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Eclogite xenoliths record a constant ocean oxygen isotope composition for 3 billion years

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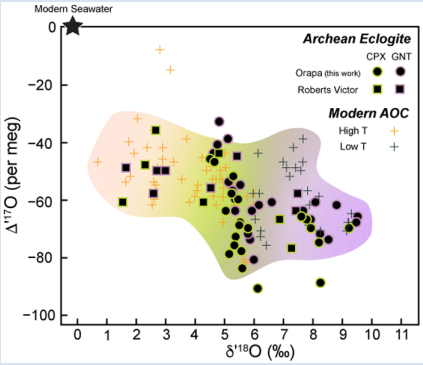
Abstract



In this work, we utilised the triple oxygen isotope (TOI) composition of Archean-age xenolithic eclogites, representative of Archean subducted altered oceanic crust (AOC) to constrain the triple oxygen isotope composition of the Earth's ancient ocean. We present new triple oxygen results of eclogite xenolith garnet and clinopyroxene mineral separates from the Orapa kimberlite mine (Botswana). In conjunction with samples from Roberts Victor (South Africa), the combined TOI range of both Archean xenolith suites overlap with modern AOC precisely, and the alteration signature suggests Archean seawater had the same TOI composition as the modern (ice-free) ocean. Using fluid–rock exchange modelling, we demonstrate that Archean seawater was similarly buffered by both high and low temperature interaction with ocean crust, like the present day system. The low $\delta^{18}O$ values of Archean cherts typically used to reconstruct Archean seawater are inconsistent with *ad hoc* suggestions of isotopically light ocean water and provide an important boundary condition for future studies of Archean sediments and models.

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Introduction

The oxygen isotope composition of the Earth's Archean ocean has principally been interpreted through the oxygen isotope composition of sedimentary rocks. As a surficial sedimentary archive, Archean cherts offer a window into the past, but only if diagenesis during or after deposition has not overprinted the original seawater oxygen isotope signature. Significant oxygen isotope modification is seen in recent (Miocene to present) deep ocean silica (Ibarra *et al.*, 2022; Tatzel *et al.*, 2022), but the trends do not match the Archean data, suggesting an alternative explanation. It remains unresolved as to whether the highest $\delta^{18}O_{chert}$ values (or the least altered) ($\delta = (R_{sam}/R_{std} - 1)1000$ in per mille notation, $R = {}^{18}O/{}^{16}O$ ratio of sample or standard) reflect either an isotopically lower ocean than the present (up to 15 ‰) (Perry, 1967) or a record of extremely hot seawater (>70 °C) (Lowe *et al.*, 2020). Deciphering the conditions of the Archean ocean and its hydrologic implications for the Earth's earliest rock record requires an independent proxy for either the $\delta^{18}O$ or the temperature of ancient seawater that is not prone to the effects of diagenesis.

Altered Oceanic Crust as a Proxy for Ancient Seawater $\delta^{18}O$

Unaltered ocean crust is emplaced at spreading centres as a stratified sequence of pillow basalts near the surface, underlain by sheeted dikes and gabbro with increasing depth. Hydrothermal alteration occurs both from a deep, high temperature hydrothermal interaction with seawater near the spreading centre and cold, off axis alteration on million year timescales. The final equilibrated whole rock $\delta^{18}O$ value of the altered oceanic crust (AOC) is a function of the initial mid-ocean ridge basalt (MORB) and seawater $\delta^{18}O$ values, as well as the temperature and degree of alteration (fluid/rock ratio). The oxygen isotope composition of fresh MORB has been constant through time (Mattey *et al.*, 1994). The low (~50 °C) and high (~350 °C) temperatures of hydrothermal alteration have also remained constant through time (Staudigel, 2003). Therefore, any differences in the triple oxygen isotope fields between ancient and modern AOC is a direct measure of changes in the $\delta^{18}O$ of seawater over geologic time.

Due to the nature of subduction, the oldest existing oceanic lithosphere is around 200 Ma with older AOC preserved almost exclusively as obducted oceanic crust, known as ophiolite sequences. Exposures of Palaeoproterozoic (2.5 Ga) ophiolites generally have $\delta^{18}O$ values that overlap with modern oceanic crust from the Deep Sea Drilling Program (DSDP) (Holmden and Muehlenbachs, 1993). Older, Archean-age hydrothermally altered mafic rocks are limited to greenstone belts which have a complicated emplacement and fluid alteration history (Gregory, 2003). The degree of diagenetic alteration, and therefore the possibility that obducted AOC sequences provide non-primary information, is uncertain.

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Xenolithic eclogites provide an attractive alternative to determining the Archean seawater $\delta^{18}O$ value because they are thought to be subducted AOC (MacGregor and Manton, 1986) that became metamorphosed to garnet- and omphacite-bearing rocks without significant isotopic modification. The eclogite xenolith record is especially relevant for interpreting the oldest oceanic lithosphere because most are of Archean age (Aulbach and Smart, 2023), and isotopic preservation is generally supported because low fluid–rock ratios are likely to prevail within the cold and thick continental lithosphere. Modification has been suggested only from intense melt-rock interaction that is identified in a subset of samples from any given eclogite xenolith suite and that is easily identified using geochemical proxies (Aulbach *et al.*, 2020).

The Overlapping Triple Oxygen Isotope Signature of Modern AOC and Ancient Eclogite Xenoliths

We report new triple oxygen isotope (TOI) values for Archean-age eclogite xenoliths from the Cretaceous Orapa kimberlite (2.99 ± 0.26 Ga) (Shirey *et al.*, 2008) located on the margin of the Zimbabwe Craton (Botswana). We measured pristine separates of garnet ($n = 23$) and clinopyroxene ($n = 22$) mineral pairs from the Orapa kimberlite mine (Table S-2). These data, combined with eclogite xenolith triple oxygen analyses from the proximal Roberts Victor kimberlite mine (2.7 ± 0.1 Ga) (Jagoutz *et al.*, 1984), located on the Kaapvaal Craton (South Africa), as well as modern AOC (McGunnigle *et al.*, 2022), are shown in Figure 1. The eclogite $\delta^{18}O$ and $\Delta'^{17}O$ values range from 1.8 to 9.8 ‰ and −35 to −85 per meg, respectively. The combined triple oxygen isotope range of both ancient suites is identical to near modern AOC. If the eclogite xenoliths analysed here have retained their original near surface oxygen isotope signature, then they serve as a proxy for the oxygen isotope composition of ancient seawater. We show how the overlapping TOI signature of modern AOC and ancient eclogite xenoliths reflects no difference between the reconstructed Archean and modern ocean oxygen isotope compositions, and has three broadly important geologic consequences: 1) Eclogite xenoliths preserve a signature of Archean altered oceanic crust that underwent subduction and recycling back to the surface *via* kimberlite volcanism without modification; 2) The composition of ancient seawater was buffered by similar ^{18}O fluxes relative to the modern hydrologic system on Earth (Muehlenbachs, 1998); and 3) The oxygen isotope composition of Archean cherts records either a hot ancient ocean or diagenesis.

Possible Mantle Modification of the Eclogite Xenolith Record

The origins of eclogite xenoliths and their utility as a recorder of ancient AOC isotope values is an ongoing debate due to the susceptibility of fluid or melt alteration within the continental lithosphere. The overlapping TOI range of eclogite xenoliths with modern AOC is evidence of a surficial origin, primarily because large isotopic variations are representative of low temperature mineral-water fractionations only. The main argument against a surficial signature is mantle metasomatism (O'Reilly and Griffin, 2013) or alteration by an externally derived fluid or melt prior to kimberlite entrainment. Two main ideas have evolved from mantle metasomatic studies. The first prescribes eclogite $\delta^{18}O$ values greater than 5.5 ‰ as interaction with a mantle metasomatic agent sourced from subducted sediments ($\delta^{18}O_{fluid}$: +15 to +30 ‰) (Gréau *et al.*, 2011). Alternatively,

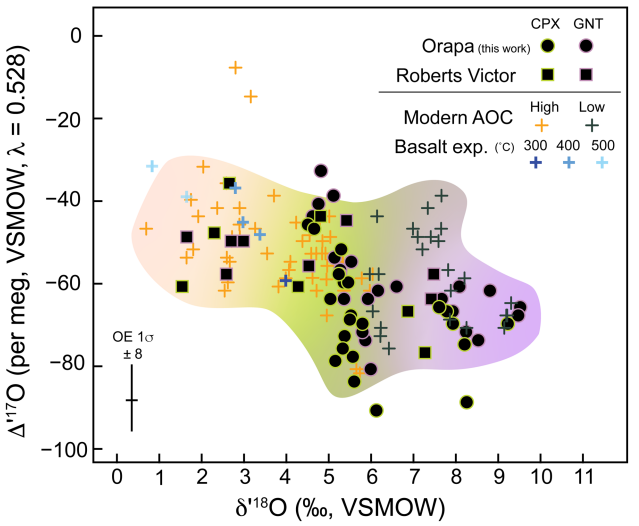

Figure 1 $\Delta'^{17}O$ *versus* $\delta'^{18}O$ modern natural and experimental altered ocean crust and ancient eclogite xenoliths. Eclogite garnet (black with purple outline) and clinopyroxene (black with green outline) mineral separate analyses are represented by the circles (Orapa: this work) and squares (Roberts Victor) (McGunnigle *et al.*, 2022). The yellow and grey crosses (+) are the high and low temperature modern altered ocean crust samples, respectively. The modern field is outlined by the coloured region. Two sample replicates for one bulk rock analysis of AOC with high $\Delta'^{17}O$ values were found to be very heterogeneous and are not included in the construction of the coloured region (note that two samples from IODP Hole 504B had similar values; Sengupta and Pack, 2018). The blue crosses are data from high temperature basalt-water equilibration experiments (McGunnigle *et al.*, 2022). The $\Delta'^{17}O$ 1σ uncertainty range is shown for the Orapa eclogite only and was calculated based on San Carlos olivine analyses ($n = 25$) for this work (Table S-1). The garnet and clinopyroxene triple oxygen isotope ranges are identical and overlap almost exactly with the modern AOC analyses.

more recent studies of intense metasomatic modification by proto-kimberlite magmatism support a mantle-like oxygen isotope 'overprinting' ($\delta^{18}O_{fluid} \sim 5.5$ ‰) on subordinate portions of the eclogite inventory for a given locality (Aulbach *et al.*, 2020).

The eclogite xenolith record in this work exhibits a $\delta^{18}O$ range both less than and greater than the canonical mantle value (Cano *et al.*, 2020). Surficial sedimentary sourced metasomatic fluids can only increase the $\delta^{18}O$ value of eclogitic materials. Therefore, the isotopically low TOI values could then only be explained by a mantle metasomatism with isotopically low primordial mantle materials ($\delta^{18}O < 5.5$ ‰). This is not supported by Archean-age peridotitic diamond inclusions, which are shielded by their chemically inert hosts, and whose similarity with modern MORB infers a constant mantle O-isotope composition since their inception (>3 Ga) (Mattey *et al.*, 1994).

High temperature isotope fractionations alone are too small to modify the $\delta^{18}O$ and $\Delta'^{17}O$ values of mantle minerals which have a TOI composition of roughly $\delta'^{18}O = 5.5$ ‰ and $\Delta'^{17}O = -55 \pm 5$ per meg based on terrestrial basalt glasses and dunites (Cano *et al.*, 2020). We calculated the TOI fractionation between garnet (grossular), clinopyroxene (diopside) and olivine (forsterite) using theoretically determined fractionation factors (Schauble and Young, 2021). Even at the relatively 'cold' sublithospheric temperatures of 900 °C, the diopside–forsterite fractionation is only +1 ‰ for $\delta^{18}O$ and +1.5 per meg for $\Delta^{17}O$, meaning that diopside will be slightly enriched in $\delta^{18}O$ relative to forsterite, with a near constant $\Delta'^{17}O$ (within analytical

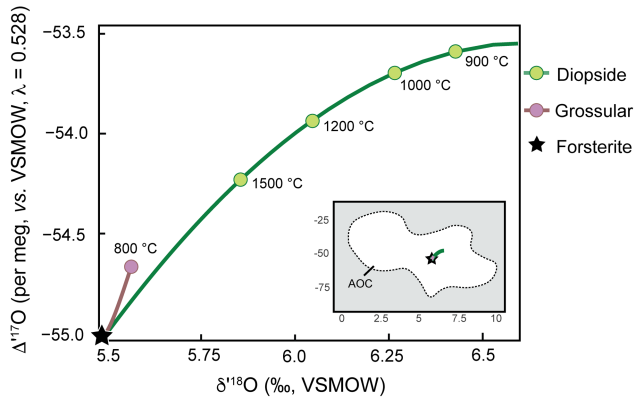

Figure 2 Δ'^{17}O *versus* δ'^{18}O triple oxygen isotope fractionation between olivine and eclogite mineral phases. Calculated theoretical equilibrium fractionation (Schauble and Young, 2021) between eclogite phases relative to mantle olivine with TOI values of δ'^{18}O = 5.5 ‰, Δ'^{17}O = −55 per meg (Cano *et al.*, 2020). The greatest Δ'^{18}O difference ($\delta'^{18}O_{diopside} - \delta'^{18}O_{forsterite}$) corresponding to equilibrium at 900 °C is only 1 ‰, and for grossular is <0.2 ‰. The inset shows these fractionations relative to the general range found in modern AOC and ancient eclogite xenoliths from this work. Equilibrium isotope fractionation cannot explain the variation of δ^{18}O and Δ'^{17}O values found in the eclogites.

uncertainty) (Fig. 2). The grossular–forsterite fractionation is essentially 0 ‰ (within analytical uncertainty). Based on these calculations, mantle metasomatism is inefficient for generating the widespread oxygen isotopic heterogeneity found in eclogite xenoliths unless a metasomatic fluid with an exotic, non-mantle fluid is assumed. Rather, metasomatism homogenises the isotopic variation closer to the bulk composition of the mantle (Aulbach *et al.*, 2020). Because the purported sources of exotic fluids capable of mantle metasomatic alteration are surficial sedimentary melts, this hypothesis is only capable of explaining a portion of the eclogite inventory, those with δ^{18}O values greater than 5.5 ‰. Despite mantle homogenisation, our combined data set remains representative of the entire modern range, suggesting isotopic preservation of altered oceanic crust within the subcontinental lithosphere throughout the multi-billion-year residence time and prevalent metasomatic alteration. While perhaps not intuitive, it is reasonable to conclude that subduction and metamorphism of oceanic crust is a good way of preserving the seawater alteration signature unobscured by diagenesis that affects the sedimentary record.

Comparing Ancient Eclogites to Modern AOC

The oxygen isotope composition of the ancient ocean is preserved in AOC and is represented by the eclogite xenolith record. Continuous seawater circulation imposes a distinct oxygen isotope signature upon the initially unaltered MORB, resulting in isotopic exchange that reflects either high temperature hydrothermal alteration or low temperature seafloor 'weathering' (Muehlenbachs, 1986). Two juxtaposing results arise from this currently active seafloor process: an imprint of the $\delta^{18}O_{ocean}$ upon the altered crustal assemblage and a buffered $\delta^{18}O_{ocean}$ value controlled by water–rock interaction between the ocean and various ^{18}O reservoirs present on Earth. Under this model, the composition of the ocean will remain at steady state so long as seafloor spreading persists, because the contribution of ^{18}O from hydrothermally altered basalt serves as the greatest flux, offset by low temperature sinks from the oceans and continents (Muehlenbachs, 1998).

The oxygen isotope composition of a complete section of ocean crust, recognised from both modern drill cores and ancient ophiolite sections emphasises the geologic significance of subsurface seawater circulation (Gregory and Taylor, 1981). To test the consistency of the buffered ocean paradigm in the Archean, the eclogite xenolith record is useful because the ancient triple oxygen isotope variation is fully analogous to the modern one, with virtually identical ranges in δ'^{18}O and Δ'^{17}O values. We therefore argue that the same processes buffering the current ocean were also active early in Earth's geologic history despite alternative hypotheses, such as minimised rates of hydrothermal alteration (Kasting *et al.*, 2006), closed system basalt-water exchange (Jaffrés *et al.*, 2007), and extreme rates of continental weathering, all of which have been proposed based on the low $\delta^{18}O_{ocean}$ estimates from sedimentary archives (Wallmann, 2001). These *ad hoc* explanations for low or high ancient δ^{18}O values of the ocean are not consistent with the near identical range of triple isotope values of ancient eclogites and modern AOC.

To show that the Archean hydrological system operated similarly to the modern one, we used a closed system, fluid–rock exchange model to calculate and compare the expected triple oxygen isotope compositions of low and high temperature AOC that would be preserved in the eclogite xenolith record if the ocean oxygen isotope value was lower than the present. The triple isotope solution is resolvable by using the modern

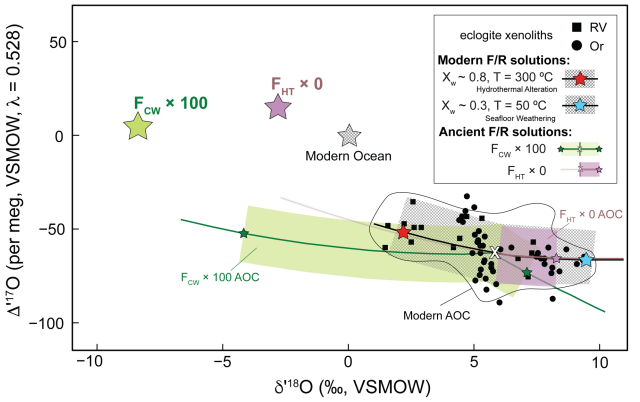

Figure 3 Closed system, fluid–rock model trajectories for modern MORB (**X**) altered by modern ocean water ($\delta'^{18}O = 0$ ‰, $\Delta'^{17}O = 0$ per meg, black crosshatched star) at low (50 °C) and high (300 °C) temperatures (see Supplementary Information, Section 5 for details). The modern AOC modelled variation is represented by the crosshatched region extending from **X** to a fluid–rock ratio (X_W) of 0.8 for high and 0.3 for low temperature alteration. The solid black line and cross hatched region shows the range for measured AOC (McGunnigle *et al.*, 2022). Each high and low temperature solution is used to solve the Δ'^{17}O basalt-water apparent fractionations for hydrothermal alteration and seafloor weathering under two different model ocean compositions (McGunnigle *et al.*, 2022): 1) Extreme continental weathering ($F_{CW} \times 100$, large green star); and 2) No high temperature hydrothermal alteration ($F_{HT} \times 0$, large purple star). The respective shaded region for each ocean composition represents the predicted ranges for AOC extrapolated to the same fluid–rock ratio as the modern one. The reduced range for HT × 0 is due to a lack of high temperature hydrothermal alteration that underlies this model. Ancient eclogite xenoliths from Orapa (Or) and Roberts Victor (RV) (black squares and circles) extend to a nearly identical range as modern AOC. These data cannot be explained from an $\delta'^{18}O$ ocean value of −8 ‰ or −4 ‰ assuming the latter shift is due to a lack of high temperature ocean crust alteration.

measured minimum and maximum $\delta'^{18}O$ (high and low temperature) altered rocks and experimentally determined 'apparent' basalt-water fractionations ($\Delta^{18}O_{basalt\text{-}water}$) (Cole *et al.*, 1987) to calculate the water fractions (X_w). We extrapolated the latter unknown to solve the $\Delta^{17}O_{basalt\text{-}water}$ apparent fractionations and applied our results using modelled triple oxygen isotope ocean compositions for the condition of 0 % hydrothermal alteration (HA × 0) and extreme continental weathering (CW × 100) (Fig. 3) (McGunnigle *et al.*, 2022). Each seawater solution has its own unique altered oceanic crust signature that is not consistent with the xenolith record. Our calculations show that the HA × 0 AOC solution, which only consists of low temperature 'weathered' crust, has a minimal TOI range, while the CW × 100 AOC solution extends far beyond the modern trajectory, with negative $\delta'^{18}O$ values. Neither low $\delta'^{18}O_{ocean}$ model prediction can explain the Archean eclogite signature in this work, leaving the explanation of a 0 ± 2 ‰ $\delta^{18}O$ Archean ocean as the only plausible explanation for the eclogite triple isotope data.

Conclusions

This work provides a significant amendment to the Archean ocean oxygen isotope record by comparing the indifferent triple oxygen isotope composition of near modern altered oceanic crust and ancient eclogite xenoliths. The resemblance suggests the ocean's $\delta'^{18}O$-$\Delta'^{17}O$ composition has remained constant through time, preserving a signature of Earth's hydrological evolution that persisted in a similar manner for the past three billion years. Establishing that the oxygen isotope composition of the Archean ocean was like the modern one reduces the interpretations for the low $\delta^{18}O$ values of Archean cherts to either 1) a hot Archean ocean, or 2) post-depositional diagenesis. Conversely, an isotopically light ocean with temperatures similar to the modern is not compatible with the eclogite record presented here.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2536.

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