked against international standard NIM-G and was better than 3%.

For Si isotope and Ge/Si ratio determinations, shale samples were milled in agate, while paleosol samples were milled in carbon steel. About 50 mg of powdered rock samples were dissolved by fusion with lithium metaborate flux (99.999% purity; American Element) at 1000 °C for 1 h in platinum crucibles with a 1:8 flux to sample ratio. The fusion beads were dissolved in 5% HNO₃ and the solutions were analyzed for Ge contents. Ge concentrations were analyzed by HR-ICP-MS (Element 2) at RMCA (Belgium) using ⁷⁴Ge isotope in medium-resolution mode with indium (In) as internal standard. Solutions are introduced into the HR-ICP-MS via a glass nebulizer at a flow rate of 1 ml/min, and then vaporized into a chilled cyclonic spray chamber and a quartz

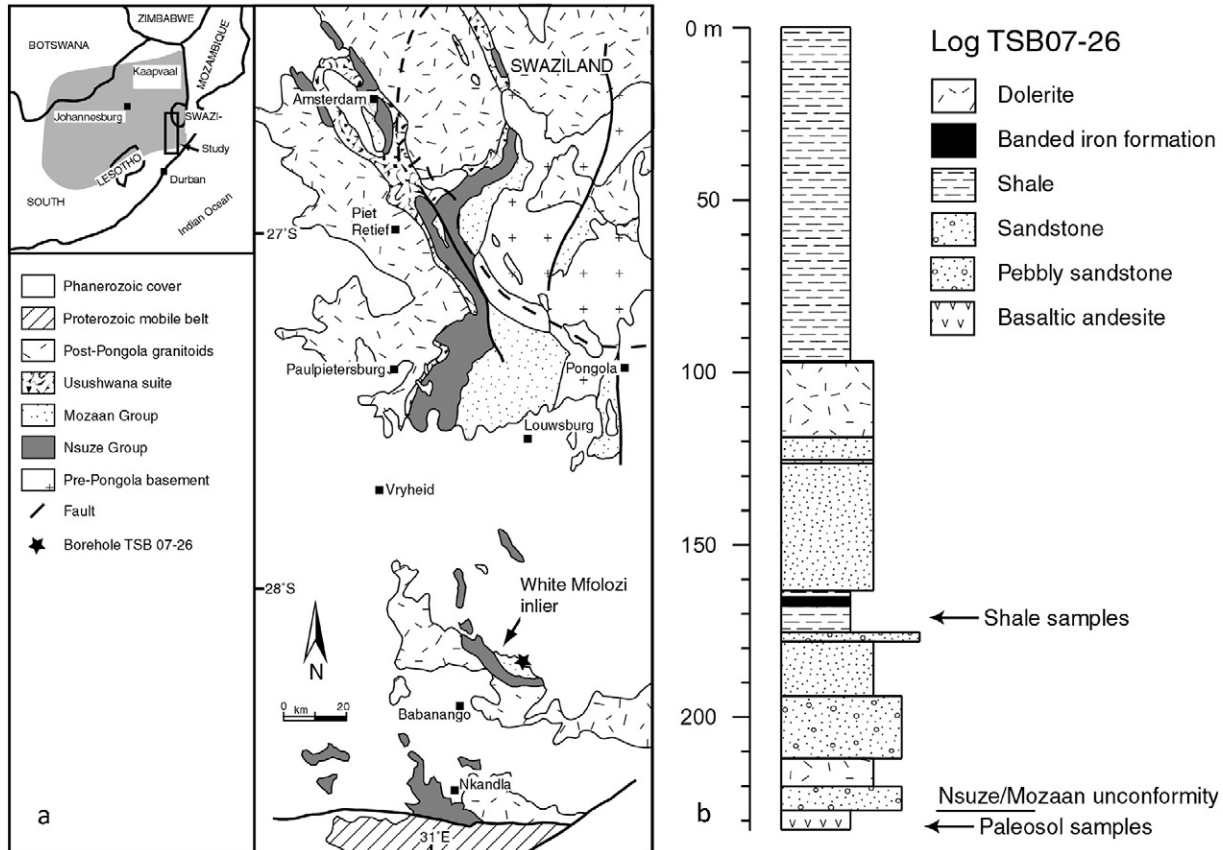


Fig. 1. (a) Geological map of the study area (modified after Bolhar et al., 2015) showing the extent of the Pongola Supergroup, location of the White Mfolozi Inlier and the borehole TSB 07-26. (b) Stratigraphic log of the drill core under study (TSB 07-26) indicating depths of shale and paleosol samples.

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