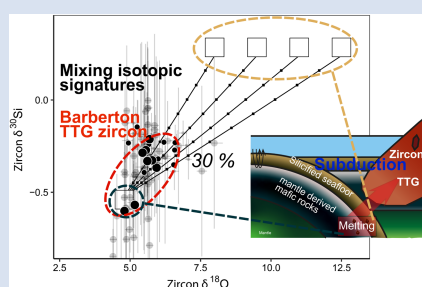


Silicified seafloor contribution to TTG formation: insights from zircon O and Si isotopes

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Abstract



Tracing the input of altered seafloor lithologies to primary melts of tonalite-trondhjemite-granodiorite suites (TTGs) is critical for understanding Archean geodynamic processes that transported these lithologies towards melting regions. Zircon oxygen and silicon isotopic compositions in TTGs are particularly useful for this end, but their interpretation remains challenged by scarce quantitative constraints on isotopic compositions reflecting partial melting of specific Archean lithologies. Here, we combine oxygen and silicon isotope measurements in 3.45 and 3.22 Ga Barberton TTGs and their zircons with numerical models simulating partial melting of different source lithologies, which predict the isotope signatures of zircons and their host melts. Measured whole rock and zircon O and Si isotope compositions reflect the presence of up to 30 wt. % of hydrothermally silicified mafic rocks derived from the seafloor in

the Barberton TTG source region. Considering the regional geological context, we propose subduction-like processes to explain the burial of the silicified seafloor into the TTG source. These results imply that subduction-like processes operated at least locally on Earth during the Paleoproterozoic.

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Introduction

Geochemical signatures of buried seafloor lithologies in Archean TTGs (tonalite-trondhjemite-granodiorite) are critical for constraining seafloor alteration and burial processes on the early Earth (Trail *et al.*, 2018; André *et al.*, 2019; Deng *et al.*, 2019). TTGs are the oldest available granitic lithologies and formed by partial melting of hydrated mafic crust (Condie, 2013). Because TTG source regions likely comprised altered lithologies derived from the surface (André *et al.*, 2019; Deng *et al.*, 2019), TTGs offer a unique opportunity to investigate surface alteration processes on the early Earth. Most TTGs still contain pristine magmatic zircons, minimally affected by post-crystallisation alteration, that are texturally and compositionally distinct from zircons crystallised or modified during late stage processes (e.g., metamorphism) (Guitreau *et al.*, 2022). These magmatic zircons typically serve as proxy for primary TTG melts (Trail *et al.*, 2018; Moreira *et al.*, 2020; Guitreau *et al.*, 2022; Wang *et al.*, 2022).

TTG zircon oxygen and silicon isotopic compositions are particularly powerful for tracing the reworking of Archean altered seafloor lithologies (Trail *et al.*, 2018; Wang *et al.*, 2022; Guitreau *et al.*, 2022; Lei *et al.*, 2023). This is because, compared to unaltered (mantle derived) mafic rocks exhibiting a narrow range of oxygen

and silicon isotopic compositions ($\delta^{18}\text{O}_{\text{wt}} = +5.6 \pm 0.6 \text{ ‰}$; Eiler *et al.*, 2000, and $\delta^{30}\text{Si}_{\text{wt}} = -0.29 \pm 0.06 \text{ ‰}$; Savage *et al.*, 2010), altered seafloor rocks display variable oxygen and silicon isotopic compositions due to low temperature seawater-rock interactions (Abraham *et al.*, 2011; André *et al.*, 2022). Particularly, sediments and basalts that were hydrothermally silicified on the Archean seafloor (hereafter silicified seafloor lithologies) exhibit elevated $\delta^{18}\text{O}_{\text{wt}}$ (+8 to +16 ‰) and $\delta^{30}\text{Si}_{\text{wt}}$ (0 to +1.5 ‰) (Abraham *et al.*, 2011; Hofmann and Harris, 2008). If such silicified seafloor lithologies partially melted to generate felsic liquids, the latter must have inherited their unique O and Si isotopic compositions. Thus, felsic melt extraction from buried silicified seafloor lithologies to Archean TTGs can be traced by bulk rock and zircon O and Si isotope compositions in TTGs (Trail *et al.*, 2018; André *et al.*, 2019). Generally, if located within the $\delta^{18}\text{O}_{\text{zrc}}$ range of $+5.3 \pm 0.6 \text{ ‰}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ range of $-0.38 \pm 0.04 \text{ ‰}$ characterising “mantle zircons” (or zircon potentially crystallised in the mantle and hosted in kimberlites; Valley *et al.*, 1998; Trail *et al.*, 2018), $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ values of TTG hosted zircons are interpreted to evidence zircon crystallisation from felsic liquids generated by partial melting of unaltered (mantle derived) mafic rocks. Contrastingly, $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ outside this “mantle zircon” range are generally ascribed to partial melting of altered mafic

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lithologies or sediments, or an input of fluids from the surface (Valley *et al.*, 2005; Moreira *et al.*, 2020; Lei *et al.*, 2023). However, the mantle zircon range was originally constrained by analysing zircons from kimberlites and peridotites (Valley *et al.*, 1998; Trail *et al.*, 2018), neglecting mineral–melt O and Si isotope fractionations during partial melting and crystallisation processes (Lackey *et al.*, 2008; Guitreau *et al.*, 2022; Murphy *et al.*, 2024). Considering these fractionations can improve the determination of TTG source lithologies from bulk rock and zircon O and Si isotope analyses and the detection of altered seafloor lithology signatures in TTGs and zircons. This is important because such signatures are critical for understanding surface alteration conditions (André *et al.*, 2019) and geodynamic processes that potentially transported altered rocks towards deeper melting regions (Deng *et al.*, 2019). These processes remain debated between 1) burial below thick volcanic successions (Hernández-Urbe, 2024), 2) burial due to density driven vertical movements (Smithies *et al.*, 2021), and 3) burial due to horizontal motions linked to modern-like subduction (Deng *et al.*, 2019). Integrating isotopic data with stratigraphic and metamorphic constraints on burial depth of reworked seafloor rocks can improve the understanding of Archean geodynamic processes.

This study aims to 1) trace the signature of silicified seafloor rocks in Archean TTG source regions using zircon O and Si isotope compositions, and 2) integrate isotopic data with knowledge of regional geology to discuss geodynamic processes that transported seafloor lithologies towards melting regions. For these purposes, we present new O and Si isotope data for magmatic zircons and new O isotope data for bulk rock samples from 3.45 and 3.22 Ga TTGs occurring in the Barberton Granitoid–Greenstone Terrain (BGGT, South Africa) (Table S-1 and Fig. S-1). These new data are combined with previously published O and Si isotopic compositions of bulk rock samples and zircons from Barberton TTG (Faure and Harris, 1991; Valley *et al.*, 2005; André *et al.*, 2019; Deng *et al.*, 2019; Guitreau *et al.*, 2022; Wang *et al.*, 2022; Lei *et al.*, 2023). We also present a petrological model simulating the O and Si isotope compositions of TTG melts and zircons that reflect the reworking of unaltered mafic lithologies and silicified seafloor mafic lithologies in the TTG source region.

Results

The $\delta^{18}\text{O}_{\text{zrc}}$, $\delta^{30}\text{Si}_{\text{zrc}}$, $\delta^{18}\text{O}_{\text{wt}}$ values, major element compositions of Barberton TTG samples and images of zircons analysed in this study (Tables S-1 to S-4 and Figs. S-1 to S-3) are provided in Supplementary Information. The TTG SiO_2 content ranges between 58.2 and 73.5 wt. % and covers a large compositional spectrum from tonalite to trondhjemitic (Fig. S-4), with a $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios of 0.22 to 0.38. Bulk $\delta^{18}\text{O}_{\text{wt}}$ values for the studied TTG samples range between +6.9 and +8.1 ‰, consistent with previously published $\delta^{18}\text{O}_{\text{wt}}$ values for Barberton TTGs (Fig. 1a) (Faure and Harris, 1991). For zircon, median $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ values *per* analysed TTG sample range respectively from $+4.8 \pm 0.59$ to $+5.91 \pm 0.96$ ‰ and from -0.21 ± 0.34 to -0.60 ± 0.57 ‰ (Fig. 1a,b). Significant $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ variation appear between crystals of the same sample with 2 s.d. values up to ± 0.96 ‰ for $\delta^{18}\text{O}_{\text{zrc}}$ and ± 0.57 ‰ for $\delta^{30}\text{Si}_{\text{zrc}}$ (Fig. 1, Tables S-1 to S-3). Important variations occur also between samples of the same pluton (*e.g.*, Nelshoogte and Stolzburg samples in Table S-1). Although samples SK-NL02 and SK-SLZ01 with the highest SiO_2 contents show the lowest zircon $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ median values, no continuous correlation is observed between bulk rock major element features like SiO_2 content (Fig. 1a) and inter-sample or intra-pluton variations of $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$.

In the thermodynamic model (modelling methods and additional results given in SI), partial melting of an average unaltered Onverwacht basalt at 700–800 °C and 0.8–1.3 GPa along different geothermal gradients (600–900 °C/GPa) provides a liquid that is similar in major element composition to primary melts of TTGs obtained experimentally (Laurie and Stevens, 2012) and calculated empirically from compiled Barberton TTG compositions (Laurent *et al.*, 2020) (*e.g.*, $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratio of ~ 0.25) (Fig. S-5). At the same P–T conditions, partial melting of the average silicified basalt produces a more potassic liquid ($\text{K}_2\text{O}/\text{Na}_2\text{O} = 3.1$ to 3.3) (Fig. S-5). Considering the proportion of residual mineral phases (Fig. S-6) and mineral/liquid fractionation coefficients (Table S-4), we modelled $\delta^{18}\text{O}$ values of +6.4 ‰ and $\delta^{30}\text{Si}$ values of -0.19 ‰ for TTG-like liquids generated by partial melting of unaltered Onverwacht basalt (or unaltered mafic source) (Fig. 2). For the liquid formed by partial melting of silicified mafic source, we obtained $\delta^{18}\text{O}_{\text{wt}}$ values of +9.5 to +14.0 ‰ (considering average $\delta^{18}\text{O}_{\text{wt}}$ variation between +10.0 and +14.5 ‰ in Onverwacht silicified rocks; Kitoga *et al.*, 2024) and $\delta^{30}\text{Si}_{\text{wt}}$ values of +0.58 to +0.56 ‰ (Fig. 2). For magmatic zircons crystallised in the melt derived from partial melting of an unaltered mafic source, we obtained $\delta^{18}\text{O}_{\text{zrc}}$ values of +4.8 to +5.0 ‰ and $\delta^{30}\text{Si}_{\text{zrc}}$ values of -0.50 to -0.60 ‰, considering zircon/liquid empirical fractionation coefficients (Lackey *et al.*, 2008; Guitreau *et al.*, 2022). Much higher $\delta^{18}\text{O}_{\text{zrc}}$ values of +8.0 to +12.5 ‰ and $\delta^{30}\text{Si}_{\text{zrc}}$ of +0.19 to +0.26 ‰ are obtained for zircons crystallised in equilibrium with the melt derived from silicified source (Fig. 2).

Discussion

Silicified seafloor lithologies in Barberton TTG source. Oxygen and silicon isotopic compositions of TTGs and their zircons are important for identifying altered seafloor lithology inputs into the TTG source regions (Trail *et al.*, 2018; André *et al.*, 2019; Deng *et al.*, 2019). Bulk rock and zircon $\delta^{18}\text{O}$ and $\delta^{30}\text{Si}$ values measured in Barberton TTGs (Fig. 1) (except in zircons from samples SK-SLZ01 and SK-NL02) are higher than isotopic values obtained in our model for felsic liquids issued by partial melting of an unaltered mantle derived mafic source and for zircon crystallised from these felsic melts (Fig. 2c). Variations of $\delta^{18}\text{O}$ and $\delta^{30}\text{Si}$ in bulk rock samples and zircons do not show a continuous correlation with bulk rock SiO_2 concentrations (Fig. 1). Such a continuous correlation typically reflects isotopic variation due to magmatic differentiation of a primary melt and/or crust assimilation (Lackey *et al.*, 2008; Savage *et al.*, 2011; Guitreau *et al.*, 2022). The lack of continuous correlation between isotopic compositions and SiO_2 in Barberton TTGs (Fig. 1) precludes an important control of magmatic differentiation on measured isotopic compositions.

In $\delta^{18}\text{O}$ versus $\delta^{30}\text{Si}$ diagram (Fig. 3), Barberton TTGs and their zircons plot on mixing curves between end member isotopic signatures reflecting partial melting of an unaltered mafic source and a silicified seafloor source. Mixing unaltered mafic lithologies with up to ~ 30 % of silicified seafloor rocks in the melting region accounts for the measured isotopic compositions, consistent with estimates of André *et al.* (2019). We note, however, that the lowest $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ values observed in samples SK-SLZ01 and SK-NL02 mimic the modelled zircon end member composition reflecting partial melting of an unaltered mafic lithology (Fig. 3). These particular zircons (and others displaying similar $\delta^{18}\text{O}_{\text{zrc}}$ values in the literature, *e.g.*, Moreira *et al.*, 2020; Smithies *et al.*, 2021) likely reflect a so far undocumented end member for Archean magmatic zircons crystallised in felsic liquids generated mainly by partial melting of an unaltered mafic lithology. This interpretation is also consistent with sample

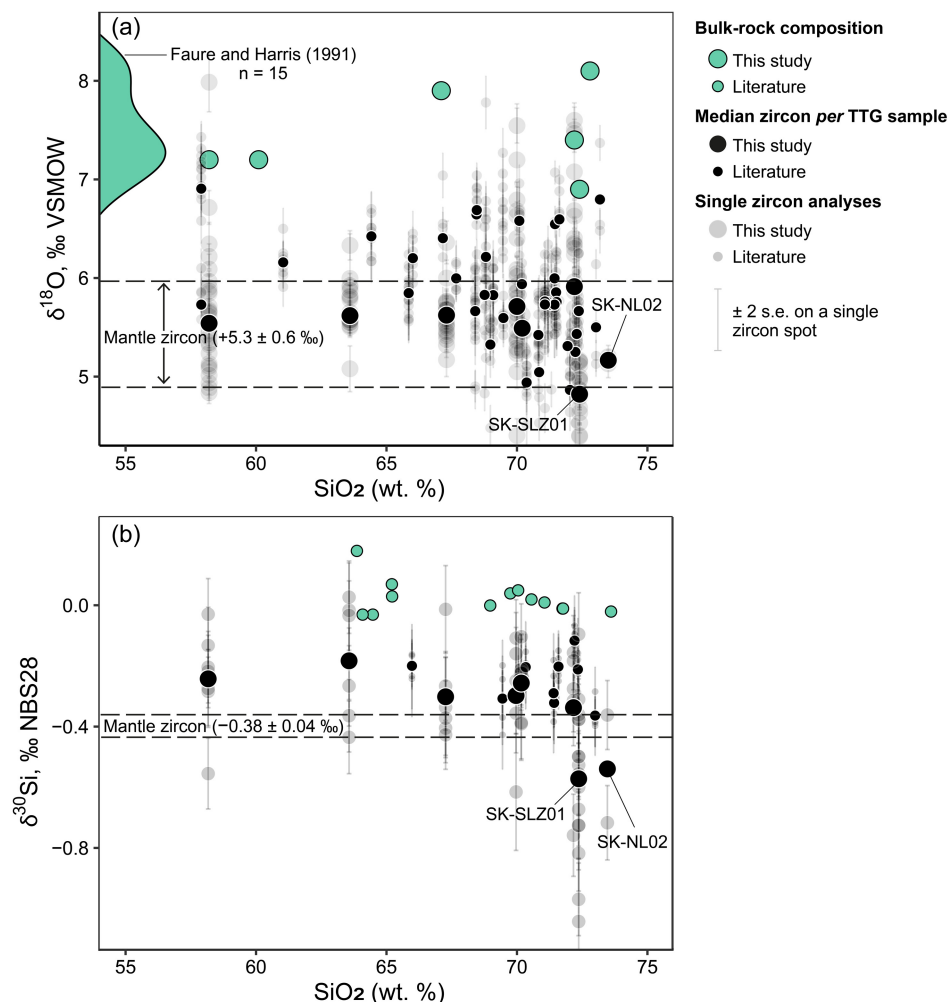


Figure 1 Oxygen and silicon isotopic composition of bulk rock and zircon samples from the BGGT plotted against bulk rock SiO_2 concentration. Error bars are 2 s.e. uncertainty for every spot. Density curve in (a) presents $\delta^{18}\text{O}_{\text{wr}}$ values in 15 samples from the Kaap Valley pluton (BGGT) from Faure and Harris (1991). Other Barberton TTG data sources: $\delta^{18}\text{O}_{\text{zrc}}$ in (a) (Valley et al., 2005; Wang et al., 2022; Lei et al., 2023), and whole rock $\delta^{30}\text{Si}_{\text{wr}}$ in (b) (André et al., 2019; Deng et al., 2019), $\delta^{30}\text{Si}_{\text{zrc}}$ (Guitreau et al., 2022; Lei et al., 2023). The ‘mantle zircon’ range represents $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ values of zircons from peridotites and kimberlites (Trail et al., 2018).

SK-SLZ01 showing the lowest whole rock $\delta^{18}\text{O}_{\text{wr}}$ of our dataset, although bulk rock compositions generally do not preserve this end member signature in Barberton TTGs.

Recently, a controversy on Barberton TTG source lithologies arose from interpretations of $\delta^{18}\text{O}$ and $\delta^{30}\text{Si}$ values of their bulk rock samples and zircons. Wang et al. (2022) interpreted 3.45 Ga Barberton TTG zircons with $\delta^{18}\text{O}_{\text{zrc}}$ signatures within the range of mantle zircons as demonstrating the absence of silicified seafloor rocks in the TTG source region. Conversely, $\delta^{30}\text{Si}_{\text{zrc}}$ (Guitreau et al., 2022; Lei et al., 2023) and $\delta^{30}\text{Si}_{\text{wr}}$ values (André et al., 2019; Deng et al., 2019) within the same Barberton TTGs were ascribed to the presence of silicified seafloor lithologies in the source regions. These contradictory interpretations result from considering that zircons with $\delta^{18}\text{O}_{\text{zrc}}$ of $+5.3 \pm 0.6 \text{ ‰}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ of $-0.38 \pm 0.04 \text{ ‰}$ (mantle zircon ranges; Fig. 1b), necessarily crystallised from melts formed by partial melting of unaltered mafic lithologies. Our model reveals that zircons reflecting an unaltered mantle derived source for TTGs are restricted to a $\delta^{18}\text{O}_{\text{zrc}}$ of $+4.8$ to $+5.0 \text{ ‰}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ of -0.5 to -0.6 ‰ (Fig. 2). When modelled end member compositions are mixed, $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ appear to be well coupled in Barberton TTGs and reflect a contribution of silicified seafloor lithologies to the TTG melt (Fig. 3). Our dataset shows no $\delta^{18}\text{O}_{\text{zrc}}$ variation over time, although Wang et al. (2022) observed

an increase of $\delta^{18}\text{O}_{\text{zrc}}$ at ~ 3.23 Ga in Barberton TTGs. The highest $\delta^{18}\text{O}_{\text{zrc}}$ (exceeding 5.9 ‰) were measured only in post-3.23 Ga Honingklip, Eerstehoeck and Uitgevonden bodies (Fig. S-1) by Wang et al. (2022) (not analysed here), and may reflect incorporation of silicified seafloor lithologies with a higher $\delta^{18}\text{O}$ value, by the source region of these specific plutons (Kitoga et al., 2024). Unlike that proposed by Wang et al. (2022), these high $\delta^{18}\text{O}_{\text{zrc}}$ values do not date the onset of seafloor recycling at ~ 3.23 Ga because seafloor derived rocks were also present in the source region of ~ 3.45 Ga old Stolzberg and Theespruit TTGs as well as 3.28–3.23 Ga old Kaap Valley and Nelshoogte TTGs investigated here (Fig. 3).

More importantly, our results demonstrate that Paleoproterozoic TTG zircons with $\delta^{18}\text{O}_{\text{zrc}}$ or $\delta^{30}\text{Si}_{\text{zrc}}$ values within the range of mantle zircon do not necessarily reflect an unaltered mafic source for TTG melts. Interestingly, the $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ values previously published for ≥ 3.0 Ga TTG hosted and detrital zircons in the Pilbara Craton, Saglek, North China Craton and Coorg block (Fig. 3; references listed in Table S-5) are also generally higher than signatures reflecting the partial melting of an unaltered mafic source. Consequently, partial melting of silicified seafloor rocks potentially contributed to Paleoproterozoic TTG melts worldwide.

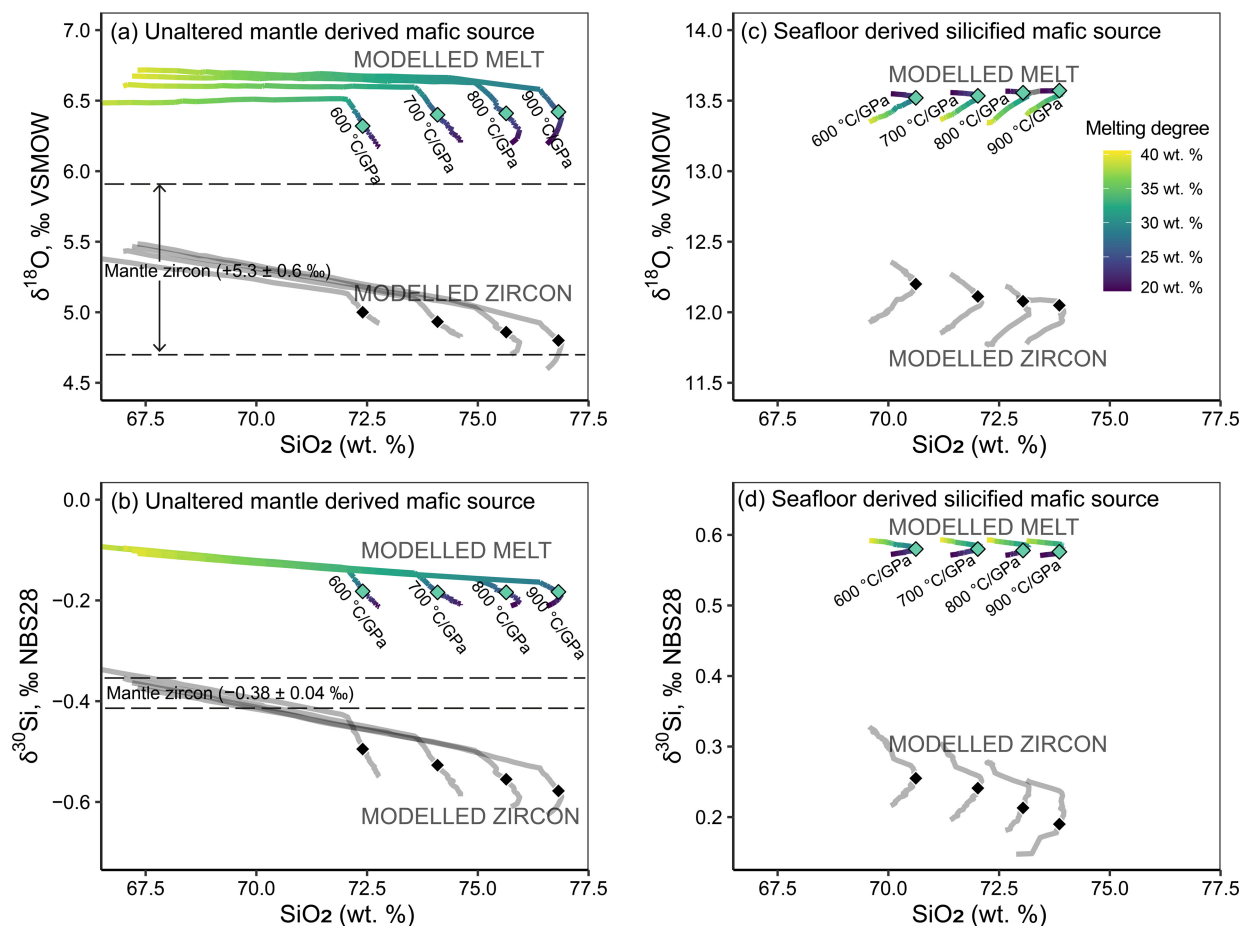


Figure 2 Modelled melt and zircon isotopic compositions reflecting partial melting of an unaltered mafic source and a silicified source along different P-T paths. We considered melting degrees between 20 and 40 wt. % (able to produce TTG-like melts) (Laurie and Stevens, 2012). Plotted SiO₂ concentrations are calculated on an anhydrous basis. Diamonds represent conditions where partial melting of a basaltic source generates a melt with TTG-like major element composition. In (c) we only considered an average δ¹⁸O_{wr} of +13.5 ‰ for a silicified mafic source (overlooking the full δ¹⁸O_{wr} variation shown in Fig. 3 for simplicity).

Evidence of Paleoproterozoic subduction-like processes in the Barberton area. The presence of silicified rocks in TTG source regions complements evidence from Nd-Hf isotopes that the mafic source of Barberton TTGs was extracted from the mantle less than 0.3 Ga before being reworked (Moyen *et al.*, 2019). These observations collectively support 3.5–3.2 Ga old rocks of the Onverwacht Group as proxies of lithologies melted to form 3.45 and 3.22 Ga Barberton TTGs (André *et al.*, 2022; Kitoga *et al.*, 2024). Thus, to constrain geodynamic processes that may have buried silicified seafloor lithologies into melting regions, we interpret our isotopic constraints in light of geological information on the stratigraphic burial and regional metamorphism of Onverwacht silicified rocks before episodes of TTG magmatism (Hofmann and Harris, 2008; Byerly *et al.*, 2019).

Within the BGGT and surrounding Paleoproterozoic terranes (Byerly *et al.*, 2019), there is no evidence (*e.g.*, particularly thick stratigraphic layers) of an important volcanic and/or sedimentary event capable of burying the silicified rocks to the depth (>20 km) required for their partial melting ~3.45 and ~3.2 Ga ago, as would be needed in a stagnant crust scenario (Moyen *et al.*, 2019). This precludes partial melting of altered basalts at the base of an over-thickened volcano-sedimentary succession (Hernández-Uribe, 2024) as a viable explanation for the formation of BGGT TTGs. At the time of TTG magmatism in the BGGT, the silicified seafloor lithologies of the Onverwacht Group were not stratigraphically buried deeper than 10 km below the surface by younger volcanic

or sedimentary piles. Thus, vertical movements of delamination or dripping (Van Kranendonk *et al.*, 2014) are also unlikely to explain their transport towards TTG source regions as these vertical movements should preferentially mobilise lower crustal material. In addition, if overturn movements proposed by Schmitz and Heubeck (2021) controlled the recycling of silicified rocks into the TTG source region, at 3.48 to 3.20 Ga, a 10 km thick volcano-sedimentary succession would not be as continuously preserved with greenschist metamorphic facies, as it actually is in Barberton (Byerly *et al.*, 2019).

Discarding alternatives favours subduction-like processes (Stevens *et al.*, 2002; Moyen *et al.*, 2006) to explain the reworking of seafloor silicified rocks in the Barberton TTG source regions. A horizontally mobile portion of the Paleoproterozoic crust was likely subducted below a relatively stagnant (and therefore more easily preserved) portion of the same crust due to vigorous mantle convection. This model accounts for both geochemical evidence of reworked silicified seafloor, preservation of a ~10 km thick, continuous volcano-sedimentary succession that has never exceeded greenschist facies in northern BGGT, and rapid subduction of sediments proposed around 3.2 Ga ago by petrochronological analyses in southern BGGT (Stevens *et al.*, 2002; Moyen *et al.*, 2006). It is, however, important to note that this proposed localised subduction does not necessarily imply global subduction worldwide in the Paleoproterozoic. In fact, in other Paleoproterozoic terrains such as the Pilbara craton, density driven

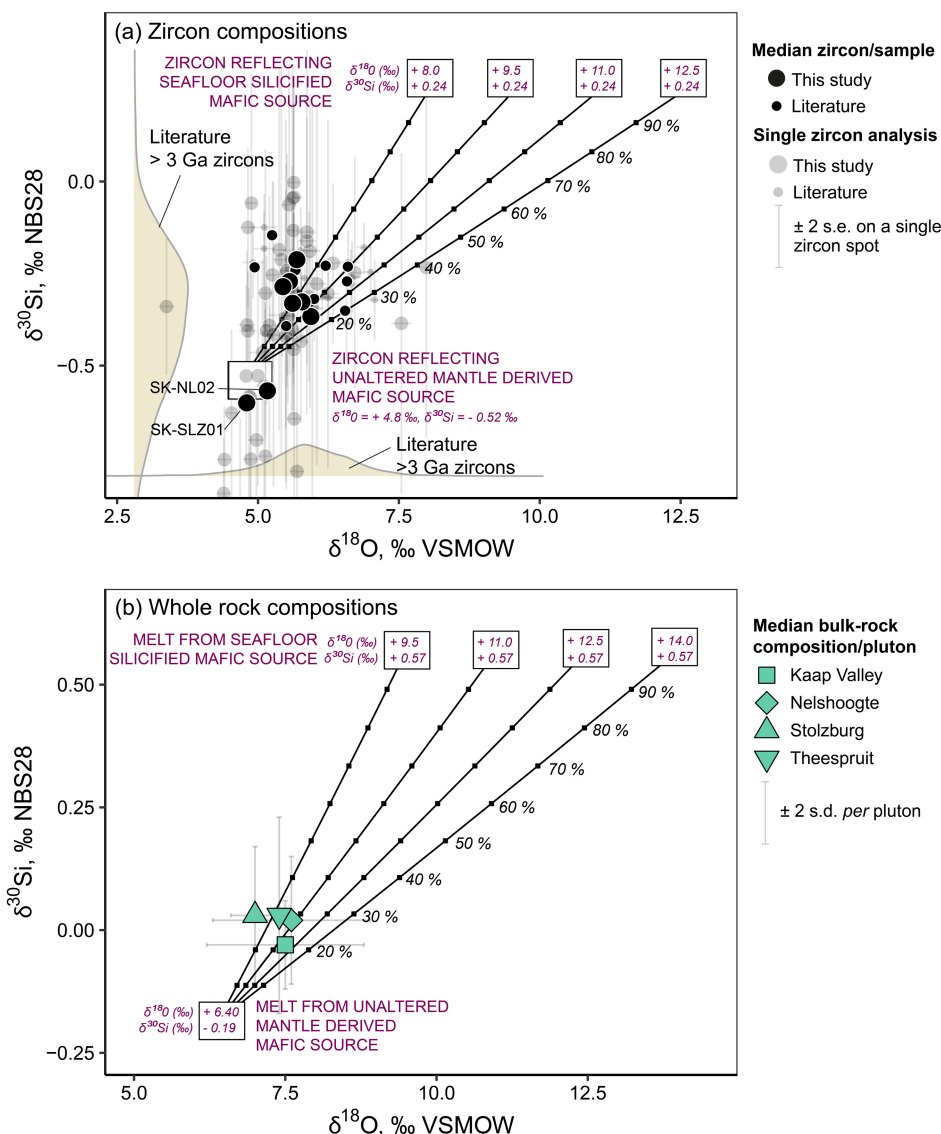


Figure 3 Mixing model explaining the origin of oxygen and silicon isotopic compositions of (a) BGGT TTG zircon and (b) whole rock samples. Density curves in (a) are distributions of $\delta^{18}\text{O}_{\text{zrc}}$ and $\delta^{30}\text{Si}_{\text{zrc}}$ in other Paleoproterozoic or older zircons (references in Table S-5). The two end member source signatures considered in the mixing calculations result from our numerical model. Zircon compositions on mixing curves represent the calculated isotopic composition of zircon crystallised from modelled liquids (end member or mixed). Average $\delta^{18}\text{O}_{\text{wr}}$ temporal variation in Onverwacht silicified lavas (Kitoga et al., 2024) is considered.

vertical transportation was proposed as the most likely geodynamic scenario by both geological and isotopic data (Smithies et al., 2019). Co-occurrence of different reworking processes on the Paleoproterozoic Earth remains a possibility.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2608>.



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