

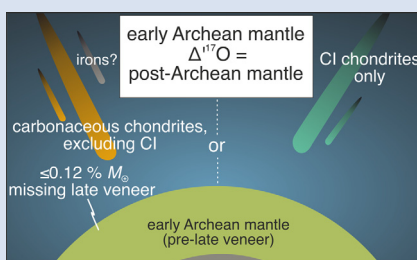
Tight bounds on missing late veneer in early Archean peridotite from triple oxygen isotopes

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Abstract



Oxygen isotopes provide a unique possibility to study Earth's late accretion phase from a lithophile element perspective, because most carbonaceous chondrites – meteorites that likely resemble the composition of the terrestrial late veneer – have markedly different $\Delta^{17}\text{O}$ values than the silicate Earth. Ultramafic rocks in the early Archean assemblage of southwest Greenland have not incorporated the full amount of late accreted materials, and therefore possibly record the $\Delta^{17}\text{O}$ of the mantle before late accretion. We measured $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ ratios of olivine from these ultramafic rocks and compared them with olivine from post-Archean mantle peridotite. A missing late veneer component was not resolved. The missing component from the early Archean mantle is therefore restricted to $\leq 0.12\%$ of Earth's mass

(M_{\oplus}) for most carbonaceous chondrite-like materials, unless the missing component resembles CI chondrites – the only carbonaceous chondrites with $\Delta^{17}\text{O}$ values similar to those of the silicate Earth. If the early Archean mantle had incorporated 60% late veneer, the overall late accreted mass would be restricted to $\leq 0.3\%$ M_{\oplus} for most types of carbonaceous chondrites, with a more massive late veneer only possible for CI-like chondrites.

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Introduction

Missing late veneer in early Archean mantle domains. Earth formed by the accretion of rocky materials in a protoplanetary disk. The final $\sim 0.5\%$ of these materials escaped metal-silicate equilibrium during core formation processes, and are commonly referred to as the late veneer (Walker, 2009). The composition of these late accreted materials is interesting to study, because they possibly contributed a large portion of volatile elements and water to the Earth's mass (Wang and Becker, 2013; Braukmüller et al., 2019).

Based on platinum and tungsten isotope work, it was proposed that some Archean mantle domains did not incorporate the full amount of late veneer (e.g., Willbold et al., 2011; Creech et al., 2017). Such mantle domains could then provide unique windows into the final stages of Earth's accretion and its earliest evolution. An unambiguous pre-late veneer signal is present in ultramafic enclaves that are entrained in the Eoarchean Itsaq Gneiss Complex and Mesoarchean Fiskefjord region of southwest Greenland. These rocks show a uniform excess in s-process Ru nuclides compared to r-process Ru nuclides relative to the bulk silicate Earth (Fischer-Gödde et al., 2020). The excess s-process Ru in the

ultramafic enclaves is best explained by a deficit in late accreted materials that carried a deficit in s-process Ru. Whereas both carbonaceous and non-carbonaceous chondrite groups carry deficits in s-process Ru, the s-process deficit is only sufficiently large in carbonaceous chondrite groups to potentially account for the observed s-process excess in the ultramafic enclaves, given the concentrations of platinum group elements in the Eoarchean mantle (Fischer-Gödde et al., 2020). This conclusion is important, because it would support a volatile-rich, carbonaceous chondrite-like late veneer, and therewith complement evidence from volatile chalcophile elements in the post-Archean mantle (Wang and Becker, 2013; Braukmüller et al., 2019; Varas-Reus et al., 2019).

Potential of studying triple oxygen isotope ratios. Oxygen stable isotope ratios ($^{17}\text{O}/^{16}\text{O}$, $^{18}\text{O}/^{16}\text{O}$) are a unique tool for studying late accretion processes from a lithophile element perspective (Rumble et al., 2013; Valley et al., 2014; Young et al., 2016; Reimink et al., 2018), because bulk asteroids in the Solar System show ~ 6000 ppm variations in $\Delta^{17}\text{O}$ (Clayton, 1993). This is large compared to the ~ 5 ppm analytical resolution of state of the art laser fluorination techniques. Henceforth, we use a definition for $\Delta^{17}\text{O}$ with:

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$$\Delta^{17}\text{O} = 1000 \left[1000 \ln \left(\frac{\delta^{17}\text{O}}{1000} + 1 \right) - 0.528 \times 1000 \ln \left(\frac{\delta^{18}\text{O}}{1000} + 1 \right) \right]$$

Eq. 1

Carbonaceous chondrites have lower $\Delta^{17}\text{O}$ values than the silicate Earth, with only CI chondrites having similar but slightly higher $\Delta^{17}\text{O}$ and much higher $\delta^{18}\text{O}$ values than the silicate Earth (Clayton, 1993). If the early Archean ultramafic enclaves from southwest Greenland would have been deprived of a late veneer component that resembles most carbonaceous chondrite types (CM, CV, CO, CK, CR, CH, CB chondrites), samples from the early Archean ultramafic enclaves would therefore be expected to have an elevated $\Delta^{17}\text{O}$ compared to the silicate Earth. Only if the ultramafic enclaves would be missing a late veneer component that resembles CI chondrites, would they have a similar or lower $\Delta^{17}\text{O}$ than the silicate Earth. In order to study mass and composition of the missing late veneer component from the early Archean ultramafic enclaves of southwest Greenland, we therefore compared their olivine $\Delta^{17}\text{O}$ values with olivine in post-Archean mantle peridotite.

Samples and Methods

The suite of post-Archean mantle peridotite sample comprises 14 lherzolitic xenoliths that were erupted in Phanerozoic magmas from diverse geological settings, and 1 dunite sample from the Beni Boussera massif (Supplementary Information S-1). The ultramafic enclaves with s-process Ru excess (Fischer-Gödde et al., 2020) that we studied are from two localities within the >3.7 Ga Isua supracrustal belt (ISB), one locality south of the Isua supracrustal belt (SOISB), the ~3.8 Ga Narssaq ultramafic body (NUB), and the ~3.8 Ga Ujaragssuit Nunât ultramafic body (Supplementary Information S-1). The early Archean ultramafic bodies were interpreted either as slivers of residual mantle peridotite, as metamorphosed cumulates from (ultra)mafic magmas, or, for one location, as an ultramafic layered intrusion. We also studied peridotite samples from the Fiskefjord region that are geologically related to the Mesoproterozoic Seqi peridotite body (Szilas et al., 2015), for which an excess in s-process Ru compared to the silicate Earth was also reported (Fischer-Gödde et al., 2020). Olivine samples were analysed for their triple oxygen isotope compositions ($\delta^{17}\text{O}$, $\delta^{18}\text{O}$) using laser assisted fluorination protocols (Supplementary Information S-2). All samples were analysed relative to San Carlos olivine, which is considered here to have a $\Delta^{17}\text{O} = -51.8$ ppm relative to VSMOW.

Results

Olivine $\delta^{18}\text{O}$ values of the post-Archean mantle peridotite samples are on average $\delta^{18}\text{O} = 5.21 \pm 0.08$ ‰ (1 s.d., $n = 15$), which is in good agreement with previous data for olivine in mantle peridotite (e.g., Mathey et al., 1994) (Table S-1). The post-Archean mantle peridotite samples have an average olivine $\Delta^{17}\text{O}$ value of -51.6 ± 2.1 ppm (1 s.d., $n = 15$; Fig. 1a), which is in line with data for mafic and ultramafic rocks from previous studies (Herwartz et al., 2014; Greenwood et al., 2018; Cano et al., 2020). No variations in olivine $\Delta^{17}\text{O}$ values were found for the post-Archean mantle peridotite samples. The peridotite samples from the Archean ultramafic enclaves in the Itsaq Gneiss Complex and Fiskefjord region show a much wider range in olivine $\delta^{18}\text{O}$ values ($\delta^{18}\text{O} = 2.8\text{--}6.2$ ‰) (Table S-2). Olivine $\Delta^{17}\text{O}$ values of the early Archean ultramafic bodies are on average $\Delta^{17}\text{O} = -50.9 \pm 3.2$ ppm (1 s.d., $n = 23$), a value that is

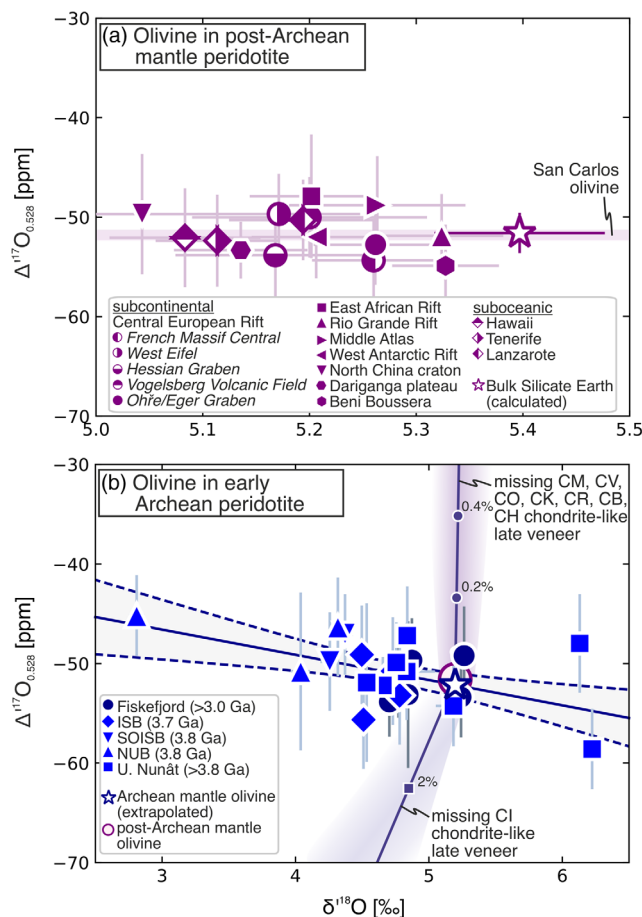


Figure 1 Plots of $\Delta^{17}\text{O}$ vs. $\delta^{18}\text{O}$ (± 1 s.e.m.) of olivine from ultramafic rocks. (a) Olivine in peridotites that were erupted as xenoliths in Phanerozoic magmas and from the Beni Boussera massif, relative to the composition of San Carlos olivine (horizontal line, $\Delta^{17}\text{O} = -51.8$ ppm; Supplementary Information S-2). The composition of the bulk silicate Earth (star) is calculated from the mean olivine compositions from this study ± 1 s.d. and is based on -0.2 ‰ fractionation in $\delta^{18}\text{O}$ and a θ value of 0.53 for fractionation between olivine and bulk peridotite. (b) Olivine from early Archean ultramafic enclaves with excess s-process Ru (Fischer-Gödde et al., 2020). The curve corresponds to a linear fit ± 1 s.d. through the Archean peridotite data in $\delta^{17}\text{O}$ versus $\delta^{18}\text{O}$ space and is used to extrapolate the compositions of the samples, which were in part altered by fluid-rock reactions, to the $\Delta^{17}\text{O}$ of pristine olivine in the early Archean mantle with $\delta^{18}\text{O} = 5.21$ ‰ (star). Shaded fields correspond to calculated olivine compositions if carbonaceous chondrites would be missing from the ultramafic enclaves, considering all carbonaceous chondrite meteorites for which data are available in the Meteoritical Bulletin Database as possible end member compositions for the late veneer (Table S-3). Curves are shown for missing late veneer with the average composition of CM, CV, CO, CK, CR, CH, CB chondrites and the average composition of CI chondrites, respectively, with the percentages of missing late veneer indicated relative to the mass of the Earth.

indistinguishable from the $\Delta^{17}\text{O}$ value of post-Archean mantle olivine. The early Archean peridotite samples show decreasing olivine $\Delta^{17}\text{O}$ values with increasing $\delta^{18}\text{O}$ values (Fig. 1b).

Discussion

Effects of fluid-rock reactions. Olivine $\delta^{18}\text{O}$ values of the early Archean peridotites predominantly deviate towards lower values



compared to post-Archean mantle olivine. We reported similarly low and variable olivine $\delta^{18}\text{O}$ values for a >2.7 Ga peridotite body that is entrained in the Kuummiut terrane in southeast Greenland, for which the low and variable $\delta^{18}\text{O}$ values reflect the effects of fluid-rock reactions that were followed by dehydration of the peridotitic protoliths at high grade metamorphic conditions (Peters *et al.*, 2020). Likewise, we suggest that the ultramafic enclaves in the Itsaq Gneiss Complex and Fiskefjord region experienced hydration and dehydration events during their metamorphic history that modified their primary olivine $\delta^{18}\text{O}$ values, a process that is likely common for ultramafic rocks in Archean high grade metamorphic terranes (Nishio *et al.*, 2019) (Supplementary Information S-1). Fluid-rock reactions cannot only have shifted the $\delta^{18}\text{O}$ values of the Archean peridotite samples, but may also have altered the $\Delta^{17}\text{O}$ values of the peridotite samples (*e.g.*, Sengupta and Pack, 2018). In order to obtain the $\Delta^{17}\text{O}$ value of pristine olivine in the ultramafic rocks before they interacted with fluids, we extrapolated the measured $\Delta^{17}\text{O}$ values to the $\delta^{18}\text{O}$ value of typical mantle olivine (this study; $\delta^{18}\text{O} = 5.21\text{‰}$) (Fig. 1b). The extrapolated $\Delta^{17}\text{O}$ value of pristine olivine in the early Archean mantle is -52.3 ± 1.7 ppm (1 s.d.); a value that is only 1.4 ppm lower than the average $\Delta^{17}\text{O}$ of the uncorrected data.

Implications for missing late veneer. The suggested $\Delta^{17}\text{O}$ value of pristine olivine in the early Archean ultramafic rocks is indistinguishable from the mean $\Delta^{17}\text{O}$ value of olivine in post-Archean mantle peridotite (Fig. 1b). A pre-late veneer signal in the Archean ultramafic enclaves is thus not resolved with respect to oxygen isotopes. This conclusion corroborates earlier results from a laser fluorination study on the Acasta Gneiss Complex and Isua supracrustal belt (Rumble *et al.*, 2013), and lower resolution SIMS data for the Jack Hills zircons, as well as zircons from the Acasta Gneiss Complex and (Valley *et al.*, 2014; Reimink *et al.*, 2018). Using the new data, we calculated the uppermost likely limits for missing late veneer from the Itsaq Gneiss Complex and Fiskefjord mantle, assuming the compositions of carbonaceous chondrite groups as the missing components (*cf.* Fischer-Gödde *et al.*, 2020). In general, and by definition, the compositions of the mantle before the late veneer (hereafter ‘pre-late veneer mantle’; PLVM), the bulk silicate Earth and the late veneer plot on a mixing line in $\delta^{17}\text{O}$ versus $\delta^{18}\text{O}$ space. The slope m and intercept i of this mixing line are given by the $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ values of the bulk silicate Earth and the composition of the late veneer. In $\delta^{17}\text{O}$ versus $\delta^{18}\text{O}$ space, the composition of the pre-late veneer mantle also plots on a line with slope 0.528 and intercept $\Delta^{17}\text{O}$ that equals the value for the pre-late veneer mantle. The point of intersection of the two relations in $\delta^{17}\text{O}$ versus $\delta^{18}\text{O}$ space is given by:

$$0.528\delta^{18}\text{O}_{\text{PLVM}} + \frac{\Delta^{17}\text{O}_{\text{PLVM}}}{1000} = 1000 \ln \left[m e^{\frac{\delta^{18}\text{O}_{\text{PLVM}}}{1000}} - m + \frac{i}{1000} + 1 \right] \quad \text{Eq. 2}$$

and can be approximated with the Newton-Raphson method. The intersection δ values then allow determining the oxygen atom fraction of missing late veneer from the pre-late veneer mantle (Supplementary Information S-4).

Based on Equation 2 we modelled possible masses of missing late veneer if carbonaceous chondrite groups would be missing from the Itsaq Gneiss Complex and Fiskefjord mantle, using a Monte Carlo approach (Fig. 2). In each Monte Carlo run, the end member composition of the late veneer was selected by randomly sampling a meteorite composition from the carbonaceous chondrite groups of interest, *i.e.* each meteorite specimen from a given selection of carbonaceous chondrites was

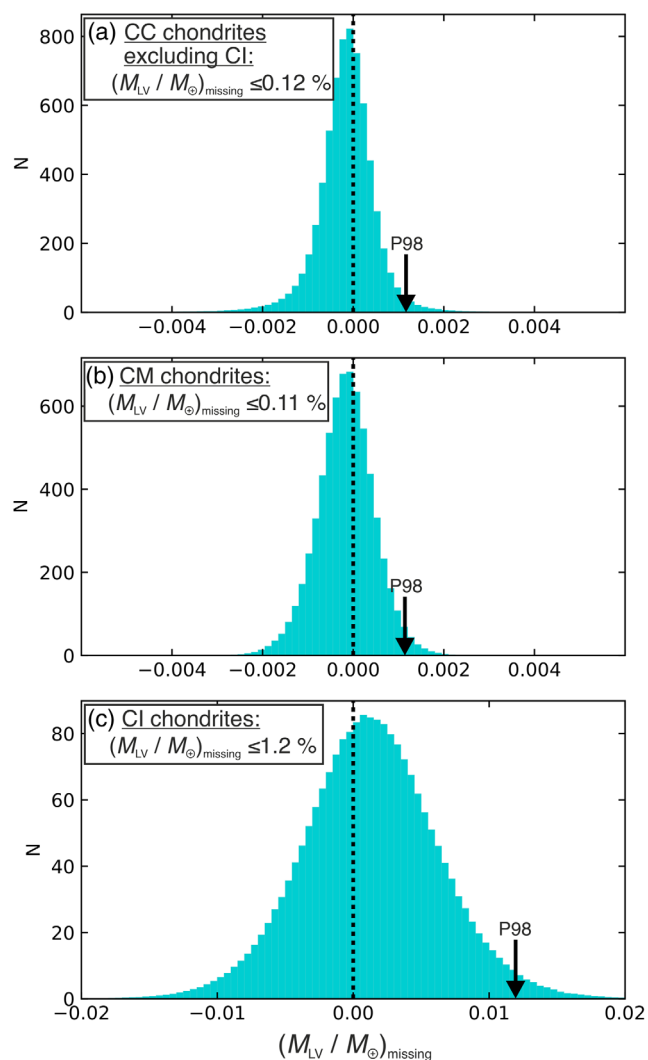


Figure 2 Missing late veneer from the Itsaq Gneiss Complex and Fiskefjord mantle relative to the mass of the Earth, $(M_{LV}/M_{\oplus})_{\text{missing}}$, calculated from the data in this study and from available triple oxygen isotope data for carbonaceous chondrites (Monte Carlo simulation; 10^6 runs). End member compositions of the late veneer were sampled randomly for given chondrite groups from a compilation of all meteorite data that are available in the Meteoritical Bulletin Database (Table S-3). The $\delta^{18}\text{O}$ value of the silicate Earth was considered to be 0.2 ‰ higher than olivine in mantle peridotite from this study, *i.e.* $\delta^{18}\text{O}_{\text{BSE}} = 5.41\text{‰}$. The 97.72nd percentile (P98) of the outcomes are shown by arrows and are given in the panel headings as percentages; and are considered uppermost bounds for missing late veneer. Outcomes with negative values correspond to hypothetical scenarios in which the Itsaq Gneiss Complex and Fiskefjord mantle contain excess late veneer. Dotted lines show outcomes where $(M_{LV}/M_{\oplus})_{\text{missing}} = 0$. (a) Missing carbonaceous chondrites, excluding CI chondrites. (b) Missing CM chondrites only. (c) Missing CI chondrites only.

considered equally likely to represent a possible end member composition of the late veneer (Table S-3). We adopted concentrations in the mixing calculations of 46.5 wt. % oxygen in carbonaceous chondrites and 44.33 wt. % oxygen in the terrestrial mantle (Palme and O'Neill, 2003). In the simulations, the $\Delta^{17}\text{O}$ values for the bulk silicate Earth and the pre-late veneer mantle were sampled from normally distributed populations that are described by the mean values and standard deviations for post-Archean mantle peridotite and the extrapolated $\Delta^{17}\text{O}$ value for pristine olivine in early Archean peridotite from this

study, respectively. We consider the 97.72nd percentile of the outcomes of the simulations as the uppermost likely values for the missing late veneer. These uppermost values correspond to a missing late veneer component relative to the Earth's mass (M_{\oplus}) of $\leq 0.12\% M_{\oplus}$ if the missing component would resemble CM, CV, CO, CK, CR, CH, CB, but not CI chondrites (Fig. 2a); $\leq 0.11\% M_{\oplus}$ if the missing component would resemble only CM chondrites (Fig. 2b); and $\leq 1.2\% M_{\oplus}$ if the missing component would resemble CI chondrites (Fig. 2c).

Our modelling results have implications for the materials that comprised Earth's late veneer. Fischer-Gödde *et al.* (2020) estimated based on their Ru isotope data that a late veneer component of up to $0.3\% M_{\oplus}$ of carbonaceous chondrite-like materials could be missing from the Itsaq Gneiss Complex and Fiskefjord mantle, and favoured CM chondrites as the missing component. The oxygen isotope data, in contrast, imply that only a much smaller component of carbonaceous chondrites ($\leq 0.12\% M_{\oplus}$) can possibly be missing from the early Archean ultramafic enclaves with respect to oxygen, unless this component comprises CI chondrites. We propose two end member scenarios that best reconcile the triple oxygen isotope data with the available Ru isotope data. In one end member scenario, the missing late veneer component indeed resembles CM chondrites with the equivalent mass of $\leq 0.11\% M_{\oplus}$ (Fig. 2b). This scenario implies that the early Archean mantle had incorporated more than *ca.* >86 % late veneer with respect to lithophile elements, which agrees with some estimates from the concentrations of highly siderophile elements in the early Archean mantle (van de Löcht *et al.*, 2018), but not with others (Dale *et al.*, 2017). The missing CM chondrite-like component was possibly complemented by a missing component of carbonaceous group iron meteorite-like materials, *e.g.*, materials similar to the IID and IVA irons that have comparable Ru isotope compositions compared to carbonaceous chondrites (Fischer-Gödde and Kleine, 2017) and high Ru concentrations, but are deficient in O. This scenario is also consistent with highly siderophile element concentrations and $^{187}\text{Os}/^{188}\text{Os}$ ratios in lunar impact rocks and in the upper mantle (Fischer-Gödde and Becker, 2012). In the second end member scenario, the missing late veneer component resembles the composition of CI chondrites (Fig. 2c). This scenario is feasible only if the s-process Ru deficit in CI chondrites is greater than was considered in the modelling by Fischer-Gödde *et al.* (2020). We consider this a reasonable suggestion, because the Ru isotope composition of CI chondrites is constrained at present by a single meteorite specimen only, whereas other carbonaceous chondrite groups show internal Ru isotope variations (Fischer-Gödde and Kleine, 2017).

The discussion of our data thus far concerns the mass and composition of a late veneer component that can possibly be missing from the early Archean mantle. It can be assumed instead that the early Archean mantle was deficient in 40 % late accreted materials (Dale *et al.*, 2017). Following this line of logic, the triple oxygen isotope data imply a maximum late accreted mass of $\leq 0.3\% M_{\oplus}$ for CM, CV, CO, CK, CR, CH, CB chondrites; and $\leq 3\% M_{\oplus}$ CI chondrites. In conjunction with the absolute concentrations of highly siderophile elements in the post-Archean mantle, this conclusion would imply that the late veneer contained abundant CI-like materials; a scenario that agrees well with volatile element patterns (Braukmüller *et al.*, 2019) and the Se isotope composition of the post-Archean mantle (Varas-Reus *et al.*, 2019).

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2120>.



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