Oxygen isotope ($\delta^{18}\text{O}$, $\Delta^{17}\text{O}$) insights into continental mantle evolution since the Archean

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Oxygen isotope ($\delta^{18}\text{O}$, $\Delta^{17}\text{O}$) insights into continental mantle evolution since the Archean

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Abstract: The proportions of stable isotopes of oxygen change little in geologic processes at high temperatures, but are strongly fractionated near the Earth's surface, and thus are robust indicators of recycling of surface materials to Earth's mantle. Here we document a subtle but significant (by 0.15-0.2‰) temporal decrease in $\delta^{18}\text{O}$ in the continental lithospheric mantle (CLM) from the Archean to the Phanerozoic eons both for its major mineral (olivine, $5.37 \pm 0.07\text{‰}$ to $5.16 \pm 0.10\text{‰}$) and bulk rocks (peridotites), which we link to the initiation of plate tectonics at the end of the Archean (~ 2.5 - 2.0 Ga). The CLM is made of accreted melting residues of the asthenosphere, usually created or transformed in mantle wedges above subduction zones, fluxed by low- $\delta^{18}\text{O}$ fluids from subducting slabs. We posit that $\delta^{18}\text{O}$ in the oldest Archean CLM, uncontaminated by subduction, provides the best $\delta^{18}\text{O}$ estimate for the BSE of 5.37‰ for olivine and 5.57‰ for bulk peridotite, values that are comparable to lunar rocks as the moon did not have plate tectonics. Younger samples document a decrease and greater heterogeneity of $\delta^{18}\text{O}$ in the geologic record due to the development and progression of plate tectonics. Furthermore, mass balance considerations suggest that, given large volume of the CLM, even small decreases in its $\delta^{18}\text{O}$ may explain increasing $\delta^{18}\text{O}$ of the continental crust since oxygen is progressively redistributed by fluids between these reservoirs.

One-Sentence Summary: $\delta^{18}\text{O}$ of the continental lithospheric mantle has decreased by 0.15‰ since the Archean due to initiation of plate tectonics and recycling in subduction zones, explaining crust-upper mantle mass balance

Introduction. Oxygen isotopes may be the best tool in addressing the effects of water-rock interaction and recycling of surface-exposed materials [1-4]. Oceanic crust, created at mid-ocean ridges (MOR), undergoes pervasive hydrothermal exchange with seawater, leading to heavy $\delta^{18}\text{O}$ (+7-13‰) at the top in pillow basalts and lower $\delta^{18}\text{O}$ (+2-5‰) at the bottom in gabbros and peridotites [1]. Olivine with higher and lower $\delta^{18}\text{O}$ than the average olivine phenocrysts from mantle-equilibrated MOR basalts has been found in several island arcs around the world [2, 5] and attributed to the dehydration or melting of this altered oceanic crust during subduction. Eclogites that represent metamorphosed basalts in subducting slabs display this range of $\delta^{18}\text{O}$ values, suggesting that hydrothermally-altered slabs were subducted in the past [6,2].

The exact O isotope composition of the Bulk Silicate Earth (BSE) is essential to understand the Earth's accretion from different types of parental meteoritic reservoirs, yet is to be well-constrained. Even small $\delta^{18}\text{O}$ variations in global reservoirs are significant because oxygen is by far the most abundant chemical element in the BSE. Estimates range from 5.1 to 5.9‰, depending on whether the value is derived from data on meteorites or terrestrial samples [7-8].

Initiation of plate tectonics on Earth happened in the Archean but the style of the earliest Archean tectonics may have been very different to today, dominated by "sagduction" due to the predominance of plume-related activity [9]. In such a regime, progressive accretion of basaltic lavas on the surface and delamination of the deep lithosphere results in a limited amount of recycling of the surface-altered material (with respect to $\delta^{18}\text{O}$) into the convecting mantle. Subduction is capable of efficient delivery of surface-altered materials via melts and/or fluids, with both low- and high- $\delta^{18}\text{O}$, into the mantle and directly under the continental lithosphere. It follows, therefore, that the Archean mantle, similar to lunar rocks, should have been less affected by subducting slabs, thus more closely reflecting the BSE.

Of particular importance is the question of potential differences between the Earth's earliest mantle, represented by xenoliths (fragments) of tentative Archean age in volcanic rocks, and younger mantle samples. As Archean mantle was most likely 100-200°C hotter than today, degrees of mantle melting were greater (up to 40%) leading to the generation of komatiites and leaving behind melt-depleted harzburgites [10, 11]. As these highly refractory residues were Fe-depleted, they were less dense and resulted in the formation of rigid and stagnant keels of Archean continental lithosphere [3]. This difference can be clearly seen in the composition (e.g., olivine Mg#, defined as $\text{Mg}/(\text{Mg}+\text{Fe})$) of Archean and modern peridotites [10, 11].

The $\delta^{18}\text{O}$ values of peridotite xenoliths from many tectonic settings have been inferred to be roughly uniform irrespective of bulk composition and mineralogy (e.g. spinel vs garnet facies, fertile vs. refractory) ever since Matthey et al. (1994) [6] employed new laser fluorination methods to generate an expansive dataset of global peridotites, and inferred these to be uniform within analytical error ($\sim\pm 0.1\%$). Later studies of even strongly metasomatized, hydrous (amphibole-bearing) peridotites reported values within the canonical mantle ranges [12].

Previous studies, including those employing secondary ionization mass spectrometry (SIMS) (within $\geq 0.2\%$ precision), found a range from 5.0 to 5.5‰ for olivine from different peridotite xenoliths, attributed to a subtle diversity of depletion/enrichment processes, but no covariation of $\delta^{18}\text{O}$ for with composition or settings [13, 4].

Laser fluorination, with its error on the order of 0.01 to 0.08‰, continues to be the most precise method to resolve small 0.1-0.3‰ differences in $\delta^{18}\text{O}$. The analytical effort and a dataset presented here for 104 individual mantle xenoliths spans 13 years but is based on a single set of standards, analytical methodologies and normalization procedures (Tables A1). The samples are previously well-characterized xenoliths with available modal, petrographic and chemical data pertinent to the depletion/enrichment events (Mg#, Al_2O_3 , CaO) as well as equilibration temperatures (14-18, and references in the supplementary).

Results

Figs. 1-2 present temporal oxygen isotopic ($\delta^{18}\text{O}$, $\Delta^{17}\text{O}$) trends for our worldwide peridotite xenolith suite from cratonic, off-craton and mantle wedge settings spanning ~ 3 Ga. The formation (melt extraction) ages for cratonic xenoliths were estimated from Re-Os isotope data [14-19, Appendix] using Re-depletion (T_{RD}) estimates that presume complete Re extraction from residues of BSE melting and thus are only valid for highly refractory rocks (harzburgites and dunites). The T_{RD} values are slightly lower than ideally more correct model Re-Os ages (T_{MA}), yet allow to account for common post-melting Re-enrichments in xenoliths due to metasomatism and host magma infiltration responsible for common aberrant T_{MA} values [3,14,17]. For consistency, we attribute the same age to coeval rock types from each locality, e.g. 2.9 Ga and 2.6 Ga for dunites vs. 2.0 Ga for harzburgites from Udachnaya. These estimates match main stages of cratonic crust formation supporting their validity [16-17, 19].

Re-Os ages cannot be systematically used to date off-craton peridotites that range broadly in composition, but are generally more fertile than in cratons, i.e. most of them lost little (if any) melt, hence Re such that valid T_{RD} ages cannot be obtained. We presume that the off-craton CLM formed during the most recent crustal growth or accretion event in a given lithospheric domain [42]. Harzburgite xenoliths from west Pacific represent residual oceanic mantle, likely melted first at mid-ocean ridges, then in mantle wedge. The Kamchatka samples are presumed to be ~ 0.2 Ga old (the oldest large-scale oceanic crust in the region). T_{RD} estimates (0.1-1.1 Ga) are used for the West Bismarck samples because some of them may come from older microcontinents.

When plotted vs age (Fig. 1) $\delta^{18}\text{O}$ values of olivine exhibit a temporal ~ 0.2 ‰ decrease from 5.38‰ in the Archean, to 5.21‰ in the Paleoproterozoic and 5.16‰ in the Phanerozoic. The $\delta^{18}\text{O}_{\text{Opx}}$ decreases as well. The bulk $\delta^{18}\text{O}$ values (estimated via modal abundances of minerals and their measured or computed $\delta^{18}\text{O}$ values at last equilibration temperatures) shows a sub-parallel trend of comparable magnitude. This precludes interpretation of this trend as due cooling or melting effects shown and discussed in Fig. A1. Importantly, the differences between the Archean and Proterozoic olivine are observed not only globally but locally in two kimberlite pipes in Siberia (Udachnaya and Obnazhennaya) (Fig 1a) [15,17]. Olivine modal abundance in our predominantly harzburgitic dataset stays nearly constant at $76 \pm 6\%$ from 3Ga to 2Ga. Mg# in our dataset displays an overall decrease from 0.93-0.94 in the Archean-early Proterozoic to ≤ 0.92 after that, consistent with greater melting degrees and pressures for the ancient mantle [10, 11]. If corrected for prior melt depletion using Mg# and Al_2O_3 from the compositional model of [11], the Archean samples come as $\sim 10\%$ more melt-depleted than post-Archean (Fig. 1c, Table A1), perhaps due to hotter regime in the Archean. This leads to a more pronounced negative trend with age (Fig. 1c), and the overall estimate of temporal $\delta^{18}\text{O}_{\text{bulk}}$ peridotite difference between the Archean and Phanerozoic samples is further increased to 0.18‰. T-test returns p values of

<0.001. Earliest Archean xenoliths return $\delta^{18}\text{O}$ values comparable to that of the Moon and are used here to estimate the BSE.

Last equilibration temperatures estimated with the Ca-in Opx thermometer are lower for the higher- $\delta^{18}\text{O}$ Archean cratonic xenoliths than for off-craton ones, but display no trend vs. age (Table A1). Lower temperature should result in lower (not higher) $\delta^{18}\text{O}_{\text{Olivine}}$ values of olivine at identical $\delta^{18}\text{O}_{\text{Bulk}}$ (cf. Fig. A1) and normalization to a single temperature would increase $\delta^{18}\text{O}_{\text{OI}}$ for the Archean, and further decrease it for hotter recent samples, amplifying the temporal trend.

Triple oxygen isotope analyses of a selection of olivines (Fig. 2) yield an overlap in values: $\Delta^{17}\text{O}_{0.5305} = -0.047 \pm 0.011\text{‰}$ ($n=9$) for Archean vs $-0.051 \pm 0.010\text{‰}$ ($n=12$) for the early-Paleoproterozoic samples. The value for San Carlos Olivine, likely sampling younger off-craton Phanerozoic (≤ 1.8 Ga) mantle of N America ([20] is -0.051‰ ([21]. Vein orthopyroxenes with minor amphibole from the Kamchatka arc, most likely formed due to subduction fluid/melt percolation [43], yield a mantle-like value $-0.052 \pm 0.005\text{‰}$ [21]. Modeling demonstrates that fluxing with fluids with any reasonable $\delta^{18}\text{O}$ (0 to $+10\text{‰}$) or $\Delta^{17}\text{O}$ ($+0.02$ to -0.02‰) values will have far greater impact on $\delta^{18}\text{O}$ than on $\Delta^{17}\text{O}$, resulting in largely constant mantle-like $\Delta^{17}\text{O}$.

Discussion

The decreasing olivine $\delta^{18}\text{O}$ trend that we observed made us search for similar trends in other published datasets, of which Matthey et al. (1994) [6] is the most comprehensive. When assigning ages to their xenolith suites (Table S2) we observe a trend similar to ours (Fig. S3): decreasing $\delta^{18}\text{O}_{\text{Olivine}}$ and $\delta^{18}\text{O}_{\text{Bulk}}$ values and constant or even decreasing $\delta^{18}\text{O}_{\text{Opx}}$ values.

We experimented with an alternative age assignment to the samples studied here and by Matthey et al. (1994) [6], by simply using model formation ages of the host lithosphere (e.g. [20] Fig. S4). This results in similarly decreasing trends, signifying the conclusion that the obtained subtle temporal trends are not fortuitous results of age assignments.

The average for $\delta^{18}\text{O}_{\text{Olivine}}$ in our entire datasets (5.224 ± 0.120 , st dev, $n=174$) is heavier by 0.04‰ to that for data presented by Matthey et al. (1994) [6], which we attribute to small differences in standardization. The computed average bulk values of $5.34 \pm 0.13\text{‰}$ is identical to [6]. We thus consider the average values of two datasets as compatible and the best current estimates of the $\delta^{18}\text{O}$ lithospheric mantle (Table A4). Below we further consider its evolution.

The 5.37‰ value for Archean olivine and 5.44‰ bulk value for mean Archean refractory peridotite of our dataset with the average 79% modal olivine and 16% modal orthopyroxene enables correction for a decrease in $\delta^{18}\text{O}$ due to prior melt depletion as is explained in Fig A1a,b. Translating this harzburgitic bulk into a typical original lherzolitic assemblage of 56% olivine and 26% orthopyroxene [22] leads to a 0.12‰ increase for the value of the original lherzolitic bulk peridotite. We thus obtain a value of 5.57‰ as the best estimate of the primary mantle source (Fig. 1c) that characterizes the original BSE prior to melting in the Archean. The olivine $\delta^{18}\text{O}$ value of $5.37 \pm 0.07\text{‰}$ (1 st dev, $n=20$) recorded in the Archean olivine would stay the same and may thus reflect the best estimate for olivine in the BSE.

Constraining $\delta^{18}\text{O}$ for mantle older than 3.0-3.5 Ga is hampered by the lack of peridotites, hence olivine, formed at the time. Here, we use crustal zircon as a proxy (see Appendix for justification) for mantle olivine, since zircons of such age exist in mantle-derived crustal rocks, and because zircon-olivine O isotope fractionation at mantle temperatures is well constrained

($0.1 \pm 0.1\%$ [23]). For example, Kaapvaal craton zircons of Archean age exhibit the range of $5.32 \pm 0.17\%$ [24]. Lunar zircon of presumably 4.4-4.3 Ga age has $\delta^{18}\text{O}$ of 5.6% [24] and lunar olivines and basalts are 0.05% and 0.18% heavier than terrestrial ones respectively [25]. As the Moon has not had plate tectonics, it thus provides a bench mark for the pre-plate tectonics Earth since both planets re-accreted after the Giant Impact (4.44 Ga) and had magma oceans [7,8]. These heavier values agree with our extrapolated estimates of $\delta^{18}\text{O}_{\text{BSE}} = 5.57\% \pm 0.07\%$ (based on propagated errors) very well.

Having discounted the possibility that melting and sub-solidus cooling can account for the observed temporal trends in $\delta^{18}\text{O}$ (Fig. A1), we next consider the possibility of time-integrated effects of recycling hydrothermally altered lithospheric materials (Fig. 3).

First, direct hydrothermal alteration (serpentinization and chloritization) of the shallow mantle in areas accessible to seawater percolation occurs today and in the geologic history at midocean ridges, as well as, importantly, in areas where plates bend in front of subduction trenches [37]. Any such hydrothermal interaction at $T > 200^\circ\text{C}$, plausible in terms of typical sub-Moho temperature gradients, would generate lower- $\delta^{18}\text{O}$ peridotites [1-2]. These low- $\delta^{18}\text{O}$ peridotites could have accreted under the continents upon initiation of horizontal motion.

Second, such low- $\delta^{18}\text{O}$ portions of subducted slabs expel low $\delta^{18}\text{O}$ fluids and thus modify the $\delta^{18}\text{O}$ of the overlying mantle wedge peridotites. Given the $\delta^{18}\text{O}$ distribution in the altered oceanic crust, which ranges from high- $\delta^{18}\text{O}$ at the top to low- $\delta^{18}\text{O}$ in the interior ([1, 5, 26], upon subduction and slab heating the high- $\delta^{18}\text{O}$ fluid will be lost first at the fore-arc, while dehydration of the low- $\delta^{18}\text{O}$ interior of the slab would occur deeper into the mantle wedge as the slab geotherm progressively increases [41]. The modern analogues to Archean subduction are modern “hot” subduction zones, such as the Central American Arc. Eiler et al. (2005) [26] argued that the subtly low- $\delta^{18}\text{O}$ value of the magmas in the central part of this arc results from low- $\delta^{18}\text{O}$ fluids, and greater (2-4%) addition of such fluids corresponded to greater (15-25%) degree of mantle melting. Our sample collection has two Phanerozoic arc-related xenolith suites: harzburgites from the Kamchatka [27] and the West Bismarck arcs [28]. Both of them occupy the low- $\delta^{18}\text{O}$ end of our sample array, supporting the proposed scenario of fluxing by low- $\delta^{18}\text{O}$ fluids. In other, shallower areas of the arc, fluids can be high- $\delta^{18}\text{O}$ and normal (mantle-like)- $\delta^{18}\text{O}$, contributing to higher $\delta^{18}\text{O}$ values of accretionary sediments and the crust. Strongly re-worked mantle peridotites (deformed and modally metasomatized at high temperature and pressures) by invoked subduction fluids shortly before magmatic entrainment, show shifts to lower (not higher) $\delta^{18}\text{O}$ values (down by 1% , [36]).

Therefore, the reported, previously unrecognized, temporal decrease in $\delta^{18}\text{O}$ value of peridotites, dominated by continental harzburgite, is likely due to a billion-year record of seawater-peridotite interaction in areas of spreading and plate bending, and the subduction-accretion of these materials to the CLM. If we consider the entire 100 km-thick section of such mantle, at present subduction rates of 8 cm/yr it would take 2 Gyr to lower the value of peridotite by 0.15% . This computation assumes that the remaining 1-2 wt% of H_2O is released from the slab into the overlying peridotite as a low- $\delta^{18}\text{O}$ fluid (0 to $+2\%$) due to the breakdown of serpentine and chlorite ~ 100 -200 km below the mantle wedge [41]. This estimate is likely robust within a factor of 2 to 3 depending on the choice of water content, its isotopic composition, and total volume of the mantle affected. Perhaps more importantly, only a certain fraction of the mantle under volcanic fronts may be affected, compared to neighboring areas, or by different fluids. This depends on the mechanisms and timescales of fluid migration/fluid-rock reaction and would potentially produce a range of $\delta^{18}\text{O}$ for xenoliths from a single locality. This explains

resolvable $\delta^{18}\text{O}$ heterogeneity observed among xenoliths from a single kimberlite pipe (Udachnaya and Obnazhennaya [Fig. 1a, 16-17, 19]), or xenoliths from variously fluxed mantle wedges in the Phanerozoic Kamchatka or West Bismarck arcs [27,28].

The trend established here for the isotopic composition of a major element, oxygen, places the initiation of “modern-style” plate tectonics at the end of the Archean [29, 9], though rudimentary on-off plate tectonic events may have existed earlier [30-31] from data on shales and granites [32]. A similar approach for other chemical and isotopic systems that record interaction with the hydrosphere may better quantify the onset of mantle fluxing and rehydration of the mantle after the beginning of modern-style plate tectonics. For example, the irreversible low- $\delta^{18}\text{O}$ flux from subducting hydrated slabs may explain the contemporaneous increase of $\delta^{18}\text{O}$ in the continental crust (Fig. 4) and remaining hydrosphere, as documented by surface sedimentary archives [32 and refs therein], a long-standing controversy in Earth sciences [34, 39]. Furthermore, this could also explain the secular drop in the sea-level [38] contributing to continental emergence after the Archean. Given that the mass of the CLM is 10-20x that of the continental crust, a decrease by $\sim 0.2\%$ in $\delta^{18}\text{O}$ of the CLM on a Ga time scale would suggest an increase of up to 4‰ in the crust (Fig. 4) without taking into account the subduction-affected materials to convecting mantle. Such a calculation assumes that most of oxygen isotope mass balance proceeds via surface tectonics and fluid transfer (hydration-rehydration) and is mostly contained to within lithospheric plates of 100-250 km. Although not considered in this work, sunken slabs with residual water would additionally transfer more ^{18}O -depleted oxygen (Fig. 3), working in the same direction of contributing to the overall increase in $\delta^{18}\text{O}$ of the crust, hydrosphere, and surface materials [32, 39].

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Methods

Oxygen isotope measurements for $\delta^{18}\text{O}$ relied on 1 to 1.5 mg of material, predominantly single crystals and were performed by laser fluorination and gas-source mass spectrometry (MAT253) at the University of Oregon, and the effort span 13 years. We used purified BrF₅ as a reagent and boiling Hg diffusion pump to get rid of excess of F₂ gas, then converted purified O₂ into CO₂ and

run it in a dual inlet mode on MAT253 mass spectrometer, integrated with the vacuum line, as this method is most precise for $\delta^{18}\text{O}$ determination (e.g. [2,5]). Samples yields were measured in a calibrated volume using a Baratron gauge are reported in Table A1. San Carlos Olivine ($\delta^{18}\text{O}=5.25\text{‰}$, UWG2 garnet, 5.80‰ [23] and an indoor UOG garnet standard calibrated relative to the other two ($\delta^{18}\text{O}=6.52\text{‰}$) were used to calibrate the data on VSMOW scale. Each session included analyses of 4 to 6 standards, and correction for day-to-day variability was 0 to 0.2‰ . Standard CO_2 gas was used as a working standard and it was periodically rerun against OZTECH CO_2 gas supplied to us by Dr. Charles Douthitt. Errors for standards in individual sessions ranged from ± 0 to $\pm 0.11\text{‰}$ and on average is $\pm 0.06\text{‰}$. Samples from sessions with worse precision on standards were rerun and most data reported in Table A1 represents duplicates with some triplicates/quadruplicates run in different sessions and are averaged with shown 2 st err on replicate measurements. Yields were close to 100‰ and when plotted vs $\delta^{18}\text{O}$ demonstrated no correlation. Coexisting orthopyroxenes, clinopyroxenes and garnets were also analyzed in many samples. For triple oxygen isotope analyses generated gas was run as O_2 in sessions with SCO standard ($\Delta^{17}\text{O} = -0.052\text{‰}$, [21] and utilized a triple O continuous flow line constructed for most precise triple O measurements [32]. The generated O_2 gas was put through the 8 ft long gas chromatographic column at room temperature for its purification from NFx compounds. Generated gases were additionally frozen on a 5A zeolite and then released into the bellow of the mass spectrometer by LN2-ethanol mix over 10 min. GC and zeolite traps were degassed with flowing He at 200°C for 15-20 min between each sample. The purified O_2 gas was run 5 times 8 cycles in a dual inlet mode, against a well calibrated oxygen gas standard on a MAT253 isotope ratio mass spectrometer. Triple O isotope values for $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$ are reported in Table A2.

Fig. 1. Temporal trends of $\delta^{18}\text{O}$ olivine (a, b), orthopyroxene and bulk (b) and lithospheric mantle CLM corrected for melt depletion (c) in studied mantle nodules. Bulk is computed based on modal mineral abundances and $\delta^{18}\text{O}$ values of olivine and orthopyroxene (Table A1). Zircon from the Archean Kaapvaal cratonic mantle is from [23]; and zircon and basalts from the moon is from [24-25], lunar mantle estimate is based on zircon and olivine (see supplementary).

Fig. 2 Triple oxygen isotope analyses of Archean and post-Archean olivines and orthopyroxenes, showing overlap with San Carlos olivine. Expected trends to higher- $\Delta^{17}\text{O}$ and lower $\delta^{18}\text{O}$ subduction fluids, derived from low- $\delta^{18}\text{O}$ interiors of subducting slabs are shown (e.g. [35]). The $\delta^{18}\text{O}$ in orthopyroxenes are higher than in equilibrium olivine by 0.5-0.7‰ and nearly identical in $\Delta^{17}\text{O}$ at mantle temperatures (Fig. A1, ref 33).

Fig. 3. A cartoon that explains cooling of the Earth and rehydration of the mantle by low- $\delta^{18}\text{O}$ fluids in spreading centers [1,4], plate bending zones [37], and subduction zones [26,33], generating progressively lower $\delta^{18}\text{O}$, and overall heterogeneous, continental lithospheric mantle due to accretion after initiation of plate tectonics. $\Delta^{17}\text{O}$ is not significantly modified by these processes (Fig. 2). Color code: yellow: low- $\delta^{18}\text{O}$ peridotites, red: high- $\delta^{18}\text{O}$ sediments and eclogites. a) Early Earth and moon regimes: degassed mantle, plume tectonics, original in $\delta^{18}\text{O}$

(=Bulk Silicate Earth) peridotites, b) modern plate tectonics regime: rehydration of mantle, plate tectonics, lower- $\delta^{18}\text{O}$ peridotites in the subcontinental lithosphere

Fig. 4 Co-evolution of continental lithospheric mantle (CLM, this work) and the Earth's crust expressed by three proxies: granites, bulk shales [32], and zircons [40]. Note change in $\delta^{18}\text{O}$ scale. Inset shows mass-balance of CLM and crust growth and evolution; 1:10 and 1:20 are assumed crust/CLM proportions based on modern average lithospheric thickness of 150 km. BSE is estimated in this work and corresponds to the starting point of crustal growth after the Giant Impact. Unidirectional temporal loss of light- $\delta^{18}\text{O}$ to the large reservoir of CLM helps to explain accumulation of heavy $\delta^{18}\text{O}$ in the crust and potentially hydrosphere (e.g., Muehlenbachs, 1998, refs. 39, 32).

SUPPLEMENTARY MATERIAL

Thermodynamic $\delta^{18}\text{O}$ trends of mantle melting

In order to understand isotopic effects related to progressive basaltic melt removal during mantle melting, we performed modeling of bulk, fractional and fluxed melting of original lherzolitic peridotite and computed the associated oxygen isotopic effects on coexisting minerals and bulk (Fig. A1a). We accounted for the effects of cooling after melt extraction to peridotite storage conditions, assuming uninterrupted equilibrium exchange between remaining minerals in the residual assemblage (Fig A1b). As the melt is 0.4-0.7‰ higher in $\delta^{18}\text{O}$ than the bulk (due to the predominant removal of higher $\delta^{18}\text{O}$ pyroxenes), fractional melting with removal of 30% of melt leads to 0.1‰ decrease in the bulk $\delta^{18}\text{O}$ values. However, the noticeable and counter-intuitive feature of the peridotite melting process is that the $\delta^{18}\text{O}_{\text{Olivine}}$ value increases during formation of melting residues (harzburgite and dunites), as $\delta^{18}\text{O}_{\text{Olivine}}$ becomes closer to the bulk

values as its modal proportion increases. The $\delta^{18}\text{O}$ values of orthopyroxene (and Cpx) also increase with melt removal, while the $\Delta^{18}\text{O}_{\text{Px-Ol}}$ remains the same, regardless if two minerals are from lherzolitic or harzburgitic assemblage. This simple peridotite behavior during its melting has not been discussed previously to our knowledge, but it is key to understand small (0.05-0.2‰) effects on $\delta^{18}\text{O}$ values discussed below. Such ranges are often found in different xenoliths and these could be due to different melt-depletion degrees and storage temperatures (Fig. A1b).

Fig. A1a. Computed isotope effects of batch (continuous lines) and fractional (dashed lines) peridotite melting from 0 to 45% on the equilibrium isotope values of melt and the residual mineral assemblage. Starting composition at low-T has 56% olivine, 26% Opx, 15% Cpx and 3% of Spinel. Isotope fractionation factors are from Chiba et al. (1989) and Rosenbaum et al (1993). For melt we assumed $A=1.3$. Isotope effects are computed against phase change and proportions (% melt) trends observed by Baker and Stolper (1994) in their peridotite melting experiments, also modeled by Ghiorso et al. (2002) in pMELTS. Fractional melting assumed removal of equilibrium fractional melt shown on the diagram. Flux melting trends are computed using Rayleigh melting by water fluxing and up to 5% fractional melt removal with shown effect on bulk peridotite and residual olivine $\delta^{18}\text{O}$ values. In this case, it is assumed that water fluxing generates silica-rich andesitic hydrous melt/fluid with $\delta^{18}\text{O}=6.1\text{--}7.3\text{‰}$ in equilibrium with the ambient assemblage. Upper bound of the flux melting triangle is computed by assuming decreasing $\delta^{18}\text{O}$ from 7.3‰ to 6.1‰, while the lower bound assumes that the hydrous melts maintains constant high $\delta^{18}\text{O}=7.3\text{‰}$. 3b. Melt depletion, then cooling effects on the residual mineral $\delta^{18}\text{O}$ values. Notice that melt depletion results in *lowering* bulk $\delta^{18}\text{O}$, but it also results in higher modal olivine and the $\delta^{18}\text{O}_{\text{Ol}}$ values which are *higher* (closer to the bulk), while $\delta^{18}\text{O}_{\text{Opx}}$ is also higher; $\Delta^{18}\text{O}_{\text{Opx-Ol}}$ are constant.

Estimating $\delta^{18}\text{O}$ value of the starting peridotite based on estimated prior melt depletion.

Thermodynamic modeling above on $\delta^{18}\text{O}$ trends as a function of melting degree and chemical composition of studied xenoliths permit estimation of the $\delta^{18}\text{O}$ value of the original

fertile peridotite that was undergoing melting. This estimation is based on Al_2O_3 , MgO and $\text{Mg\#}$ by using experimental data of Herzberg (2004, ref. 11). The estimated melting degrees are approximate. Nonetheless, we gave 45% to AR dunites and lowest-Al (<0.5) harzburgites, 40% for Archean-Proterozoic low-Al, high-Mg# (>0.925) harzburgites, 30-40% for PR harzburgites with 0.5-1% Al_2O_3 , 20-25% for moderate-Al harzburgites, 30% to all Avacha harzburgites (28% to those with 0.8% Al_2O_3). We consider these melting degree estimates as sufficiently robust because estimation of $\Delta^{18}\text{O}_{\text{depleted-original}}$ peridotite only produces small effects on the $\delta^{18}\text{O}_{\text{OI}}$ or $\delta^{18}\text{O}_{\text{bulk}}$ (Fig. A1a), based on Fig. A1a and the estimates are presented in Table A1 are plotted on Fig. 1. The 25% prior melt removal leads to a 0.05-0.06‰ increase in $\delta^{18}\text{O}_{\text{OI}}$ and 0.09‰ decrease in $\delta^{18}\text{O}_{\text{bulk}}$ peridotite values. Absolute majority of subcontinental peridotites including those studied in this work are represented by comparably melt-depleted harzburgites with 28-40% estimated melt depletion (ref 11), so a relative change between $\delta^{18}\text{O}_{\text{bulk}}$ peridotite of different groups is 0.05-0.06‰. As Archean peridotites are more depleted than younger, applying this melt depletion correction to $\delta^{18}\text{O}_{\text{bulk}}$ peridotite leads to a more pronounced negative trend with age as compared to bulk harzburgite (Fig. 1) and the overall estimate of temporal $\delta^{18}\text{O}_{\text{bulk}}$ peridotite depletion is increased by 0.03-0.05‰. Accepting the melt depletion thus enhance trends of lowering of $\delta^{18}\text{O}_{\text{bulk}}$ peridotite with time. This leads to more accurate $\delta^{18}\text{O}_{\text{BSE}}$ estimate and brings it closer to the lunar values.

Fig. A1b effect of melt depletion along the melting path shown in Fig A1a for $\delta^{18}\text{O}_{\text{OI}}$ or $\delta^{18}\text{O}_{\text{bulk}}$. Notice nearly linear change.

Comparing lunar and terrestrial $\delta^{18}\text{O}_{\text{basalt}}$, $\delta^{18}\text{O}_{\text{Zircon}}$, and $\delta^{18}\text{O}_{\text{mantle}}$ values

As there are no fresh terrestrial olivines and peridotites older than ~ 3 Ga, Fig. 1 in main text compares lunar and terrestrial $\delta^{18}\text{O}_{\text{Zircon}}$ with $\delta^{18}\text{O}_{\text{mantle}}$. We rely on the below arguments involving high-temperature isotopic fractionations between these materials on earth and the moon. Isotopic fractionations $\Delta^{18}\text{O}_{\text{melt-mineral}}$ are functions of temperature (as $1/T^2$), mineral identity, and composition of melt involved (Chiba et al. 1989; Eiler, 2001; Valley et al. 2003). The $\delta^{18}\text{O}$ of lunar zircons were measured by Valley et al. (2014) and are 5.61‰; these values are 0.3‰ heavier than typical terrestrial mantle zircons (5.3‰, Valley, 2003), most of which are Phanerozoic. Lunar zircon are measured to be lighter by 0.08-0.09‰ than lunar basalts (Valley et al. 2014), however typical 4.3-3.8 Ga low-Ti lunar basalts are 5.69‰, 0.12‰ heavier than the mode of modern terrestrial MORB (5.57‰). There are no coeval 4.3-3.8 Ga terrestrial basalts to compare with lunar basalts, thus lunar basalts can be used to suggest that coeval terrestrial basalts were also higher in $\delta^{18}\text{O}$. Such conclusion derived from compilation and measurements by Valley et al (2014) for lunar zircons and basalts directly supports the conclusion of our

present study that mantle peridotites (parental to basalts) evolved temporally to lower $\delta^{18}\text{O}$ values, and thus the signature of basalts is directly inherited from peridotites. Both planets had magma oceans after the Giant impact 4.44 Ga and thus likely had initially similar upper mantle, and basaltic upper crust with respect to major element oxygen. Triple oxygen isotopic effort to find differences between the earth and the moon resulted in almost complete overlap (Cano et al. (2020)).

The $\delta^{18}\text{O}$ composition of typical 4.3-3.8 Ga lunar basalts and zircons can be used to estimate $\delta^{18}\text{O}$ value of the lunar (and coeval terrestrial) mantle by a simple isotopic mass balance approach involving computing CIPW mineral norm of basalts, taking weighed average of $\Delta^{18}\text{O}_{\text{CIPW mineral} - \text{zircon}}$ fractionations using published fractionation factors (Chiba et al. 1989; Valley et al. 2003 (ref 23; Eiler, 2001) at assumed temperatures (e.g. Eiler, 2001 (ref 2), see Fig. A1). Such computation results in $\Delta^{18}\text{O}_{\text{lunar basalt-peridotite}}$ of 0.02 to 0.12‰ at 1400-1450°C by targeting common lunar basaltic magma types with lower values being characteristic for high-Ti and higher for low-Ti basalts. As $\Delta^{18}\text{O}_{\text{lunar zircon-basalt}}$ is -0.08-0.09‰ (Valley et al. 2014, ref 24), the $\Delta^{18}\text{O}_{\text{lunar zircon-peridotite}}$ is 0.01-0.02‰, indicating essentially that $\delta^{18}\text{O}_{\text{lunar zircon}}$ is identical to $\delta^{18}\text{O}_{\text{peridotite}}$. Therefore, lunar zircon plotted on Fig. 1 in the main text serves as a very good proxy for lunar (and terrestrial) mantle at 4.3-3.8 Ga. Our estimate of the BSE based on regression of a straight line that yields 5.57‰, nearly perfectly agrees with average lunar zircon of 5.61‰ measured by Valley et al. (2014, ref 24).

Fig. A2. Data from this study plotted vs different age assignment (Age 2, Table A1) based on the age of the lithosphere and the youngest possible age assignment assuming latest rejuvenation. Compare to Fig 1 in main text. Different age assignment does not change the decreasing trend.

Fig. A3 Data from Matthey et al. (1994) plotted vs continental lithospheric age. Notice decreasing $\delta^{18}\text{O}$ trends similar to what we found in this paper. Notice that both $\delta^{18}\text{O}_{\text{Ol}}$ and $\delta^{18}\text{O}_{\text{Bulk}}$ are decreasing with age while $\delta^{18}\text{O}_{\text{Opx}}$ stay constant or even decrease, while $\Delta^{18}\text{O}_{\text{Opx-Ol}}$ stay nearly constant contrary to the expectation of Fig. A1 of increasing $\delta^{18}\text{O}_{\text{Opx}}$. Panel a) assumes age of the mantle nodule as the age of the lithosphere they are collected from using Sm-Nd model ages (e.g. Bennett and DePaolo, 1987), while panel b) assumes that some of them represent the age of the crust modification by subduction/orogenesis, for example S Dish Hills (California): is plotted at 0.05 Ma (Luffi et al. (2009)).

Fig A4 Estimation of the bulk $\delta^{18}\text{O}$ value based on measured $\delta^{18}\text{O}$ values of Olivine and Opx ($\pm$ Cpx, where available) at indicated temperatures in Table A1 vs bulk $\delta^{18}\text{O}$ value computed only on $\delta^{18}\text{O}$ values of Olivine (computed). Both calculated and measured values use measured modal proportions of minerals in each peridotite. Note 1:1 agreement.

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Figures

Figure 1

Figure 2. Triple oxygen isotope analyses of Archean and post-Archean olivines and orthopyroxenes, showing overlap with San Carlos olivine. Expected trends to higher- $\Delta^{17}\text{O}$ and lower $\delta^{18}\text{O}$ subduction fluids, derived from low- $\delta^{18}\text{O}$ interiors of subducting slabs are shown (e.g. [2]). The $\delta^{18}\text{O}$ in orthopyroxenes are higher than in equilibrium olivine by 0.5-0.7‰ and nearly identical in $\Delta^{17}\text{O}$ at mantle temperatures (Fig. A1, ref 43).

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