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Constraints from uranium and molybdenum isotope ratios on the origin of enriched mid-ocean ridge basalts

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Abstract

Most mid-ocean ridge basalts (MORB) are depleted in highly incompatible elements relative to the primitive mantle and are termed normal (N)-MORB. Some MORB, erupted at ridge segments distal from mantle hot-spots, are enriched in incompatible elements. The origin of these enriched (E)-MORB is debated, although many studies have proposed that recycled oceanic crust shapes their compositions. Uranium (U) and molybdenum (Mo) isotope ratios have been argued to trace the contribution of recycled oceanic crust in the source of N-MORB, which has high $\delta^{238}U$ and low $\delta^{98/95}Mo$ relative to the bulk silicate Earth (BSE). Here, we provide U and Mo isotopic data on E-MORB samples from the northern mid-Atlantic ridge (13° & 45° N). We analysed hand-picked, leached MORB glass, yielding $^{234}U/^{238}U$ near secular equilibrium, therefore reflecting samples unperturbed by surface processes. Samples have uniform $\delta^{238}U$ and $\delta^{98/95}Mo$, with means of -0.307 ± 0.032 ‰, 2sd, and -0.14 ± 0.04 ‰, 2sd, respectively, both within uncertainty of BSE, and distinct from N-MORB. These data, as well as unremarkable Ce/Pb and radiogenic Pb isotopic compositions in E-MORB globally, are incompatible with their sources containing recycled oceanic crust or continental derived sediments. Instead, our data fit with a model of low degree partial melting of the uppermost mantle that metasomatises the sub-oceanic lithosphere. Given BSE-like U isotopic compositions of E-MORB, that are isotopically unfractionated during low degree partial melting, we suggest that the initial melting event must have occurred prior to the recycling of isotopically distinct in U oceanic crust into the upper mantle (i.e., prior to ca. 600 Ma, the estimated time of deep ocean oxygenation). Metasomatised portions of oceanic lithospheric mantle preserve these ≥600Ma U isotopic compositions, which are subducted and stirred back into the convecting upper mantle, ultimately to be sampled at ridges as E-MORB. Molybdenum

isotopic compositions of E-MORB are in line with such a model but also reflect isotopic fractionation to higher $\delta^{98/95}$Mo during low degree partial melting of ≥600 Ma upper mantle, that counter acts the lowering of $\delta^{98/95}$Mo in the upper mantle by an on-going process of plate recycling.

Keywords: U isotopes; Mo isotopes; Enriched MORB; Crustal recycling; low degree partial melting

1. Introduction

Mid-ocean ridge basalts (MORB), magmatic samples of the upper mantle, are chemically heterogenous and commonly split into two groups according to their 'incompatible' element compositions (Fig. 1) (e.g., Schilling, 1975; Gale et al., 2013). Along the majority of mid-ocean ridge (MOR) segments, basalts erupted have 'depleted' signatures, with ratios of more to less incompatible elements lower than estimates of the primitive mantle (e.g., La/Sm normalized to primitive mantle, $(La/Sm)_N < 1$) (Fig. 1) (McDonough and Sun, 1995). Rarer, enriched (E)-MORB have incompatible element abundances markedly higher than N-MORB and are associated with elevated ratios of more to less incompatible elements (e.g., $(La/Sm)_N \geq 1$) and distinctive isotopic signatures (e.g., radiogenic $^{87}Sr/^{86}Sr$). While their presence is well documented, the exact definition of E-MORB varies in different studies (Fig. 1).

Some E-MORB locations are from topographically elevated MOR sections and are linked to enrichments from hot-spots, upwellings from greater depth (Schilling, 1975; Schilling et al., 1985). However, for E-MORB that occur at MOR segments far from the influence of hot-spots, the origin of the chemical enrichment is debated. It has been proposed that recycling of oceanic crust and/or continental sediments into the upper mantle causes enrichment (e.g., Allègre and Turcotte, 1986; Prinzhofer et al., 1989; Hémond et al., 2006; Waters et al., 2011; Ulrich et al., 2012; Yang et al., 2020), similar to models of source enrichment in ocean island basalts (OIB). Other works argue for low degree partial melting and two stage melting models, either with or without recycled crustal material. Donnelly et al. (2004) argue for low degree partial melting of subducting crust that enriches the convecting mantle wedge at subduction zones. This enriched mantle is stirred into the

93 wider upper mantle and can be sampled at ridges giving rise to E-MORB. Nielsen et al., (2018)
94 propose a similar model but also argue for the addition of subducted sediment to the overlying
95 mantle that induces low degree partial melting to produce the E-MORB reservoir that is ultimately
96 sampled under ridges. Scenarios that do not invoke recycled crustal material in the E-MORB
97 source argue for low degree partial melts of the uppermost mantle that metasomatise oceanic
98 mantle lithosphere, enriching portions of mantle that are subsequently subducted back into the
99 upper mantle and sampled at ridges (e.g., Green, 1971; Kostopoulos and Murton, 1992; Halliday
100 et al., 1995; Niu et al., 2002; Chen et al., 2022; Guo et al., 2023).
101
102 Measurements of novel stable isotope ratios can be used to investigate upper mantle chemical
103 heterogeneity and enrichment. Uranium (U) and molybdenum (Mo) are useful for tracing
104 processes of crustal recycling due to large low temperature isotopic fractionations that occur during
105 seafloor alteration of the oceanic crust, sediment deposition (Andersen et al., 2015; Freymuth et
106 al., 2015; Ahmad et al., 2021), and during the dehydration of subducting slabs in subduction zones
107 (Andersen et al., 2015; Freymuth et al., 2015, 2019; König et al., 2016; Gaschnig et al., 2017;
108 Villalobos-Orchard et al., 2020; Ahmad et al., 2021). Here we express Mo isotope ratios as
109 $\delta^{98/95}Mo_{NIST\ SRM3134}$ (the relative difference in $^{98}Mo/^{95}Mo$ between samples and standard reference
110 material NIST SRM3134, hereafter written $\delta^{98/95}Mo$ in the text), and U isotope ratios as
111 $\delta^{238}U_{CRM145}$ (the relative difference in $^{238}U/^{235}U$ between samples and certified reference material
112 CRM-145, hereafter written $\delta^{238}U$ in the text).
113
114 Slab dehydration during subduction releases oxidising fluids with high $\delta^{98/95}Mo$ and low $\delta^{238}U$ into
115 the overlying mantle wedge, as inferred from the compositions of volcanic arc lavas (Andersen et

al., 2015; Freymuth et al., 2015, 2019; König et al., 2016; Gaschnig et al., 2017; Villalobos-Orchard et al., 2020) (Fig. 2). Exhumed eclogites and metasediments with low $\delta^{98/95}$Mo also reflect this process (Fig. 2a) (Chen et al., 2019; Ahmad et al., 2021). Additionally, in some cases mafic oceanic crust has acquired a low $\delta^{98/95}$Mo through seafloor alteration before subduction zone processing (Ahmad et al., 2021). The U isotopic system further reflects the importance of seawater alteration of the oceanic crust (Fig. 2b). Seawater alteration of oceanic crust strongly increases its U concentration (e.g., Staudigel et al., 1995) with the added U, on average, being isotopically heavy (Andersen et al., 2015, 2024). Subducting slab dehydration and seawater alteration therefore result in residual slabs with compositions that are isotopically light in Mo and isotopically heavy in U. The recycling of Mo and U from this crustal material into the upper mantle is inferred from compositions of N-MORB samples that are isotopically lighter in Mo and heavier in U than the bulk silicate Earth (BSE) as defined by chondrites (Burkhardt et al., 2014; Andersen et al., 2015; Hin et al., 2022) (Fig. 2). This can also be seen in higher Ce/Mo ratios and lower Th/U ratios of N-MORB than the BSE. Cerium and Th are of similar incompatibility to Mo and U respectively during mantle melting, but with different fluid mobility; Mo and U are aqueous fluid mobile, Ce and Th are not.

Following the onset of the first major rise in atmospheric oxygen (~ 2.3 Ga), there would have been a supply of continental derived U to the oceans due to oxidative weathering. The recycling of oceanic crust with excess U relative to immobile Th has been used to explain the lower measured $^{232}Th/^{238}U$ in MORB relative to the time integrated $^{232}Th/^{238}U$ ratio calculated from Pb isotopic compositions (e.g., Zartman and Haines, 1988; McCulloch, 1993; Collerson and Kamber, 1999; Elliott et al., 1999). Isotopically perturbed U, however, may only have been recycled into the

mantle since the Neoproterozoic oxygenation event, ~ 600 Ma (e.g., Lyons et al., 2014), given that imparting high $\delta^{238}U$ to altered oceanic crust (AOC) requires oxygenated deep oceans (Andersen et al., 2015, 2024). Isotopically perturbed Mo, in contrast, has potentially been recycled into the N-MORB source since the onset of modern day like plate tectonics and mass balance models suggest that at least ~ 1 Gyr of crustal recycling is needed to cause the observed shift in N-MORB $\delta^{98/95}Mo$ from BSE compositions (Hin et al., 2022).

Molybdenum and U isotopes offer a way to investigate if recycled oceanic crust is mixed into the E-MORB source and provide constraints on the timescales it takes to 'pollute' the upper mantle with surface derived material. Recent studies have shown some E-MORB have chondritic or slightly higher values of $\delta^{98/95}Mo$ and are resolvable from N-MORB (Bezard et al., 2016; Chen et al., 2022). To assess if this is a ubiquitous feature, there is need for data from different geographic regions, as well as for measurements of both $\delta^{98/95}Mo$ and $\delta^{238}U$ on the same samples. Herein we combine Mo with U isotopic measurements on a set of E-MORB samples from the northern Mid-Atlantic ridge (MAR) as a diagnostic test for recycled crustal components in the E-MORB source.

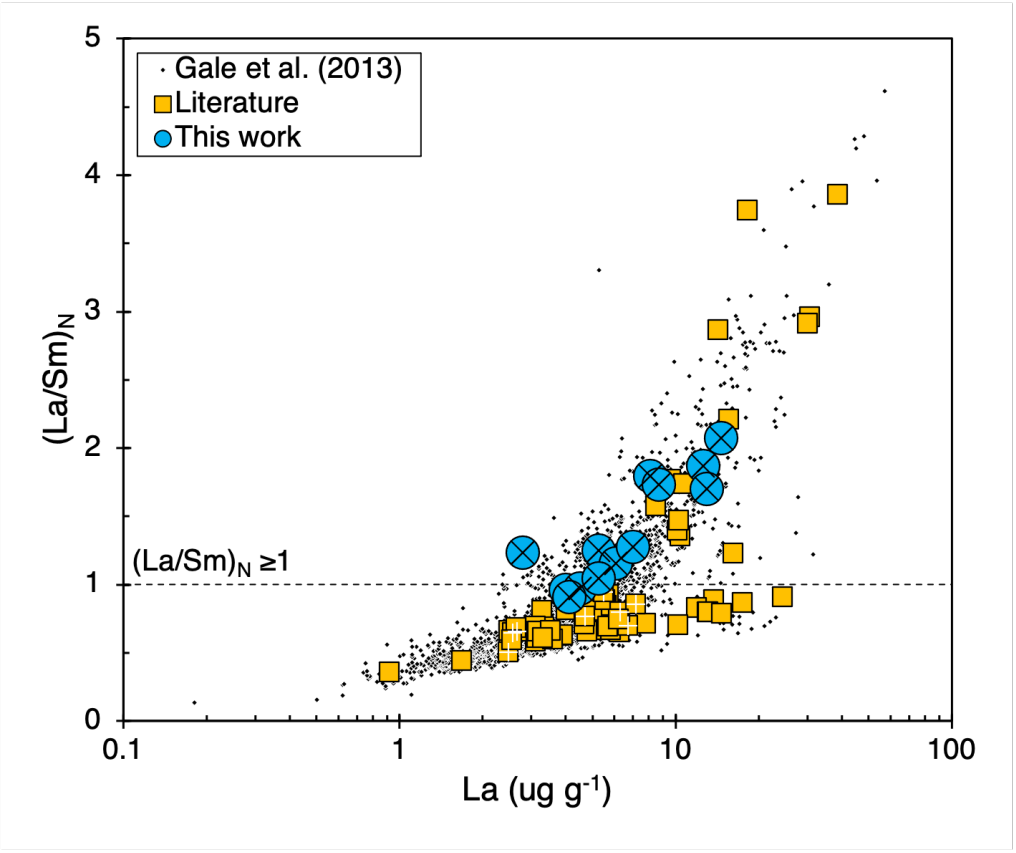
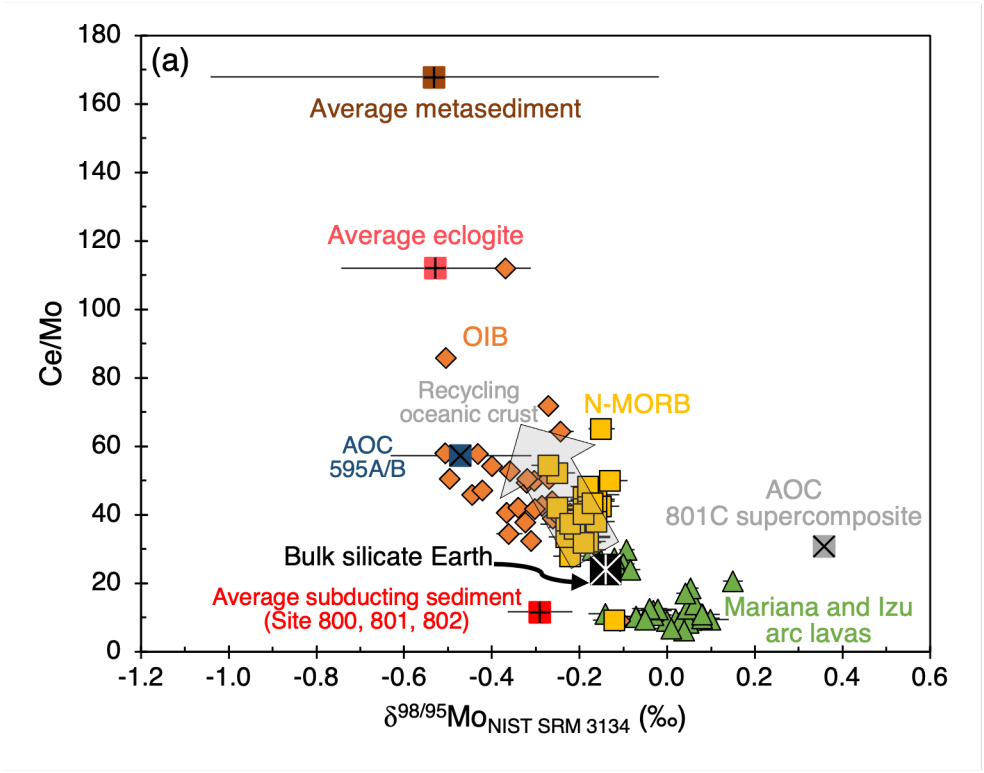


Fig. 1. Global MORB variations in chemical enrichment based on $(La/Sm)_N$. Mid-ocean ridge basalts database from Gale et al. (2013) shown as small black diamonds. Literature MORB data with Mo and/or U isotopic data are shown as yellow squares (Andersen et al., 2015; Bezard et al., 2016; Chen et al., 2022; Hin et al., 2022). Mid-ocean ridge basalt samples analysed in this study are shown as larger blue circles. Samples in this work and literature data with both Mo and U isotope data are shown with black crosses (E-MORB) or white plus signs (N-MORB).


(a)
Average metasediment
Average eclogite
OIB
Recycling
oceanic crust
N-MORB
AOC
595A/B
AOC
801C supercomposite
Bulk silicate Earth
Average subducting sediment
(Site 800, 801, 802)
Mariana and Izu
arc lavas
Ce/Mo
δ98/95MoNIST SRM 3134 (‰)
180
160
140
120
100
80
60
40
20
0
-1.2
-1.0
-0.8
-0.6
-0.4
-0.2
0.0
0.2
0.4
0.6

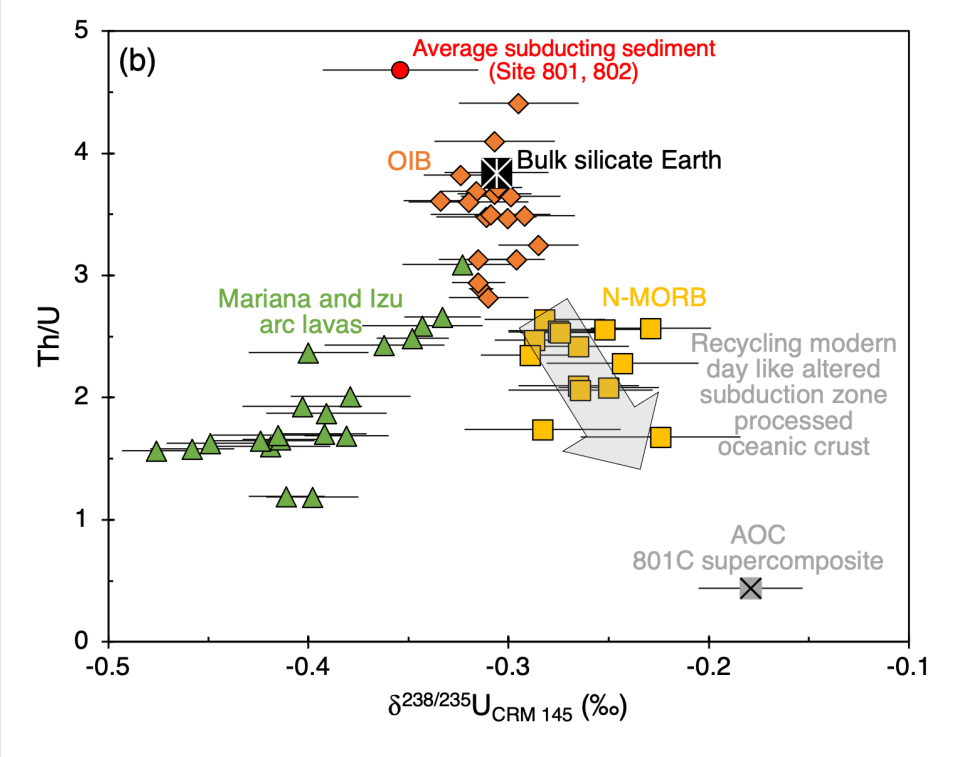

(b)
Average subducting sediment
(Site 801, 802)
OIB
Bulk silicate Earth
Mariana and Izu
arc lavas
N-MORB
Recycling modern
day like altered
subduction zone
processed
oceanic crust
AOC
801C supercomposite
Th/U
δ238/235UCRM 145 (‰)
5
4
3
2
1
0
-0.5
-0.4
-0.3
-0.2
-0.1

Fig. 2. (a) $\delta^{98/95}$Mo versus Ce/Mo and (b) δ^{238}U versus Th/U for mantle derived basalts, AOC, subduction processed N-MORB-like eclogite and subducting sediment. Ocean island basalts (orange diamonds) are from Willbold and Elliott (2023) and Andersen et al. (2015). Volcanic arc lavas from the Mariana and Izu arc (green triangles) are from Freymuth et al. (2015, 2019), Andersen et al. (2015), and Vilalobos-Orchard et al. (2020). Bulk silicate Earth compositions (black starred square) are from Hin et al. (2022) and Andersen et al. (2015). Average Western Pacific subducting sediment composition (red square with a plus sign) is from Ocean Drilling Programme sites 800, 801, and 802 from Andersen et al. (2015) and Freymuth et al. (2015). We use the global average subducting sediment Th/U ratio from Plank (2014) GLOSS-II. Average compositions of AOC from the 801C supercomposite (grey crossed square) are from Andersen et al. (2015) and Freymuth et al. (2015). Average AOC from 595A/B (blue crossed square) is the average composition, ± 1sd, weighted by Mo concentration from Ahmad et al. (2021). Average composition, ± 1sd, weighted by Mo concentration of a set of exhumed eclogites (pink square with a plus sign) are from Chen et al. (2019) and Ahmad et al. (2021). Average composition, ± 1sd, weighted by Mo concentration of metasediments (brown square with a plus sign) is from Ahmad et al. (2021). Mid-ocean ridge basalt data (yellow squares) are from Andersen et al. (2015), Bezard et al. (2016), Chen et al. (2022), and Hin et al. (2022). For MORB Mo data we follow the filtering of Hin et al. (2022), where two anomalous samples from Bezard et al. (2016) are excluded, as they do so themselves. We also exclude all data from Liang et al. (2017), whose data cannot be reproduced and show markedly higher $\delta^{98/95}$Mo compositions for MORB samples than Bezard et al. (2016) and Chen et al. (2022), see Hin et al. (2022) for details. Grey arrows show the effect of mixing recycled oceanic crust into the mantle.

2. Geological location and samples

We report U and Mo elemental and isotopic compositions for sixteen MAR E-MORB samples, located at ~ 13° N 44° W (five samples) and 45° N 28° W (eleven samples) that span MgO contents from ~ 10 to 7 wt. %. Our samples are chemically enriched, with either $(La/Sm)_N \geq 1$ and or $K_2O/TiO_2 \geq 0.11$. Enriched-MORB in the 13° N 44° W segment are common, and basalts have up to forty times higher concentrations of highly incompatible elements than N-MORB from segments nearby (Bougault et al., 1988). Samples from 13° N 44° W were collected by dredging in the RSS James Cook JC007 cruise in March – April 2007 (Wilson et al., 2013) (Table S1). They represent a group of basalts erupted before the formation of oceanic core complexes in the area, and now sit off-axis, having been erupted at ~ 0.5 – 1 Ma. Samples from ~ 45° N 28° W are also from a region where E-MORB commonly occurs (Bougault et al., 1988) and samples were collected *in situ* during RSS James Cook cruise JC024 May – June 2008 and are all <3 Ma (Table S1) (Searle et al., 2010). Major and trace element data for both sample sets were collected following the methods in Wilson et al. (2013).

The Azores hot-spot at ~ 38° N 28° W, which forms the Azores Island chain sitting to the east of the MAR, is the closest mantle hot-spot to both sample sites. The Azores hot-spot interacts with the MAR, causing nearby ridge segments to become broader and shallower. Material from the Azores hot-spot flows southwards along the ridge, with elevated La/Sm ratios between 35° to 40° N. More N-MORB like compositions occur below 30° N and above 40° N, with no detectable effects of the hot-spot further south than 26° N (e.g., Maia et al., 2007). Our sample sites sit outside

the zone of influence of the Azores hot-spot, and geochemical enrichment is not linked to hot-spot-ridge interaction (Bougault et al., 1988).

3. Methods

Fresh MORB glass was crushed and processed to ~ 600 μm size chips, using an agate pestle and mortar. To avoid samples potentially affected by seawater alteration (e.g., Fe-Mn oxide coatings), samples were hand-picked under a binocular microscope to ensure samples were optically clear and devoid of potential alteration. While this process has long been employed, notably in U-series disequilibrium studies (e.g., Reinitz and Turekian, 1989), it is laborious, and considering the quantities needed for isotopic analysis (e.g., >1 g), it is a rate limiting step. Hand picking MORB glass is also subjective, and it is unclear what defines an acceptable limit of quality. A reliable check of sample alteration in young samples is given by measurements of $^{234}U/^{238}U$ activity ratios. If unaffected by recent seawater alteration, the ^{238}U decay chain will be in secular equilibrium, and so the activity ratio of $^{234}U/^{238}U$, typically expressed as ($^{234}U/^{238}U$), will be unity. Seawater has ($^{234}U/^{238}U$) ~ 1.14 (e.g., Kipp et al., 2022) and elevated ($^{234}U/^{238}U$) in samples may indicate the addition of seawater U and other elements onto Fe-Mn oxide coatings (Siebert et al., 2003; Hin et al., 2022). We explored varyingly stringent picking strategies on samples. Up to three different splits of glass of varying quality (A, B, and C in decreasing order of quality) were prepared (classifications are detailed in Supplementary Material: Section 1) (Fig. S1). In some cases, different splits were combined to ensure there was enough sample to measure.

Samples also underwent a reductive leaching step (Supplementary Material: Section 1) prior to dissolution to remove secondary coatings. Samples were leached with a mixture of 0.05 M hydroxylamine hydrochloride, 15 % acetic acid and 0.03 M Na-EDTA buffered to pH 4 with NaOH (Gutjahr et al., 2007). Andersen et al. (2015) and Hin et al (2022) note that leaching can result in

some glass dissolution, and minor U and Mo loss (Fig. S2). Ratios of the concentrations of elements that absorb to Fe-Mn coatings such as U and Mo to those little affected, such as Th, that would only be removed during glass dissolution, were monitored to examine the effects of leaching. Samples of JC24-82-21 were prepared and analysed before other samples to calibrate methods. This sample was leached three times and results indicated that one to two leaching steps were sufficient to remove any apparent chemical signature of Fe-Mn coatings (Fig. S3). For other samples we opted for two leaching steps, although note that one step is likely sufficient.

Uranium isotopic measurement methods followed Andersen et al. (2015), and Mo isotopic measurement methods followed Willbold et al. (2016) and Hin et al. (2022), as detailed fully in Supplementary Material: Section 2. Uranium and Mo isotope analyses were conducted in the Bristol Isotope Group labs, University of Bristol. Approximately 0.5 to 1 g of MORB glass was dissolved and after achieving full dissolution, a ~ 1 % fraction of samples was measured on an Element2 ICP-MS for Th, U, and Mo concentration (Andersen et al., 2014). Measured reference materials are in good agreement with literature values (Supplementary Material: Section 2) (Table S2).

Samples were spiked with the IRMM3636 ^{236}U – ^{233}U 50:50 double spike (Richter et al., 2008) aiming for a $^{236}U/^{235}U$ ratio of 5. Samples were also spiked with an in-house ^{97}Mo – ^{100}Mo double spike, with a $^{97}Mo/^{95}Mo$ ratio of 47.58 and $^{100}Mo/^{95}Mo$ ratio of 58.32, aiming for a natural Mo-double spike Mo proportion of 0.5.

Purification and U separation used a two-column method, with TRU resin to separate most matrix elements, including all Mo, followed by UTEVA resin to separate Th from U. An aliquot containing Mo from the first separation column was collected for later processing. Final U aliquots were dissolved in 0.2 M HCl (aiming for U concentration of 100 – 300 ng g^{-1}) for isotopic analysis. Procedural blanks were <30 pg U, negligible compared to amount of U consumed per measurement, 30 – 80 ng.

Uranium isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS (serial no. 1002) in low mass resolution (M/ΔM ~ 2000, 5 to 95 % peak height definition). Samples were introduced into the plasma using a ~ 40 ul min^{-1} micro-concentric PFA nebuliser connected to a Cetac Aridus (1st generation) desolvating system. Masses 232 (^{232}Th), 233 (^{233}U), 234 (^{234}U), 235 (^{235}U), 236 (^{236}U), and 238 (^{238}U) were measured simultaneously. Each sample was preceded and followed by a measurement of the double-spiked CRM-145 standard. Individual measurements consisted of 80 cycles, with 4.194 s integration time.

Uranium isotope ratios for $^{238}U/^{235}U$ and $^{234}U/^{238}U$ were calculated using the exponential mass fractionation law and double spike $^{233}U/^{236}U$ ratio (Richter et al., 2008). Data reported are normalised to the average of the bracketing CRM-145 standard, with $^{234}U/^{238}U$ ratios reported in delta notion relative to secular equilibrium where secular equilibrium is 0 and CRM-145 has a $\delta^{234}U$ value of −38.6 ‰ (Cheng et al., 2013).

External reproducibility of all samples has been determined from the long-term external reproducibility of BHVO-2 measured at various intensities (Supplementary Material: Section 2).

343 This results in an estimated external reproducibility of $\delta^{238}U$ and $\delta^{234}U$ from ± 0.09 to 0.03 ‰,
344 2sd, and ± 4 to 0.9 ‰, 2sd, for ^{238}U intensities from 200 – 1000 pA ranges respectively (Fig. S4).
345 Uranium isotopic measurements of international reference materials (BHVO-2, BCR-2, BIR, W-
346 2A, and CZ-1) agree with literature values (Table S3).
347
348 Collected Mo fractions from the TRU resin U chemistry were dried and dissolved for Mo
349 chemistry using Eichrom AG 1-X8 anionic resin. Final Mo collections were dried and re-dissolved
350 in 0.4 M HNO_3 – 0.4 M HF for a Mo concertation of 200 ng g^{-1} for isotopic analysis. Procedural
351 blanks were <400 pg Mo, negligible compared to the amount of Mo consumed per measurement,
352 ~ 30 ng.
353
354 Molybdenum isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS
355 (serial no. 1020) in low mass resolution (M/ΔM ~ 1600, 5 to 95 % peak height definition). Samples
356 were introduced to the plasma using a ~ 40 ul min^{-1} micro-concentric PFA nebuliser connected to
357 a Cetac Aridus (1st generation) desolvating system. Masses 91 (^{91}Zr), 92 (^{92}Mo), 95 (^{95}Mo), 96
358 (^{96}Mo), 97 (^{97}Mo), 98 (^{98}Mo), 99 (^{99}Ru), 100 (^{100}Mo), and 101 (^{101}Ru) were measured
359 simultaneously. Each sample was preceded and followed by a measurement of the double-spiked
360 standard NIST SRM3134. Individual measurements consisted of 30 cycles, with 4.194 s
361 integration time.
362
363 Measurements were internally normalised with a double spike inversion using the isotopes ^{95}Mo,
364 ^{97}Mo, ^{98}Mo, and ^{100}Mo. Samples were then externally normalised to the spiked bracketing standard
365 NIST SRM3134 to calculate $\delta^{98/95}Mo$. Data were corrected for ^{98}Ru and ^{100}Ru interreferences using

both ^{99}Ru and ^{101}Ru respectively as monitors of Ru intensity; both corrections yielded the same Mo isotope ratios within uncertainty of one another and uncorrected data. We take a homoscedastic approach to determine our external reproducibility, pooled 2sd, on any single stable Mo isotopic measurement (i.e., one standard-sample-standard measurement) (Table S4). Using this approach, we define an external reproducibility of $\delta^{98/95}Mo \pm 0.05$ ‰, 2sd, for a single measurement in a given run. This pooled 2sd is then used to calculate the standard error for a given sample given the number, n, of repeat measurements, typically 4 to 6 for unknown samples. This is identical to the 2sd, ± 0.05 ‰, of 35 repeats of W-2A measured over 4 digestions across 4 measuring sessions, and is similar to that reported in Chen et al. (2022) and Hin et al. (2022). Molybdenum isotopic measurements of international reference materials (BHVO-2, BCR-2, and W-2A) agree with literature data (Table S5).

4. Results

Our MAR samples have chemical enrichments from $(La/Sm)_N$ 0.91 to 2.07 (K_2O/TiO_2 0.11 to 0.37) (Fig. 1, table S6). Uranium concentrations range from 101 to 443 ng g^{-1}, all enriched relative to average N-MORB (83 ng g^{-1}, Gale et al., 2013) (Table S6). Molybdenum concentrations range from 189 to 967 ng g^{-1}, which is above and below average N-MORB (360 ng g^{-1}, Gale et al., 2013) (Table S6). There are positive correlations between U and Mo concentrations with $(La/Sm)_N$ (Fig. 3a, b).

Values of $\delta^{238}U$ show little variation between −0.331 ± 0.019 ‰, 2se, and −0.263 ± 0.028 ‰, 2se, with a concentration weighted average of −0.307 ± 0.032 ‰, 2sd, (Fig. 3c, table S6). The variability is similar to our long-term external reproducibility of samples measured at similar conditions ~ ± 0.03 ‰ and reflects a near uniform composition of our sample set. The $\delta^{238}U$ compositions of the different qualities of glass picked and leached are all, bar one sample, within analytical uncertainty (Fig. S5a). Also, samples bar JC-24-89-13, are within uncertainty of secular equilibrium (Fig. S5b). Sample JC-24-89-13, which is only +2.5 ‰ in $\delta^{234}U$ (Fig. S5b), also has a similar $\delta^{238}U$ to other samples (Fig. S5a). Our E-MORB $\delta^{238}U$ average is lower than global N-MORB, which has a concentration weighted average of −0.259 ± 0.041 ‰, 2sd, (Andersen et al., 2015) (Fig. 3c). We also report $\delta^{238}U$ data for four N-MORB samples from the Indian ocean basin (04/13C, 05/15G, 08/26F, 12/37F) additional to the N-MORB samples in Andersen et al. (2015), but reported in Hin et al. (2022) for $^{234}U/^{238}U$ data. These data are provided in the supplementary material and were collected following methods in Andersen et al. (2015). Our E-MORB $\delta^{238}U$ average is indistinguishable from BSE, $\delta^{238}U$ −0.306 ± 0.026 ‰, 2se, (Andersen et al., 2015).

Molybdenum isotopic compositions show little variation with $\delta^{98/95}$Mo ranging between −0.11 ± 0.01 ‰, 2se, and −0.19 ± 0.02 ‰, 2se, and are within analytical uncertainty of a concentration weighted average of −0.14 ± 0.04 ‰, 2sd, (Fig. 3d, table S6). The variability, ± 0.04 ‰, 2sd, is smaller than our long-term external reproducibility and reflects the near uniform composition of our sample set. Our E-MORB concentration weighted average $\delta^{98/95}$Mo is indistinguishable from the value reported in Hin et al., (2022) for global E-MORB, −0.12 ± 0.03 ‰, 95 % c.i. The $\delta^{98/95}$Mo compositions of the different qualities of glass picked and leached are all within analytical uncertainty (Fig. S5c). Our E-MORB $\delta^{98/95}$Mo average is higher than global N-MORB, $\delta^{98/95}$Mo −0.19 ± 0.01 ‰, 95 % c.i. and indistinguishable from BSE, $\delta^{98/95}$Mo −0.14 ± 0.02 ‰, 95 % c.i. (Fig. 3d) (Hin et al., 2022).

There are no correlations of δ^{238}U and $\delta^{98/95}$Mo in our E-MORB samples with tracers of chemical enrichment (Fig. 3c, d). There are no correlations between U and Mo isotopic compositions and indicators of magmatic differentiation (Fig. S6). Samples from 13° N and 45° N show no resolvable differences and we find no reason to treat each site differently. In summary, the concentration weighted averages of δ^{238}U and $\delta^{98/95}$Mo for our E-MORB samples is distinct from global N-MORB, but indistinguishable from BSE (Fig. 3c, d) (Andersen et al., 2015; Hin et al., 2022).

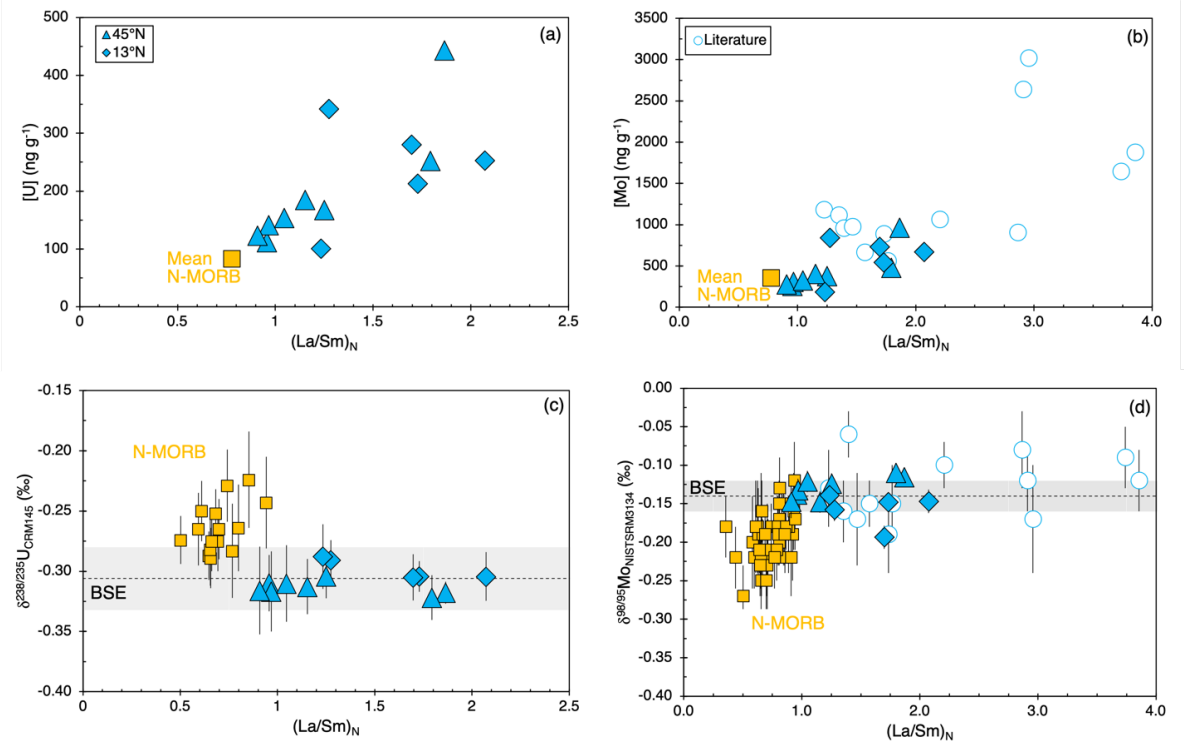


Fig. 3. (a) U and (b) Mo concentrations and (c) $\delta^{238}U$ and (d) $\delta^{98/95}Mo$ versus $(La/Sm)_N$ of MORB samples. Enriched-MORB from this study are shown as filled blue symbols and grouped into samples from 45°N (triangles) and 13°N (diamonds) and shown with ± 2se uncertainties. Average N-MORB (yellow squares) concentrations are from Gale et al. (2013). Isotopic data for BSE and N-MORB are from the same sources as in figure 2, and literature E-MORB (hollow circles) are from Bezard et al. (2016), Chen et al. (2022) and Hin et al. (2022). Grey shaded regions represents isotopic compositions of BSE (± 2se).

5. Discussion

5.1 Potential alteration of U and Mo isotopes

The reductive leaching process removed some U but little Mo, reflecting the presence of minimal secondary mineral hosted Mo (Supplementary Material: Section 1) (Fig. S2). The different qualities of picked glass for each sample measured showed similar patterns in leaching and are all largely within uncertainty in $\delta^{238}U$, $\delta^{98/95}Mo$, and $\delta^{234}U$ (all near secular equilibrium) (Fig. S3, S5). We therefore average all different splits measured for samples into overall $\delta^{238}U$, $\delta^{98/95}Mo$, and $\delta^{234}U$ compositions for each sample (Supplementary material: section 1). We further test that our hand-picked MORB glass samples reflect primary magmatic compositions by examining mixing relationships between our average E-MORB compositions and predicted compositions of Fe-Mn crusts. Iron-Mn crusts acquire U from seawater, with elevated $\delta^{234}U$, and have low $\delta^{238}U$ and $\delta^{98/95}Mo$, ~ −0.69 ‰ and −0.92 ‰ respectively (Siebert et al., 2003; Goto et al., 2014). In a binary mixing calculation between our average E-MORB composition and Fe-Mn crusts in $\delta^{238}U$-$\delta^{234}U$ and $\delta^{98/95}Mo$-$\delta^{234}U$ space, our samples do not form arrays towards the composition of Fe-Mn crusts (Fig. 4). The minor variability in $\delta^{238}U$ and $\delta^{98/95}Mo$ appears unrelated to $\delta^{234}U$, and the samples with minor deviations in $\delta^{234}U$ from secular equilibrium do not show compositions systematically perturbed towards Fe-Mn crusts in either U or Mo isotopic compositions (Fig. 4). We therefore infer that the $\delta^{238}U$ and $\delta^{98/95}Mo$ of the samples represent primary values.

481 5.2 Fractional crystallisation

482

483 Our data span a narrow range of MgO content (10.2 to 6.9 wt. %) and show no correlation in
484 $\delta^{98/95}$Mo and δ^{238}U with magmatic differentiation (Fig. S6). This is in accordance with other studies
485 that show no resolvable correlation of $\delta^{98/95}$Mo in MORB with MgO over a wider range of
486 compositions, 1.8 to 9.5 wt.% (Bezard et al., 2016; Chen et al., 2022). Our samples also reflect the
487 similar incompatibilities of U & Th and Mo & Ce during magmatic differentiation (Fig. S6), with
488 near constant ratios of ~ 3 and ~ 35 respectively, in keeping with wider data for seafloor basalts
489 (e.g., Gale et al., 2013).

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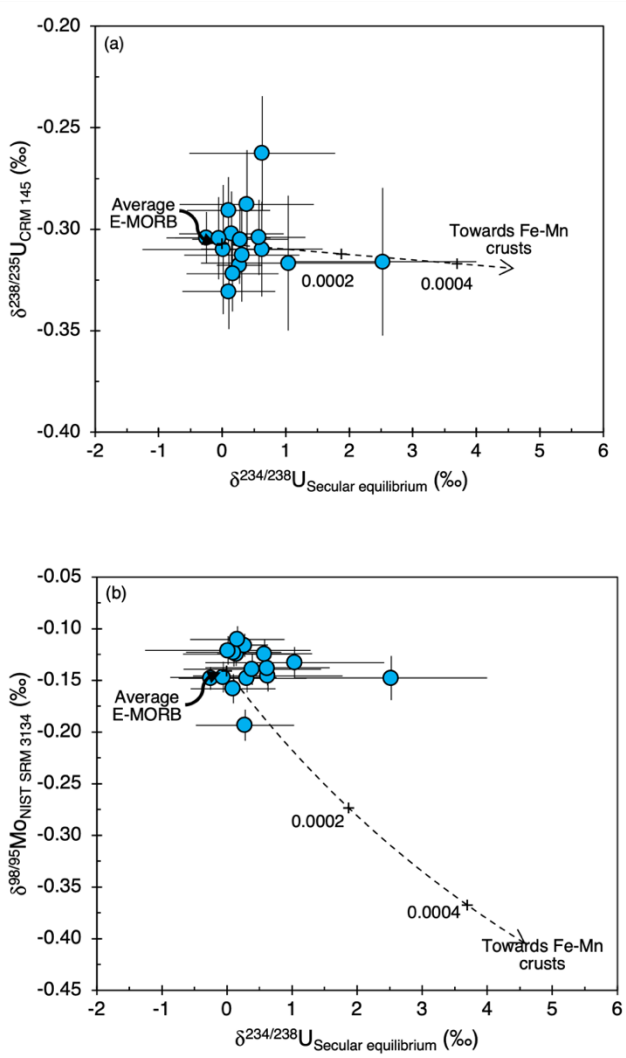


Fig. 4. Modelled mixing curves of $\delta^{234}U$ versus (a) $\delta^{238}U$ and (b) $\delta^{98/95}Mo$, showing the trajectory of Fe-Mn crust addition to our average E-MORB composition (dashed line). Values along the mixing curves show the mass fraction of Fe-Mn crust in the mixture. Compositions used in the mixing calculation are, E-MORB, $\delta^{238}U = -0.307$ ‰, $\delta^{234}U = 0$ ‰, [U] = 202 ng g^{-1}, $\delta^{98/95}Mo = -0.14$ ‰, and [Mo] = 465 ng g^{-1}. Fe-Mn crust, $\delta^{238}U = -0.69$ ‰, $\delta^{234}U = 146.3$ ‰, [U] = 13100 ng g^{-1}, $\delta^{98/95}Mo = -0.92$ ‰, and [Mo] = 477000 ng g^{-1} (Henderson and Burton, 1999; Siebert et al., 2003; Goto et al., 2014).

5.3 Recycled crustal material in the E-MORB source

Recent work from Yang et al. (2020) suggests, based on compatible element abundances, that E-MORB geochemistry is explained by the mixture of low degree partial melts of garnet-clinopyroxene pyroxenite (i.e., recycled oceanic crust), with depleted MORB like melts. Melting of this recycled material and mixing with depleted MORB melts generates distinct compositions of element ratios, such as lower Ge/Si, in E-MORB relative to depleted MORB (Yang et al., 2020). The dehydration of subducting slabs during subduction zone processing strips oceanic crust of fluid mobile elements, resulting in high fluid mobile/fluid immobile element ratios in arc lavas. Complementary compositions should then be seen in E-MORB if they contain recycled crustal components, but E-MORB are enriched in both fluid mobile and immobile elements, and have no depletions in elements such as Nb, Tb, and Ti as seen in arc lavas (Niu et al., 2002, their figure 9). To reproduce some of the such E-MORB characteristics in their mixing models, Yang et al. (2020) suggest the recycling of upper continental crust material, that is enriched in elements such as Rb, Ba, and Pb, along with recycled oceanic crust to explain high Rb/Sr, Ba/La, and low Zr/Pb ratios of E-MORB.

Subducted sediments, a proxy for upper continental crust material, are distinctly enriched in Pb and have low Ce/Pb ratios and high $^{207}Pb/^{204}Pb$ ratios relative to $^{206}Pb/^{204}Pb$ (e.g., White and Dupré, 1986; Plank, 2014). Therefore, we explore the mixing relationships defined by mixing subduction zone processed subducted sediments and recycled oceanic crust (Stracke et al., 2003) into the depleted MORB mantle (DMM) for Pb isotopic compositions and Ce/Pb ratios. Yang et al. (2020) argue for a significant amount of recycled material (in a 95-5 % mixture of recycled oceanic crust

550 and upper continental crust material) mixed into the E-MORB source (e.g., 10 to 30 %). Such
551 amounts of subducting sediment and recycled oceanic crust would decrease the Ce/Pb ratio and
552 increase the $^{207}Pb/^{204}Pb$ ratio of the upper mantle source to unobserved compositions (Fig. 5).
553 Recycling of a subducting sediment component, also has implications for the Mo and U isotopic
554 compositions, and current combined Mo-U isotopic data on subducting sediment, and Mo data on
555 metasediments are an unlikely candidate for creating E-MORB, with the meta-/sediment being too
556 isotopically light in $\delta^{98/95}Mo$ (Freymuth et al., 2015; Ahmad et al., 2021) (Fig. 6, 7a). However,
557 we note that sediment compositions can be variable (e.g., Freymuth et al., 2015; Ahmad et al.,
558 2021) and that further characterisition of combined Mo-U isotopic data on subducted sediments is
559 needed to show the potential influence of subducting sediments on the composition of the MORB
560 source. Nonetheless, our inferences from global averaged Ce/Pb and Pb radiogenic compositions
561 still preclude the mixing of subducted sediments into the E-MORB source (Fig. 5).
562
563 Other studies also suggest a role for recycled crustal material in the formation of E-MORB sources.
564 Donnelly et al. (2004) argue for low degree partial melts of subducted oceanic crust as eclogite at
565 depth that metasomatises the mantle wedge, creating necessary trace element enrichments. This
566 material is subsequently recirculated through plate motion into the upper mantle over time, ≥300
567 Myr, to allow for radiogenic isotope ratio ingrowth, before being sampled again under MOR's in
568 a larger degree melting event. Our data preclude this model, however, as the eclogitic residues of
569 oceanic crust post subduction zone processing have isotopically light $\delta^{98/95}Mo$ and high Ce/Mo,
570 which is not seen in E-MORB (Chen et al., 2019; Ahmad et al., 2021) (Fig. 7a). This is also likely
571 the case for U given the compositions of volcanic arc lavas and AOC (Fig. 7b). Nielsen et al.
572 (2018) suggest a similar model (i.e., two stage melting, with low degree melt metasomatism, and

radiogenic ingrowth followed by sampling under MOR's) but require the addition of subducted sediment, to fit with Ba isotopic data. However, as detailed above, subducted sediments in the E-MORB source are incompatible with Ce/Pb and Pb isotopic data of E-MORB (Fig. 5), as well as with Mo and U elemental and isotopic data (Fig. 6).

Some MORB samples with high "arc-like" $\delta^{98/95}$Mo, low $(La/Sm)_N$, and other arc lava geochemical signatures have been linked to mantle sources with substantial contributions from fluid-modified mantle wedge components that formed during subduction processes such as enrichments in U and Pb that result in lower Nb/U (33) and Ce/Pb (19) (Chen et al., 2025) than canonical mantle values (47 ± 10 and 25 ± 5 respectively) (Hofmann et al., 1986). These MORB samples that preserve so called ghost-arc signatures reflect a way of generating high $\delta^{98/95}$Mo in upper mantle MORB sources with recycled crustal components (Chen et al., 2025). However, our E-MORB samples with high $(La/Sm)_N$ do not show evidence of arc lava signatures. For example they have average Nb/U (48) and Ce/Pb (28) near canonical mantle values (Hofmann et al., 1986). Therefore, the high $\delta^{98/95}$Mo our E-MORB samples cannot be explained by recycled fluid-modified mantle components.

The simplest interpretation of our new E-MORB data from the North Atlantic Ocean is that they do not contain a recycled, subduction zone processed, crustal component (Fig. 6, 7). Therefore, we argue for a model that does not invoke recycled crustal material to explain the Mo and U elemental and isotopic compositions of E-MORB samples.

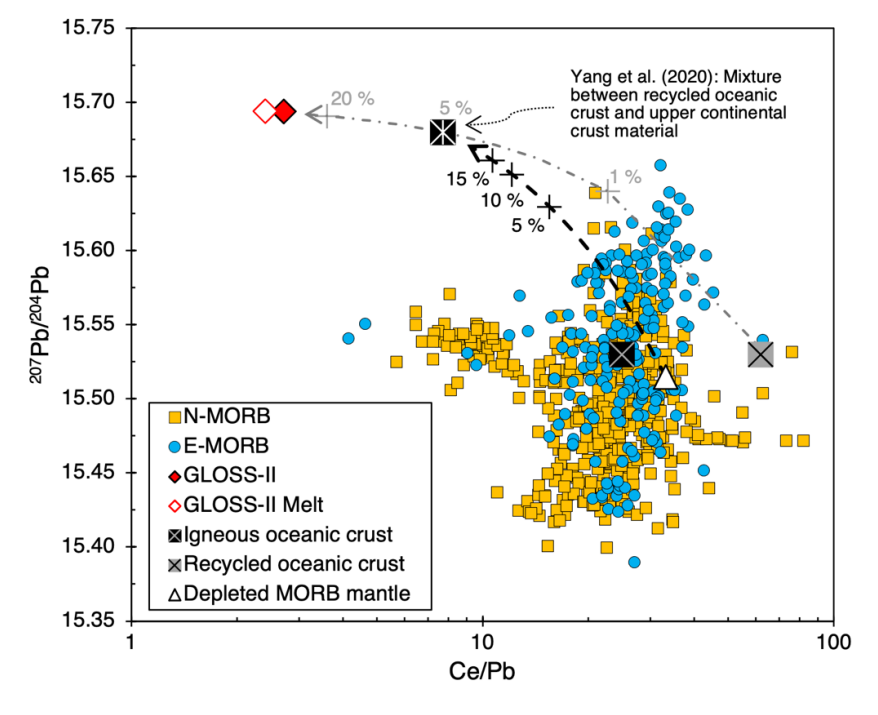


Fig. 5. Mixing model (dashed black line) for $^{207}Pb/^{204}Pb$ versus Ce/Pb between the depleted MORB mantle and subduction zone processed recycled oceanic crust mixed with recycled melted sediment (GLOSS-II, Plank, 2014) in a 95-5 % mixture following Yang et al. (2020) (dash-dotted grey line). Composition of recycled oceanic crust and melted subducted sediment composition have been calculated from Stracke et al. (2003). Mixing models have been calculated using parameters and sources in table S8. Filtered global MORB database from Gale et al. (2013) (only including data obtained by ICP-MS methods).

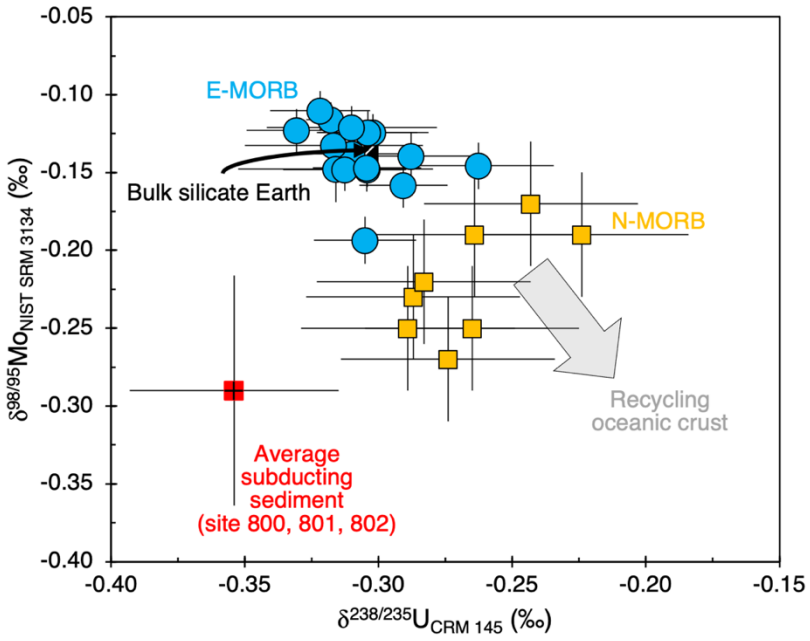


Fig. 6. $\delta^{238}U$ and $\delta^{98/95}Mo$ composition of E-MORB samples measured in this work (blue circles) and literature N-MORB (yellow squares) with U and Mo isotopic data. Symbols and sources for literature data are the same as used in figure 2. The grey arrow shows the effect of mixing recycled oceanic crust into the mantle, which does not explain composition of E-MORB relative to N-MORB.

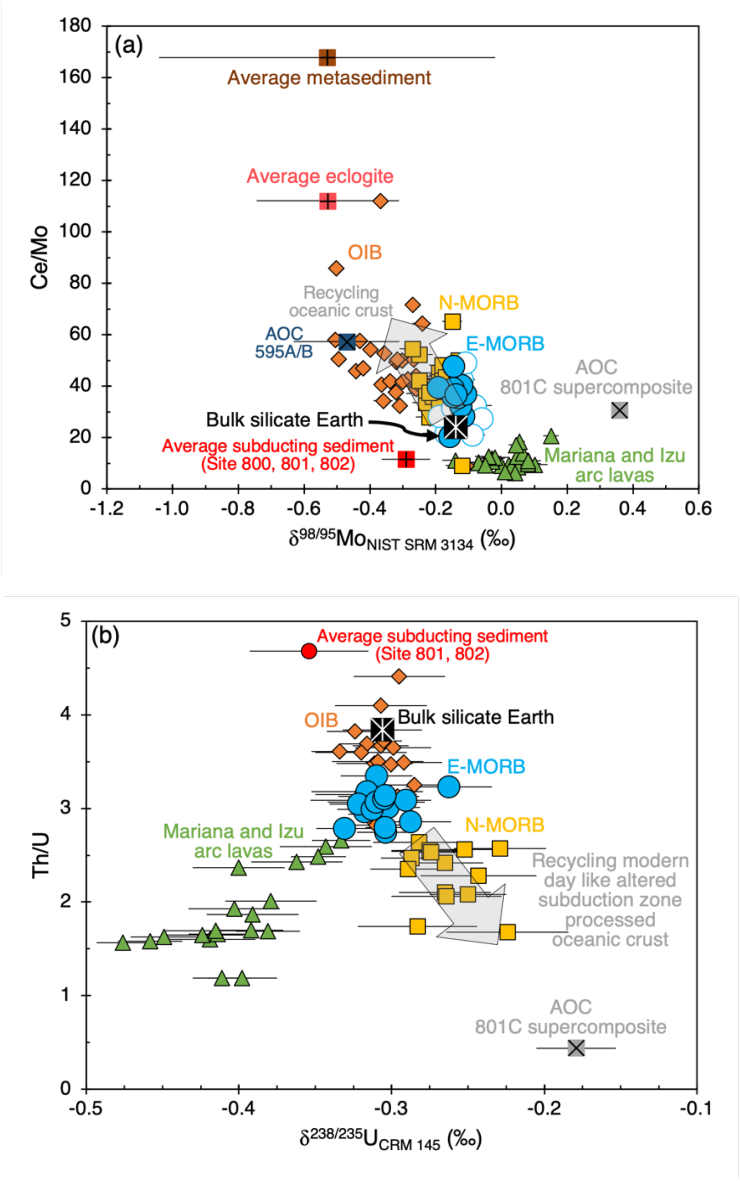


Fig. 7. (a) $\delta^{98/95}$Mo and (b) δ^{238}U versus Ce/Mo and Th/U for N-MORB, E-MORB, AOC, Mariana and Izu arc lavas, OIB, eclogite, and subducting sediment and metasediment. Symbols and sources for literature data are the same as used in figure 2. Literature E-MORB data, shown as hollow blue circles, are from Bezard et al. (2016), Chen et al. (2022) and Hin et al. (2022). Grey arrows show the effect of mixing recycled oceanic crust into the upper mantle, which does not account for composition of E-MORB.

665 5.4 Low degree partial melting and mantle metasomatism

666

667 Low degree partial melting has been suggested to explain the incompatible element enrichment
668 of fluid and non-fluid mobile elements in the E-MORB source (e.g., Niu et al., 2002). Uranium is
669 highly incompatible during mantle melting, $D_{Cpx/melt}$ U^{4+} = ~ 0.02 (Fonseca et al., 2014), and
670 therefore, any potential isotopic fractionation between melt and initial source from partial melting
671 of peridotite will be insignificant as virtually all U will enter the melt. The lower $\delta^{238}U$ of E-MORB
672 than N-MORB can therefore not be explained by isotopic fractionation during partial melting. The
673 $\delta^{238}U$ compositions of our E-MORB samples thus represent the compositions of their upper mantle
674 sources. Their chondritic values reflect mantle compositions little- or un-affected by the recycling
675 of AOC since the onset of oxic deep oceans (~ 600 Ma), which has been argued to increase the
676 $\delta^{238}U$ of the upper mantle as sampled by N-MORB. We suggest that our $\delta^{238}U$ compositions of E-
677 MORB can hence be explained by a model where ancient (≥600 Ma) mantle components are
678 preserved and unaffected by more recent (<600 Ma) crustal recycling.

679

680 This is compatible with a model of low degree partial melting and peridotite metasomatism, as
681 outlined in Niu et al. (2002), and applied from a Mo isotopic perspective in Chen et al. (2022) and
682 for other isotope systems such as Fe (e.g., Guo et al., 2023). Enriched domains in the upper mantle
683 could exist as volumetrically minor lithologies of low degree melts dispersed as (frozen) dykes or
684 veins in a depleted peridotitic matrix (Niu et al., 2002). Low degree melting may occur at the
685 boundary between the thickening and cooling oceanic lithosphere and the asthenosphere. This
686 region is marked by a low velocity zone (e.g., Green, 1971; Niu et al., 2002), which likely reflects
687 the presence of small amounts of melt (e.g., Hirschmann, 2010). These small degree, low volume

688 melts would have low thermal inertia, and will freeze as they migrate into the base of the oceanic
689 lithosphere (McKenzie, 1989). Such metasomatised lithosphere has also been invoked in the
690 source of some alkaline lavas (e.g., Pilet et al., 2008). The existence of alkali volcanism with a
691 chemistry that reflects small degree melting in the presence of garnet, far from any plate boundary
692 or hot-spot in the north-western Pacific plate (Petit spots) (Hirano et al., 2006), has been used as
693 evidence of small degree melts that are actively forming in the modern asthenosphere. The Petit
694 spots anomalously result in a surface expression of this process due to lithospheric fractures from
695 plate flexure during subduction allowing the melts to ascend (Hirano et al., 2006).
696
697 Uranium isotopic data of E-MORB samples requires that the initial low degree melting event
698 happened ≥600 Ma, metasomatising the uppermost mantle and freezing in upper mantle
699 compositions with enriched trace element compositions and chondritic $\delta^{238}U$. Thus, domains
700 enriched in U are isolated from the evolution of ambient upper mantle to higher $\delta^{238}U$ by recycling
701 of oceanic crust altered in oxic deep ocean conditions (<600Ma). A full mechanistic explanation
702 of the model of recycling and mixing of metasomatized oceanic lithosphere into the mantle is not
703 the aim of our work and we refer readers to Niu (2002) and Niu and O'Hara (2003) for details. In
704 brief, these metasomatised portions of the oceanic lithosphere would be subducted, and since they
705 are in deep portions of lithosphere, they would not undergo dehydration. Heating of this material
706 increases its buoyancy, and the crust and mantle separate by buoyancy contrast with crust sinking
707 and thermally buoyant peridotitic mantle rising with metasomatised sections of dykes and veins
708 (Niu et al., 2002; Niu and O'Hara, 2003). Such metasomatised lithosphere is stirred into the upper
709 mantle but remains chemically distinct until ultimately sampled by melting beneath ridges to give
710 rise to E-MORB (Fig. 8) (Niu et al., 2002; Niu and O'Hara, 2003).

Molybdenum isotope data of E-MORB are also compatible with such a model (Chen et al., 2022). but unlike U, the Mo isotopic compositions of E-MORB may be fractionated from its source. In pyroxene, the major upper mantle Mo host, Mo sits in the octahedral M1 site (Leitzke et al., 2017), while in melt Mo^{6+} is coordinated tetrahedrally (Holzheid et al., 1994; O'Neill and Eggins, 2002; Farges et al., 2006). While a minor species at modern ambient upper mantle oxygen fugacity, $Mo^{4+}$is significantly less incompatible than Mo^{6+} in pyroxenes ($D_{Cpx/melt}$ Mo^{4+} = ~ 2) (Leitzke et al., 2017) and is coordinated octahedrally in both mineral and melt (Farges et al., 2006). Heavier isotopes are concentrated in phases with stiffer and stronger bonds, which form between ions with lower co-ordination number and higher valence state (e.g., Schauble, 2004). Melts are therefore predicted to become isotopically heavier in Mo than residues during partial melting (McCoy-West et al., 2019). The fractionation between Mo isotopes becomes larger at smaller degrees of melting and lower Mo^{6+}/Mo_T (Mo total) ratios (i.e., more reduced compositions, unlikely for modern oxidised mantle) (Fig. S7a). Chen et al. (2022), following the melting models of McCoy-West et al. (2019), proposed that at low degrees of melting, ~ 0.2 %, of modern-day depleted mantle, $\delta^{98/95}Mo = -0.2$ ‰ (Hin et al., 2022), at a modern-day redox state, $Mo^{6+}/Mo_T = 0.99$ (O'Neill and Eggins, 2002), results in a melt composition, $\delta^{98/95}Mo$ = ~ −0.01 ‰ and $(La/Sm)_N$ ~ 5.4, that is sufficient to explain the high $\delta^{98/95}Mo$ values and chemical enrichments seen in global E-MORB when mixed with a depleted MORB component (Fig. 9a).

However, we find that with our new data on E-MORB the resulting isotopic compositions from such low degree melting are too isotopically heavy to explain some of our data, even at higher Mo^{6+}/Mo_T ratios, ~ 0.999 (Fig. 9a). McCoy-West et al. (2019) used the 'ionic model' and literature

734 bond length data to derive a fractionation factor between ^{98}Mo and ^{95}Mo during mantle partial
735 melting, $\alpha^{98/95}Mo_{Melt\text{-}Silicate}$, ~ 0.99977. Such a value has not been directly, experimentally verified
736 and notably predicts larger fractionation than for another redox sensitive system Cr (e.g., Jerram
737 et al., 2022) for example. We note that our data imply a smaller melt-silicate fractionation of Mo
738 isotopes, and we illustrate this using $\alpha^{98/95}Mo_{Melt\text{-}Silicate}$ of 0.9999 (Fig. S7b, 9b). Experimental work
739 is required to assess if this empirical reassessment of the fractionation factor is justified.
740
741 Our model fit to $\delta^{98/95}$Mo data is further improved given inferences from δ^{238}U compositions of E-
742 MORB that imply the initial low degree melts form not from a modern-day depleted mantle, but
743 ancient ≥600 Ma depleted mantle compositions (Fig. 8). Isotopic perturbation of Mo in the upper
744 mantle by crustal recycling is inferred to have occurred for longer timescales than for U. Hin et al.
745 (2022) show that at least ~ 1 to 1.4 Gyr of oceanic crust recycling is needed to lower the $\delta^{98/95}$Mo
746 value of the upper mantle from −0.14 ‰ (BSE) to −0.2 ‰ (modern-day depleted upper mantle).
747 Although, as we note, if the melt-silicate fractionation of Mo isotope is smaller than estimated by
748 McCoy-West et al. (2019), these timescales would be shorter, which is possible, as the U isotopic
749 composition of N-MORB suggests that upper mantle compositions can be perturbed by recycled
750 crust within 600 Myr. The isolation of small degree melts from a convecting upper mantle at
751 various ages ≥600 Ma can help explain the variably elevated $\delta^{98/95}$Mo of global E-MORB. We
752 show this on fig. 9b, where low degree melt compositions form from an upper mantle less affected
753 by crustal recycling with higher $\delta^{98/95}$Mo than modern day, resulting in an enriched melt end
754 member composition of $\delta^{98/95}$Mo = ~ −0.1 ‰ and $(La/Sm)_N$ ~ 5.4 at Mo^{6+}/Mo_T = 0.99.
755

In summary, the metasomatism of depleted, oceanic lithosphere with low degree melts formed ≥600 Ma would create a source dominated by a U-rich component with $\delta^{238}U \approx$ BSE and a range of $\delta^{98/95}Mo$ compositions ≥BSE, with variable enrichment in $(La/Sm)_N$ (Fig. 8). Low degree melting would also create variable degrees of enrichment in other chemical tracers of enrichment, such as Nb/Zr, Th/Yb, and Sm/Nd, that also show trends with $\delta^{238}U$ and $\delta^{98/95}Mo$, that distinguish E-MORB from N-MORB (Fig. S8). This enriched component, when stirred back into the upper mantle and sampled by melting beneath a MOR in a larger degree melting event, can explain the chemical compositions of non-hot-spot influenced E-MORB (Fig. 3c, d, 9b). The timescale of ≥600 Ma inferred for this recycling process is about twice as long as the residence time of E-MORB sources calculated by Donnelly et al. (2004) from radiogenic isotope pseudo-chrons. It will be of interest to rationalise these different timescales using a consistent set of radiogenic isotope and $\delta^{238}U$ measurements on the same samples, which is currently not possible.

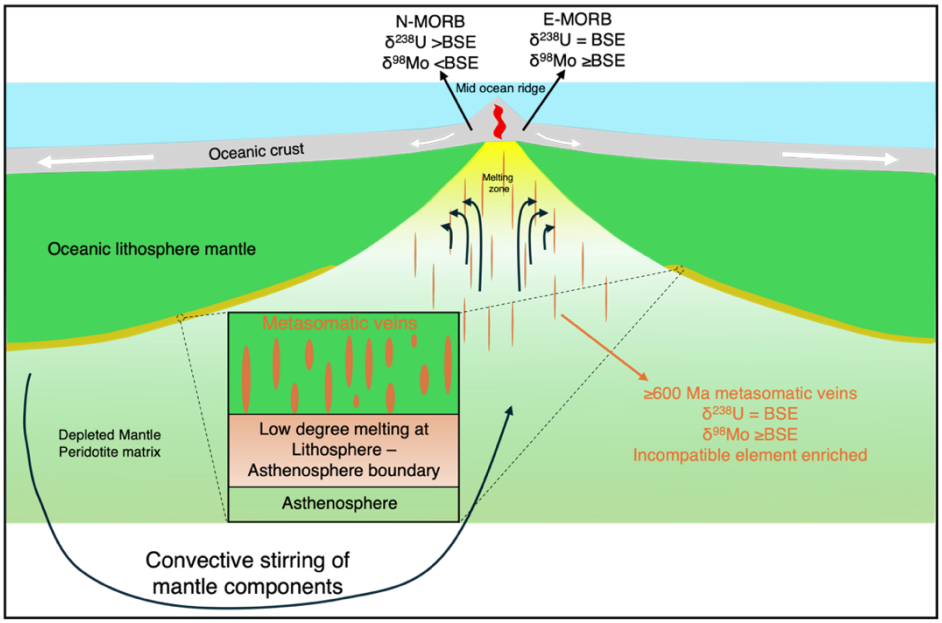
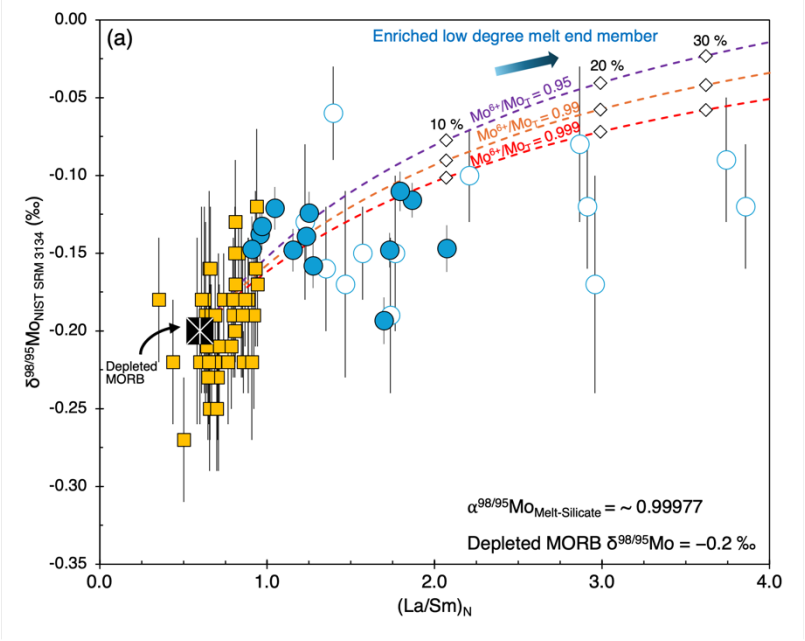


Fig. 8. Cartoon schematic of E-MORB source formation based on and modified from Niu et al. (2002) and Guo et al. (2023). Low degree partial melts enriched in incompatible elements (orange lenses) form ≥600 Ma in the low velocity zone at the boundary between the lithosphere and asthenosphere (shaded orange region at base of oceanic lithosphere mantle) and become trapped by migration and freezing in the cooling and thickening overlying oceanic lithosphere. These components preserve older mantle compositions with BSE $\delta^{238}U$. As oceanic lithosphere is subducted, the metasomatised lithosphere is stirred back into the upper mantle and some is entrained beneath MOR's and melts to produce E-MORB.


(a)
Enriched low degree melt end member
30 %
20 %
10 %
Mo6+/MoT = 0.95
Mo6+/MoT = 0.99
Mo6+/MoT = 0.999
Depleted MORB
δ98/95MoNIST SRM 3134 (‰)
α98/95MoMelt-Silicate = ~ 0.99977
Depleted MORB δ98/95Mo = –0.2 ‰
(La/Sm)N
0.00
-0.05
-0.10
-0.15
-0.20
-0.25
-0.30
-0.35
0.0
1.0
2.0
3.0
4.0

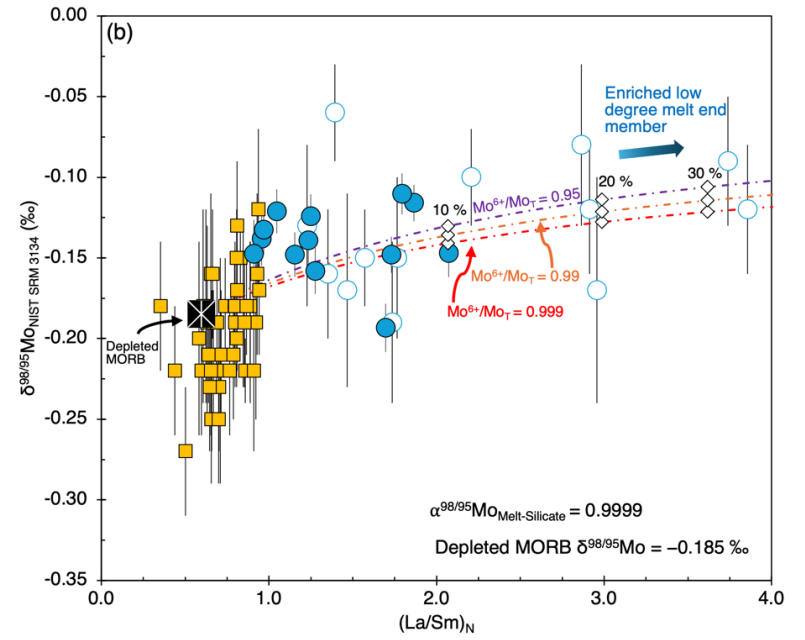

(b)
Enriched low degree melt end member
30 %
20 %
10 %
Mo6+/MoT = 0.95
Mo6+/MoT = 0.99
Mo6+/MoT = 0.999
Depleted MORB
δ98/95MoNIST SRM 3134 (‰)
α98/95MoMelt-Silicate = 0.9999
Depleted MORB δ98/95Mo = –0.185 ‰
(La/Sm)N
0.00
-0.05
-0.10
-0.15
-0.20
-0.25
-0.30
-0.35
0.0
1.0
2.0
3.0
4.0

Fig. 9. Non-modal, batch melting modelling of $(La/Sm)_N$ and Mo isotopic composition of melt. Mixing lines are shown between a depleted MORB (represented by 20 % melting of DMM) and an enriched low degree melt end member generated by 0.2 % melting of DMM at different mantle fO_2 (Mo^{6+}/Mo_T = 0.999 to 0.95). Symbols and sources for data are the same as used in figure 2. Model calculations follow McCoy-West et al. (2019) and Chen et al. (2022). Parameters used are given in table S9. White diamonds indicate mixing proportions. (a) $\alpha^{98/95}Mo_{Melt\text{-}Silicate}$ ~ 0.99977, Depleted MORB $\delta^{98/95}Mo$ = −0.2 ‰ (b) $\alpha^{98/95}Mo_{Melt\text{-}Silicate}$ ~ 0.9999, Depleted MORB $\delta^{98/95}Mo$ = −0.185 ‰.

6. Conclusions

A set of hand-picked E-MORB glasses from the Northern mid-Atlantic ridge picked to different degrees of stringency all have $^{234}U/^{238}U$ ratios close to secular equilibrium, indicating that samples likely reflect primary U and Mo isotopic composition. These E-MORB samples show a limited range of $\delta^{238}U$ and $\delta^{98/95}Mo$ from ~ −0.33 to −0.26 ‰ and −0.19 to −0.11 ‰ respectively and are both, on average, indistinguishable from the bulk silicate Earth (chondritic) U and Mo isotopic compositions. These E-MORB therefore have contrasting sources to modern N-MORB, which are super-chondritic in U and sub-chondritic in Mo isotope ratios, thought to be due to the recycling of modern day like subduction zone processed altered oceanic crust (Andersen et al., 2015; Hin et al., 2022). Our E-MORB data are inconsistent with models that explain their enrichment with recycled oceanic crust and or recycled sediment (e.g., Allègre and Turcotte, 1986; Prinzhofer et al., 1989; Donnelly et al., 2004; Hémond et al., 2006; Waters et al., 2011; Ulrich et al., 2012; Nielsen et al., 2018; Yang et al., 2020) but are consistent with a model of recycled oceanic lithospheric mantle, metasomatised by small degree asthenosphere melts (Niu et al., 2002; Chen et al., 2022; Guo et al., 2023). This process effectively isolates the composition of the upper mantle at the time of the small degree melting until the metasomatised sources are resampled as E-MORB by melting beneath ridges. Uranium isotopic compositions (which are not affected by low degree partial melting) show that this initial low degree melting event occurred in an upper mantle with a chondritic $\delta^{238}U$, namely prior to its contamination with isotopically heavy recycled altered oceanic crust. This corresponds to an age ≥600 Ma, set by the estimated timing of deep ocean oxygenation (Andersen et al., 2015), providing a minimum bound on the antiquity of the E-MORB source. Molybdenum isotopic compositions of global E-MORB are compatible with such

a model but are also potentially fractionated during low degree partial melting to isotopically heavier compositions (McCoy-West et al., 2019; Chen et al., 2022).

Credit authorship contribution statement

Joel B. Rodney: Data curation, formal analysis, investigation, methodology, validation, visualisation, writing – original draft. **Morten B. Andersen:** Supervision, project administration, funding acquisition, conceptualization, methodology, writing – review and editing. **Bramley J. Murton:** Resources, writing – review and editing. **Tim Elliott:** Supervision, project administration, funding acquisition, conceptualization, writing – review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data are available through Mendeley data at:
https://data.mendeley.com/datasets/wm84d8m8js/2

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Appendix A. Supplementary Material

Supplementary Material related to this article can be found online at:

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Constraints from uranium and molybdenum isotope ratios on the origin of enriched mid-ocean ridge basalts

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Supplementary Information

Section 1: MORB glass picking and leaching

Section 2: Full detailed method for U and Mo isotopic analysis

Section 3: Supplementary figures and tables

Section 1:

MORB glass picking and leaching

Hand specimen samples of E-MORB were crushed and processed to ~ 1000 μm size glass chips. Glass chips were further crushed using an agate pestle and mortar, and sieved, aiming for a size of ~ 600 μm. Mid-ocean ridge basalt glasses potentially experience seawater alteration, including Fe–Mn coatings precipitated on their surfaces, which can contain high concentrations of Mo and U. For example, it has been shown that the inclusion of Fe-Mn coatings that have high Mo contents ~ 450 ug g^{-1}, with extremely fractionated Mo isotopic compositions, $\delta^{98/95}$Mo –2 ‰ (Siebert et al., 2003) can decrease $\delta^{98/95}$Mo up to ~ 0.07 ‰ (Hin et al., 2022).

To avoid this, MORB glass samples were hand-picked under a binocular microscope to ensure samples were optically clear and devoid of any potential alteration. While this process has long been employed in various studies of MORB glass, notably in U-series disequilibrium studies (Reinitz and Turekian, 1989; Bourdon et al., 2000), it is laborious, and considering the large quantities of sample often needed for trace element isotopic analysis (e.g., >1 g), a rate limiting step. Hand picking MORB glass can also be a relatively subjective task, and therefore it is unclear exactly what an acceptable limit of "quality" is.

A reliable check of sample alteration is given by $^{234}U/^{238}U$ activity ratios. If unaltered by any recent seawater alteration (that would likely affect U compositions), the ^{238}U decay chain will be in secular equilibrium, and so the activity ratio of ($^{234}U/^{238}U$), will be at unity. Seawater has ($^{234}U/^{238}U$) ~ 1.14 (Kipp et al., 2022) and so elevated ($^{234}U/^{238}U$) in glass samples indicates addition of seawater U onto Fe/Mn coatings, which could also indicate adsorption of isotopically distinctive Mo. We explore the effect of varyingly stringent picking strategies on samples with abundant glass. Different splits of glass, classed A, B, C, and D in decreasing degree of quality were prepared. Samples of quality A is those that are most optically clear and devoid of all alteration and coatings (Fig. S1). Samples of quality B is less optically clear, but still mostly devoid of alteration and coatings (Fig. S1). Samples of quality C is even less optically clear or have inclusions and minor amounts of coatings (Fig. S1). Glass quality D, with large amounts of inclusions, coatings or noticeably different to the bulk sample was rejected and not measured (Fig. S1). While this

increased sample preparation time, it does potentially increase the amount of measurable sample and allows an investigation into how selective MORB glass picking needs to be. The amounts of glass picked for each quality (A, B or C) for each sample varied and are listed in table S7. In some cases, different splits were combined to ensure there was enough sample to measure.

All samples underwent a reductive leaching step prior to dissolution to remove any Fe-Mn coatings that picking had failed to exclude. Picked glass samples were poured into 12 ml centrifuge tubes and rinsed thrice with >18 M Ω.cm water (hereafter milli-Q water). Samples were shaken vigorously overnight using a vortex shaker with a 10 ml mixture of 0.05 M hydroxylamine hydrochloride, 15 % acetic acid and 0.03 M Na-EDTA buffered to pH 4 with NaOH (Gutjahr et al., 2007). The leachate was collected, and samples rinsed thrice with 10 ml milli-Q water that was also added to the collected leachate solution. Collected leachate solutions were analysed in 0.3 M HNO_3 for elemental concentrations using an ICP-MS Element2, at the University of Bristol. As noted by Andersen et al. (2015) and Hin et al. (2022) leaching can result in small amounts of glass dissolution, and thus U and Mo loss. Ratios of the concentrations of elements that absorb to Fe-Mn coatings e.g., U and Mo to those little affected, such as Th, Sc, Ti, and Zr, that would only be removed during glass dissolution, were monitored to examine the effects of leaching.

Uranium and Mo are removed during the leaching process (Fig. S2), and it is possible that the removed U and Mo is either from secondary coatings (Fe-Mn coatings) or from the dissolution of the MORB glass. Three leachates from MORB glass sample JC24-82-21, glass qualities A, B and C were measured before leaching other MORB samples to test the leaching procedure. Uranium was lost mostly in the first leaching step on all qualities of glass, ~ 10 % loss, with then only 1 % or less lost in leaching steps 2 and 3 (Fig. S2). This was associated with systematic changes in U/Th of the leachates (Fig. S3). Comparing U to less mobile elements such as Th (which is only likely to be released during glass dissolution) shows the effects of the leaching procedure (Fig. S3). In the first leachate (sample JC24-82-21) U/Th are elevated, e.g., U/Th >4, and likely reflects the release of secondary coating hosted U (Fig. S3). In the second and third leachates, these ratios all drop, e.g., U/Th <2 and show little change between leach two and three (Fig. S3). This likely reflects only small amounts of U being released, also seen in the similar amount of U loss compared to the bulk sample in leachates 2 and 3 (Fig. S2). Given the large change between the first and

second leaches seen and the little variation between the second and third, it seems reasonable that one to two leaching steps are enough to remove any potential secondary coatings. We used two leaching steps for other samples but note that one is likely sufficient.

When comparing the rest of the data set, for which only two leaching steps were done, similar trends are seen for the amounts of U and Mo loss. For Mo, however, leaching loss is more consistently <5 % and is relatively consistent between leaching steps (Fig. S2). This suggests very little secondary phase hosted Mo, which should be removed in the first leach (as for U), and minimal glass dissolution. The main concern with the leaching process is that it may alter isotopic compositions, however Andersen et al. (2015) and Hin et al. (2022) show that the leaching procedure does not fractionate U or Mo isotopes of fresh glass, from comparing a leached and un-leached BHVO glass sample. Therefore, even if some of U and Mo loss is from glass dissolution rather than the dissolution of secondary coatings, isotopic compositions should not be perturbed.

Leaching all the different qualities of glass picked for a sample allows us to compare if lower quality glass (e.g., C quality) as determined visually had more secondary alteration. However, all different qualities of glass showed largely the same patterns in U and Mo loss (Fig. S2, S3). Given this, our screening for glass quality may have been too broad, i.e., what we assessed as quality C (lowest measured quality), may have been just as high enough quality as quality B or A. This is further seen in the largely indistinguishable isotopic compositions of MORB glass samples of different quality (Fig. S5).

The $\delta^{234}U$ compositions of different qualities of glass picked and leached for each sample, A, B and C (in decreasing order of perceived quality), was used to assess the acceptable quality limit to use in future sampling of quenched MORB glass. The $\delta^{238}U$ compositions of the different qualities of glass picked and leached are all, bar one sample, within analytical uncertainty (Fig. S5a) and bar JC24-89-13, are also within uncertainty of $\delta^{234}U = 0$ (Fig. S5b). Sample JC24-89-13, which is only elevated above 0 by 2.5 ‰ in $\delta^{234}U$ (Fig. S5b), also has a similar $\delta^{238}U$ to other samples (Fig. S5a). The $\delta^{98/95}Mo$ compositions of different qualities of MORB glass are also all within analytical uncertainty (Fig. S5c). For samples with multiple splits, the fact that different qualities of glass are all within secular equilibrium and within uncertainty of one another has two important benefits:

firstly, it allows us to assess the level of MORB glass quality needed to ensure minimal secondary alteration, i.e., quality C; secondly, we can average all the splits (A, B, and C) for individual samples for overall $\delta^{238}U$, $\delta^{234}U$, and $\delta^{98/95}Mo$ compositions (Table S6, S7). This approach is reasonable given that the majority of samples with different splits (A, B, and C) have $\delta^{238}U$ and $\delta^{98/95}Mo$ compositions largely indistinguishable from one another (Fig. S5a, c).

Section 2:

Full detailed method for U and Mo isotopic analysis

For the U isotopic measurements, sample preparation and analysis followed Andersen et al. (2015) with some modification, and for Mo isotopic measurement followed Willbold et al. (2016) and Hin et al. (2022).

Uranium and Mo isotope analysis was conducted in the University of Bristol, Bristol isotope group labs. Approximately 1 g of MORB glass was digested in pre-cleaned Teflon PFA beakers in ~ 24 ml of acid using a mixture of 5:1 15.6 M HNO_3 and Romil UpA 28.1 M HF and placed on a tabletop hotplate at 120 °C for at least 48 hours. Samples were then evaporated to dryness and re-dissolved twice in 6 M HCl to remove fluoride precipitates and achieve full sample dissolution. The double spike tracer, IRMM3636 ^{236}U – ^{233}U, 50:50 (Richter et al., 2008), was added to samples prior to dissolution. The double spike was added to samples according to their U concentrations aiming for a $^{236}U/^{235}U$ ratio of 5. Samples were also spiked with a ^{97}Mo – ^{100}Mo double spike, prepared by the Bristol Isotope group, with a $^{97}Mo/^{95}Mo$ ratio of 47.58 and $^{100}Mo/^{95}Mo$ ratio of 58.32, aiming for a natural Mo-double spike Mo proportion of 0.5. For measurement of Th, U, and Mo, concentrations (non-isotope dilution), small fractions of samples (~ 1 %) were taken and measured on an Element2 at the University of Bristol following Andersen et al. (2014). Measured concentrations of U, Th, and Mo of reference materials measured on the Element2 are in good agreement with literature values (Table S2).

Samples were first processed for U isotopic analysis. Samples were loaded in 40 ml of 1.5 M HNO_3 onto 1 ml of TRU resin (100 – 150 mesh) in commercially available Bio-Rad Poly-Prep columns. Matrix was eluted in 30 ml of 1.5 M HNO_3, before U was collected in 10 ml of 0.3 M HF – 0.1 M HCl. The 40 ml of sample load and first 10 ml of wash was collected and kept for Mo chemistry. Collected U fractions were dried and fluxed in a 1 ml 50:50 15.6 M HNO_3: 30 % Romil SpA H_2O_2 mixture to eliminate any organic material that may have leached off resin into samples. Samples were loaded in 5 ml 3 M HNO_3 onto 0.5 ml of UTEVA resin (100 – 150 mesh), for Th and U separation, with 10 ml of 3 M HNO_3 washed through to elute any residual matrix, before washing through 15 ml of 5 M HCl to elute Th, before collection of U in 6 ml of 0.3 M HF – 0.1

M HCl. Again, any potential organic material was eliminated from samples. Final U collections were then dried and re-dissolved in a requisite amount of 0.2 M HCl (aiming for U concentration of 100 – 300 ng g^{-1}) for isotopic analysis. This procedure achieved efficient removal of Th and Th/U ratios during isotopic analysis were typically <0.005.

Uranium isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS (serial no. 1002) at the Bristol Isotope group in low mass resolution ($M/\Delta M \sim 2000$, 5 to 95 % peak height definition), using the setup detailed in Andersen et al. (2015). Samples were introduced to the plasma using a ~ 40 ul min^{-1} micro-concentric PFA nebuliser connected to a Cetac Aridus (1^{st} generation) desolvating system. A standard sample cone plus X-skimmer cone set up was used. Masses 232 (^{232}Th), 233 (^{233}U), 234 (^{234}U), 235 (^{235}U), 236 (^{236}U), and 238 (^{238}U) were collected in faraday cups, with most cups connected to feedback amplifiers with 10^{11} Ω resistors, apart from 234 which was connected to a 10^{13} Ω resistor and 238 which was connected to a 10^{10} Ω resistor. Before each session instrumental baselines were measured and amplifier gains intercalibrated. Measurements consisted of 80 s of solution uptake to allow the ion beams to stabilise. Sample and standard measurements were preceded by 90 s of washing with 0.4 M HCl – 0.05 M HF, followed an on-peak baseline measurement of reagent blank, 0.2 M HCl, for 20 cycles of 4.194 s integration time. Individual sample and standard measurements consisted of 80 cycles each, with 4.194 s integration time. Each sample was preceded and followed by a measurement of the double-spiked (with a double spike proportion similar to samples) standard CRM-145. Samples and standard were measured at varying concentrations, generally between 100 to 300 ng g^{-1}, correlating to U consumption between ~ 30 to 80 ng per measurement. Procedural blanks were <30 pg U, an insignificant amount compared to amount of U consumed per measurement. Ion beam intensities were corrected for low mass tailings of ion beams and high mass plus hydride tailings of ion beams following Andersen et al. (2015). Each solution was corrected for solution blank and intensities re-calculated.

The measured double spike isotope ratio of $^{233}U/^{236}U$ (Richter et al., 2008) was used with the exponential mass fractionation law to correct for mass fractionation of isotope ratios in samples and bracketing standards. Ratios were also corrected for the minute ^{238}U, ^{235}U, and ^{234}U contributions from the IRM-3636 double spike (Condon et al., 2010; Hiess et al., 2012). Uranium

isotope ratios for $^{238}U/^{235}U$ and $^{234}U/^{238}U$ are reported in δ notation with $\delta^{238}U = [(^{238}U/^{235}U_{Sample} / {}^{238}U/^{235}U_{CRM\text{-}145}) - 1]$ and $\delta^{234}U = [(^{234}U/^{238}U_{Sample} / (^{234}U/^{238}U_{CRM\text{-}145}/ (1–0.0386)) - 1]$. By normalising sample measurements to the average of bracketing CRM-145 analyses, this removes second order non-exponential mass bias effects from the analyses. Note that $\delta^{234}U$ values are reported relative to secular equilibrium, where the CRM-145 standard has a $\delta^{234}U$ of −38.6 ‰ relative to secular equilibrium (Cheng et al., 2013).

Long term external reproducibility at various measured U intensities has been estimated using aliquots of the well characterised reference material BHVO-2 measured during different analytical sessions. The external reproducibility of $\delta^{238}U$ and $\delta^{234}U$ for BHVO-2 at various intensities (e.g., ^{238}U = 200 to 1000 pA) ranges from ± 0.09 to 0.03 ‰, 2sd, and ± 4 to 0.9 ‰, 2sd, respectively. The external reproducibility of unknown samples has been determined from the long-term external reproducibility of BHVO-2 measured at various intensities. As samples were measured at varying intensities (~ ^{238}U = 200 to 1000 pA) depending on the U concentration, BHVO-2 was also ran at varying intensities. Repeat measurements of BHVO-2 were then used to construct error curves using the two standard deviations of measurements in ranges of intensities. A power law was fit to the data for $\delta^{238}U$ and $\delta^{234}U$ at the different intensities and used to approximate errors for unknown samples. This relationship was then used for samples of a given concentration, that corresponds to a given intensity, from which an approximate 2sd could be calculated and an external 2se calculated based on the number of repeats (Fig. S4).

Uranium isotopic measurements of international reference materials analysed (BHVO-2, BCR-2, BIR, uraninite, and CZ-1) agree well with values reported by other studies (Table S3). We also report data for a set of in-house reference materials (LP-45d, GUG-11, and IT-3a) that agree well with previous data, and report data on international reference material W-2A, such that it can be used for comparison in further studies. Full list of reference material data is provided in table S3.

Collected Mo fractions from the TRU resin U chemistry (40 ml of sample load and first 10 ml of 1.5 M HNO_3 wash) were dried and dissolved for chemistry using Eichrom AG 1-X8 100 – 200 mesh anionic resin, following Willbold et al. (2016). Samples were dissolved in 22.5 ml 3 M HCl and 1.25 ml 6 M HCl ready for column chemistry using 2 ml of Eichrom AG 1-X8 100 – 200 mesh

anionic resin in Bio-Rad Poly-Prep columns. At least an hour before samples were loaded onto columns, 1.25 ml of 1 M Ascorbic acid was added to samples and allowed to react to reduce Fe^{3+} to Fe^{2+} which is identified by a colour change of samples from yellow to green/colourless. Matrix was eluted using 6 ml 3 M HCl, 26 ml 0.5 M HCl- 0.5 % H_2O_2, 20 ml 1 M HF and 6 ml milli-Q water, before collection of Mo in 24 ml of 1 M HCl. Collected Mo fractions were dried and fluxed in a 1 ml 50:50 15.6 M HNO_3: 30 % Romil SpA H_2O_2 mixture to eliminate any organic material that may have leached off resin into samples. Samples were dried and re-dissolved in the requisite amount of 0.4 M HNO_3 – 0.4 M HF for a Mo concertation of 200 ng g^{-1} for isotopic analysis.

Processing samples through the U chemistry first has the potential to increase the Mo procedural blank, however procedural blanks for Mo that had been processed through the TRU resins and AG 1-X8 columns were ~ 340 and 380 pg (2 separate procedural blanks); on the same order of magnitude as other studies (Willbold et al., 2016; Chen et al., 2022) and negligible compared to amount of Mo measured (~ 30 ng).

Molybdenum isotopic compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS (serial no. 1020) at the Bristol Isotope group in low mass resolution (M/ΔM ~ 1600, 5 to 95 % peak height definition). Samples were introduced to the plasma using a ~ 40 ul min^{-1} micro-concentric PFA nebuliser connected to a Cetac Aridus (1^{st} generation) desolvating system. Nitrogen and argon flow rates were tuned at the start of each session for optimal sensitivity and stability. A standard sample cone plus H-skimmer cone setup was used. Masses 91 (^{91}Zr), 92 (^{92}Mo), 95 (^{95}Mo), 96 (^{96}Mo), 97 (^{97}Mo), 98 (^{98}Mo), 99 (^{99}Ru), 100 (^{100}Mo), and 101(^{101}Ru) were collected in faraday cups L4 to H4 respectively. All cups would ideally be connected to an amplifier with a 10^{11} Ω resistor, however due to the limited number of 10^{11} Ω resistors available on the Neptune, ^{91}Zr, ^{92}Mo, and ^{101}Ru were connected to 10^{10} Ω resistors. This should not be an issue as ^{91}Zr and ^{101}Ru are interferences and removed in chemistry and ^{92}Mo is not used in the double spike inversion for the calculation of ^{98}Mo/^{95}Mo. Before each session instrumental baselines were measured and amplifier gains intercalibrated. Measurements consisted of 80 s of solution uptake to allow the ion beams to stabilise. Sample and standard measurements were preceded by 90 s of washing with 0.4 M HNO_3 – 0.4 M HF, followed an on-peak baseline measurement of reagent, 0.4 M HNO_3 – 0.4 M HF, blank for 30 cycles of 4.194 s integration time.

Individual sample and standard measurements consisted of 30 cycles of 4.194 s integration time, consuming ~ 30 ng Mo per measurement for solutions measured at 200 ng g^{-1}. Each sample was preceded and followed by a measurement of the double-spiked (with a double spike proportion similar to samples) standard NIST SRM3134. Each solution was corrected for solution blank and intensities re-calculated.

Measurements were internally normalised using a double spike inversion using ^{95}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo, and $^{98}Mo/^{95}Mo$ ratios calculated. Samples were externally normalised to the bracketing standard and $\delta^{98/95}Mo$ calculated ($\delta^{98/95}Mo = [(^{98}Mo/^{95}Mo_{Sample} / {}^{98}Mo/^{95}Mo_{NISTSRM3134}) - 1]$). Ruthenium has interferences with ^{98}Mo (^{98}Ru) and ^{100}Mo (^{100}Ru). Therefore, ^{99}Ru and ^{101}Ru were monitored to allow for corrections to be applied to ^{98}Mo and ^{100}Mo. Ruthenium doping experiments (Chen et al., 2019) show that corrections using ^{99}Ru can accurately correct Mo data. However, there is the potential for overcorrection due to species such as $^{64}Zn^{35}Cl$ and $^{40}Ar_2{}^{19}F$ giving signals at mass 99 (Chen et al., 2019). Therefore, ^{101}Ru is also monitored and can be used for correction. Data was corrected using both ^{99}Ru and ^{101}Ru, and when compared, both methods give the same answer within uncertainty. If the total Ru correction was ever over 0.1 ‰ in $\delta^{98/95}Mo$ the datum was rejected (no data in this study was rejected).

We take a homoscedastic approach to determine our external reproducibility, pooled 2sd, on any single stable Mo isotopic measurement, i.e., one standard-sample-standard measurement (Table S4). Using this approach, we define an external reproducibility of $\delta^{98/95}Mo \pm 0.05$ ‰, 2sd, for a single measurement in a given run. This pooled 2sd is then used to calculate the standard error for a given sample given the number, n, of repeat measurements, typically 4 to 6 for unknown samples. This is identical to the 2sd, ± 0.05 ‰, of 35 repeats of W-2A measured over 4 digestions across 4 measuring sessions, and is similar to that reported in Chen et al. (2022) and Hin et al. (2022). Molybdenum isotopic measurements of international reference materials analysed (BHVO-2, BCR-2, and W-2A) agree well with literature data (Table S4), as well as our data for other internal standards (CPI and GUG-11). A full list of data for reference materials is provided in table S5.

Section 3:

Supplementary figures and tables

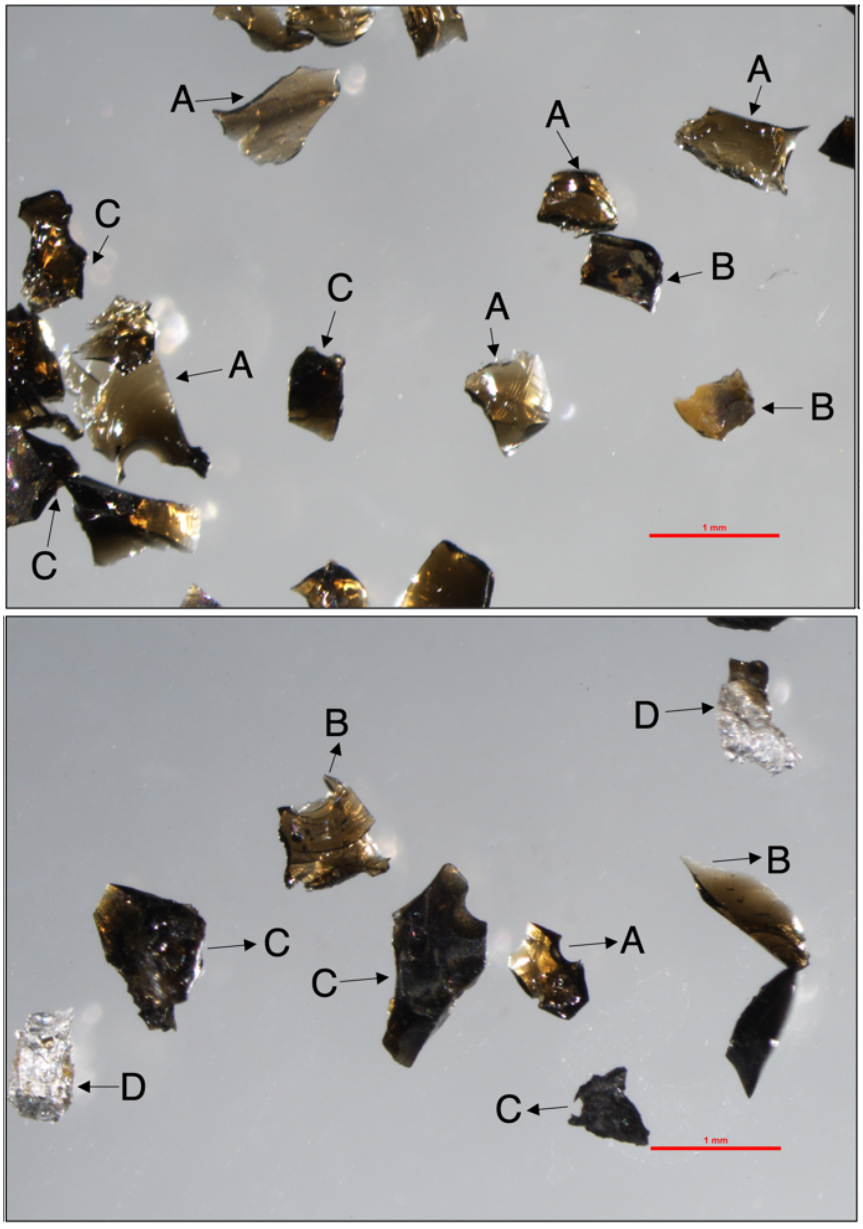


Fig. S1. Example of different qualities of MORB glass picked for sample JC24-80-23. Red scale bar is 1 mm. The letter assigned denotes the quality of glass described in the supplementary text.

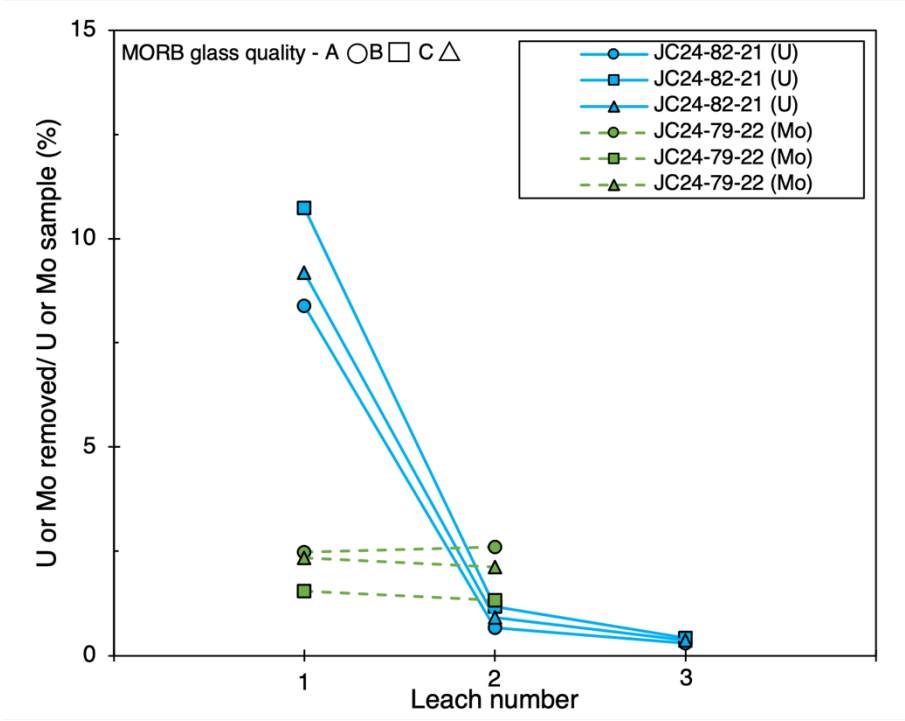


Fig. S2. Percentage of total U (blue) and Mo (green) lost in successive leaches of MORB glass for different glass qualities picked.

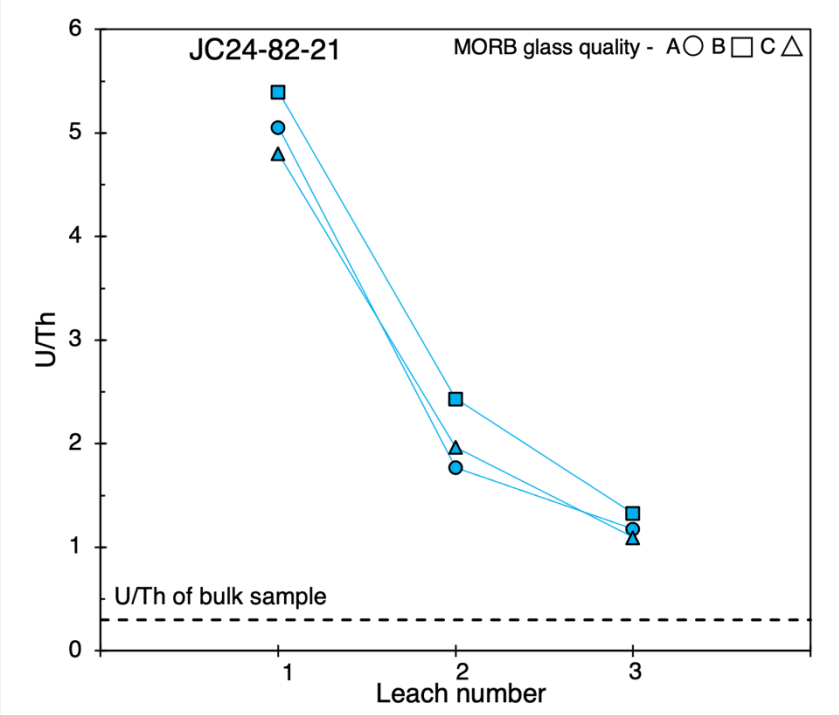


Fig. S3. U/Th ratios in successive leaches of MORB glass for sample JC24-82-21.

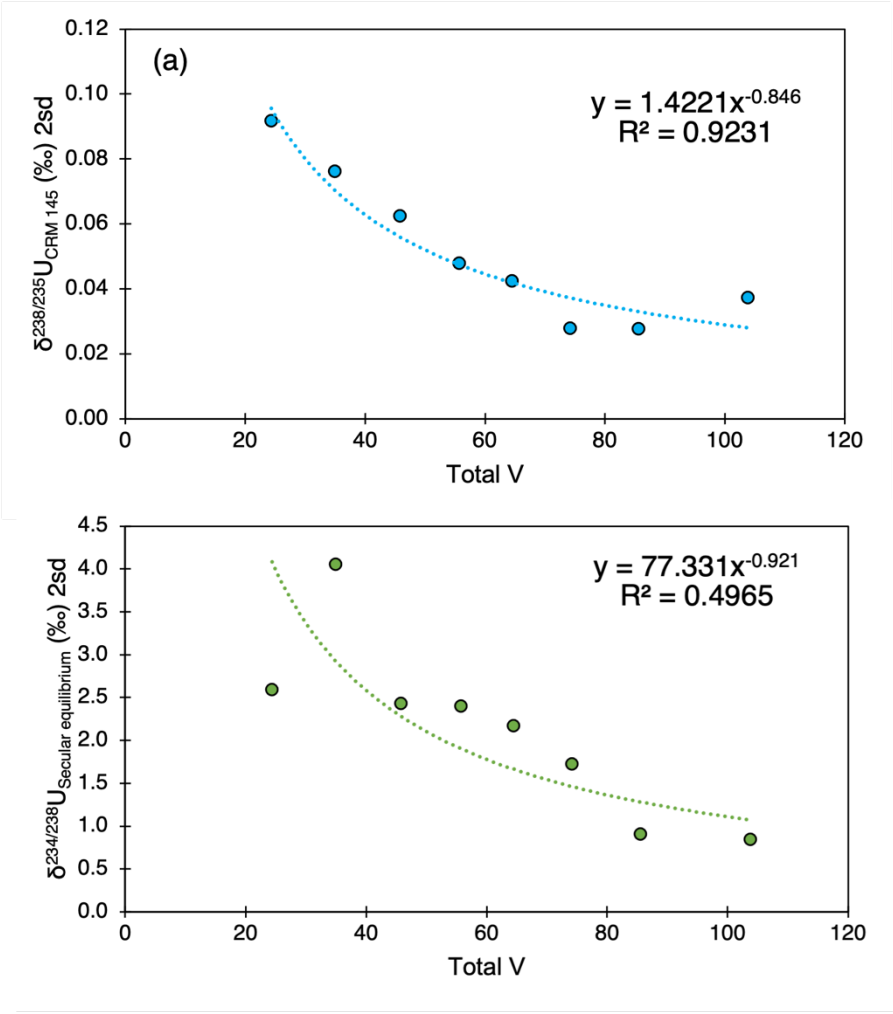
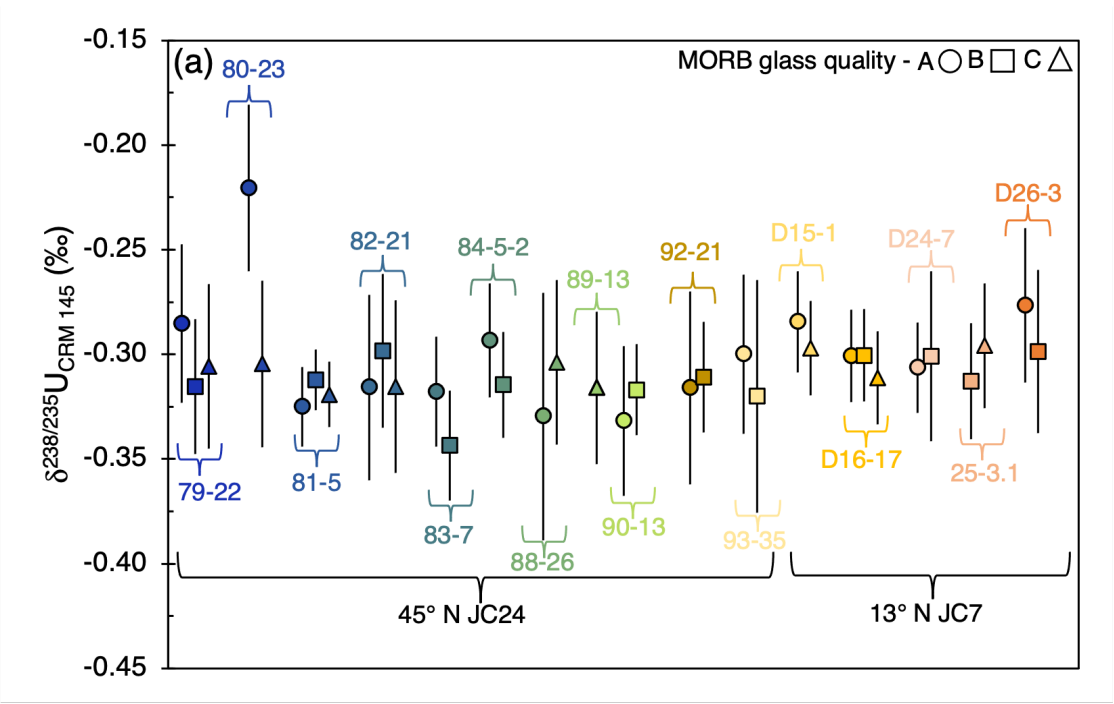
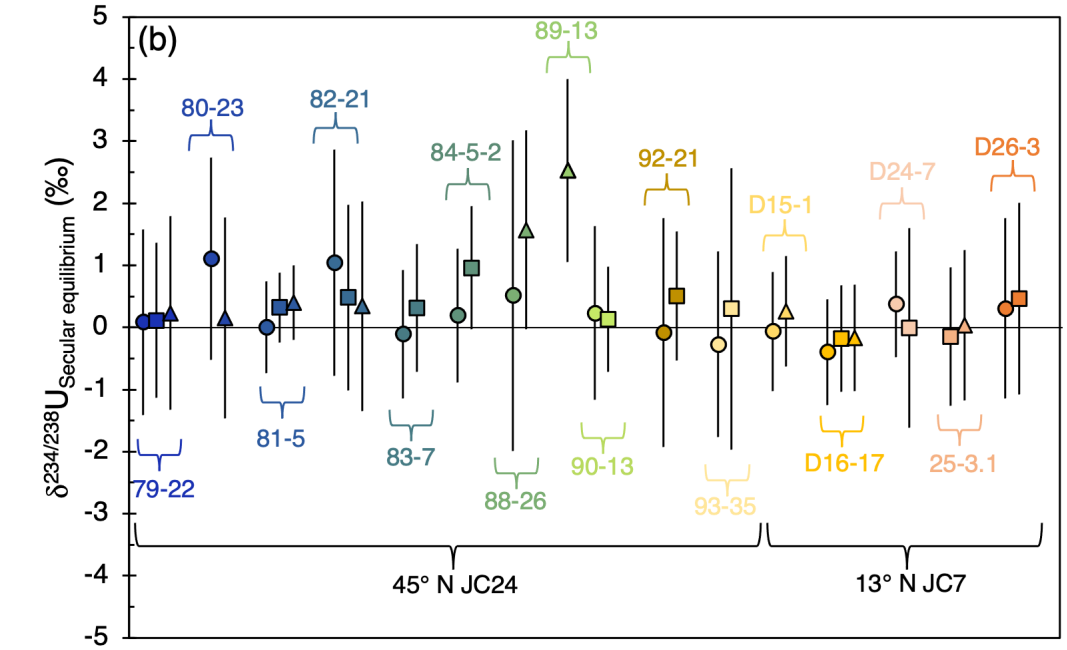


Fig. S4. Error curves for BHVO-2 constructed using the external 2sd of repeat measurements of BHVO-2 at various intensity brackets, ≥20 <30 V, ≥30 <40 V, ≥40 <50 V, ≥50 <60 V, ≥60 <70 V, ≥70 <80 V, ≥80 <90 V and ≥90 V. A power law relationship is fit through the external 2sd. The relationship defined by the power law is then applied to approximate errors for unknown samples. (a) Error curve for $\delta^{238}U$ measurements, (b) Error curve for $\delta^{234}U$ measurements.


(a)
MORB glass quality - A ○ B □ C △
$\delta^{238/235}U_{CRM\ 145}$ (‰)
-0.15
-0.20
-0.25
-0.30
-0.35
-0.40
-0.45
80-23
79-22
81-5
82-21
83-7
84-5-2
88-26
89-13
90-13
92-21
93-35
D15-1
D16-17
D24-7
25-3.1
D26-3
45° N JC24
13° N JC7


(b)
$\delta^{234/238}U_{Secular\ equilibrium}$ (‰)
5
4
3
2
1
0
-1
-2
-3
-4
-5
80-23
79-22
81-5
82-21
83-7
84-5-2
88-26
89-13
90-13
92-21
93-35
D15-1
D16-17
D24-7
25-3.1
D26-3
45° N JC24
13° N JC7

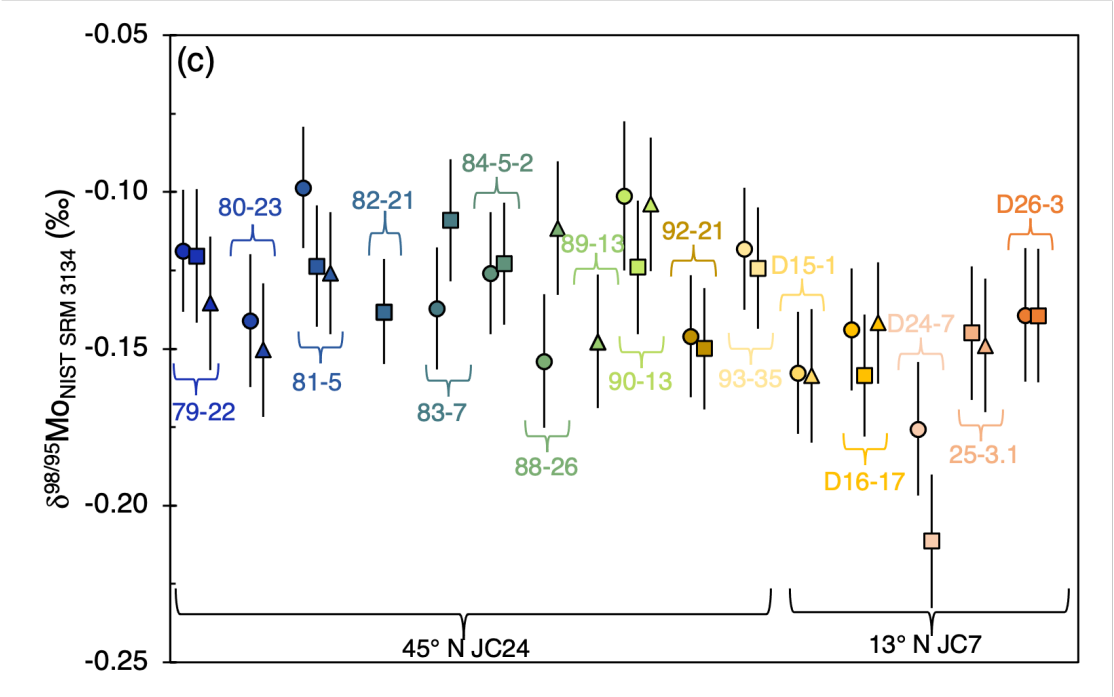


Fig. S5. Isotopic composition of different qualities of MORB glass samples, (a) $\delta^{238}U$ (b) $\delta^{234}U$ and (c) $\delta^{98/95}Mo$. Error bars are 2se.

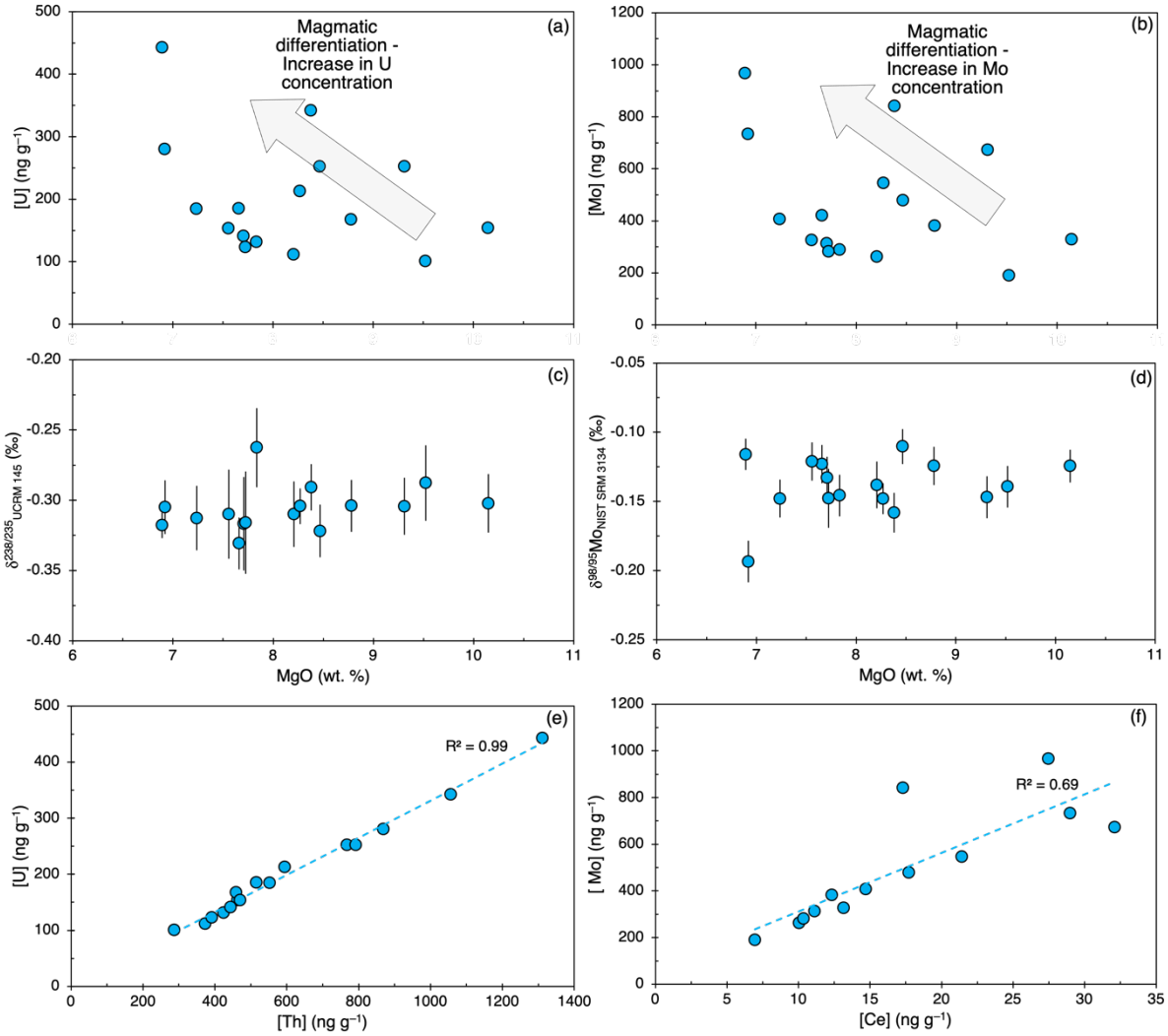


Fig. S6. Variations of E-MORB samples in this study of (a) U concentration, (b) Mo concentration (c) $\delta^{238}U$ and (d) $\delta^{98/95}Mo$ with MgO as a tracer of magmatic differentiation. Linear variation in (e) the U and Th concentration and (f) Mo and Ce concentration of E-MORB samples, trend line is shown as a dashed line with its associated R^2 value.

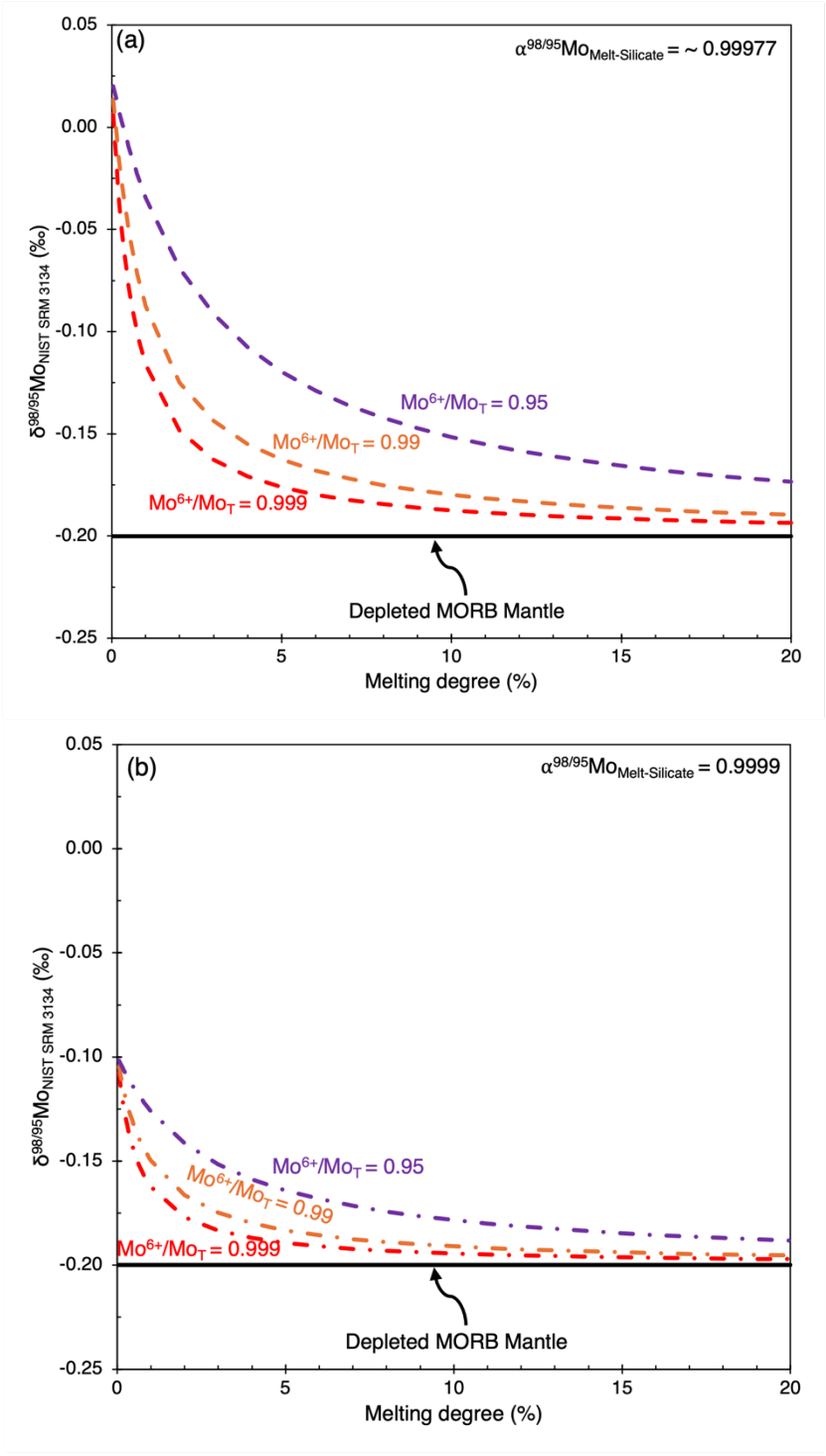


Fig. S7. Non-modal batch melting modelling of Mo isotope composition of melt as a function of melting degree. Model calculations follow McCoy-West et al. (2019) and Chen et al. (2022), parameters used are given in table S8. Compositions are calculated at different mantle fO_2 (Mo^{6+}/Mo_T = 0.999 to 0.95) and start from DMM compositions of $\delta^{98/95}Mo$ = −0.2 ‰ with $\alpha^{98/95}Mo_{Melt\text{-}Silicate}$ of (a) ~ 0.99977 and (b) 0.9999.

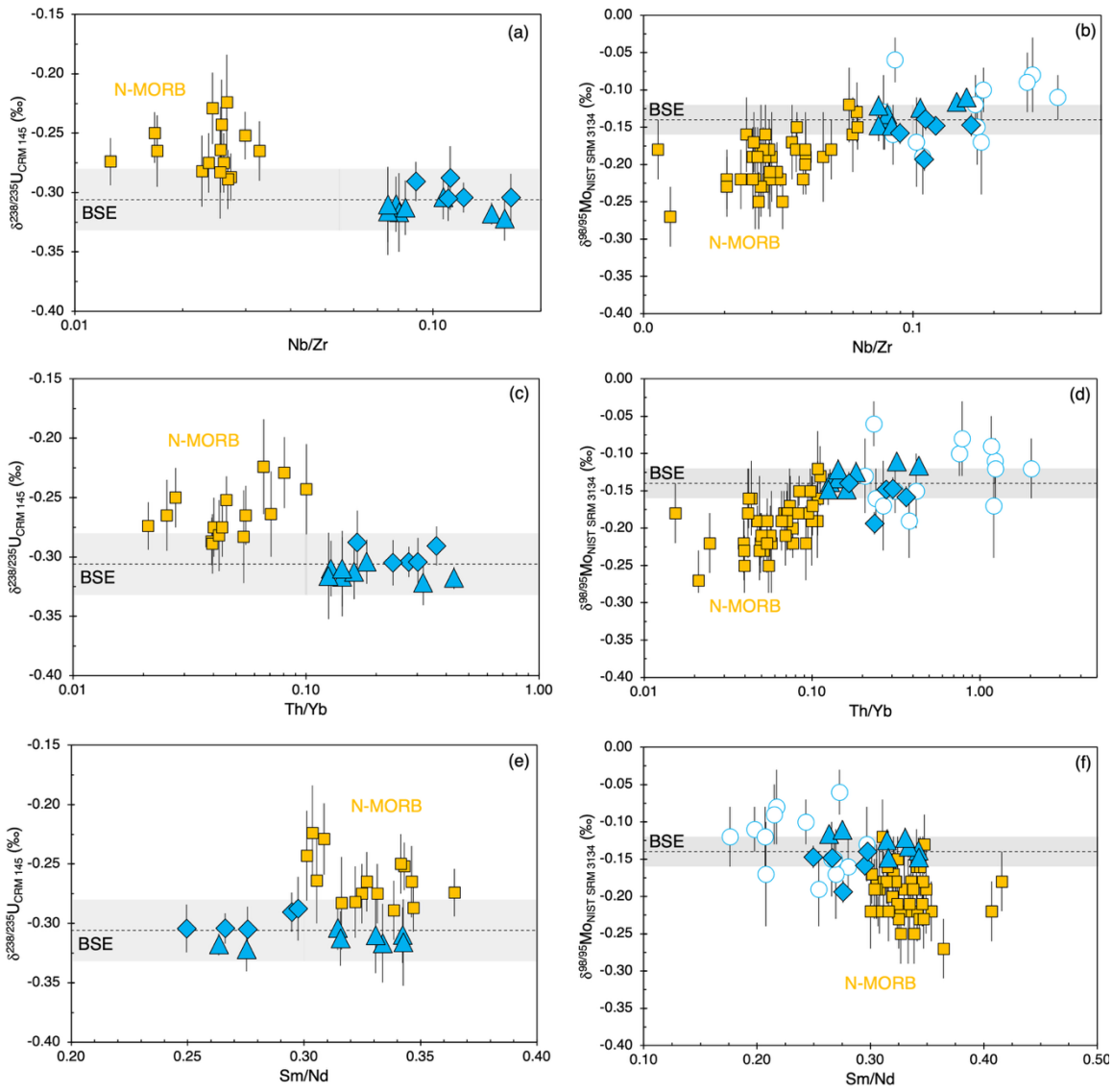


Fig. S8. (a, c, e) $\delta^{238}U$ and (b, d, f) $\delta^{98/95}Mo$ versus (a, b) Nb/Zr, (c, d) Th/Yb, (Sm/Nd) of MORB samples. Enriched-MORB from this study are shown as filled blue symbols and grouped into samples from 45°N (triangles) and 13°N (diamonds). Average N-MORB (yellow squares) concentrations are from Gale et al. (2013). Isotopic data for BSE and N-MORB are from the same sources as in figure 2, and literature E-MORB (hollow circles) are from Bezard et al. (2016), Chen et al. (2022) and Hin et al. (2022). Error bars are ± 2se. Grey shaded regions represents isotopic compositions of BSE (± 2se).

Region	Sample		Latitude (°N)	Longitude (°W)	Depth (m)	Age (Ma)
MAR 45 °N	JC24	79-22	45.49	27.86	2568	1
		80-23	45.44	27.88	2651	2.5
		81-5	45.71	27.78	3457	3
		81-21	45.62	27.85	3062	2.5
		83-7	45.55	27.89	3326	1.5
		84-5-2	45.55	27.88	3088	1
		88-26	45.39	27.88	3048	1.5
		89-13	45.59	27.81	3511	2
		90-13	45.60	27.84	2949	1.5
		92-21	45.48	27.89	3032	2
		93-35	45.48	27.87	2632	1.8
MAR 13 °N	JC7	D15-1	13.07	44.09	4496	0.5 – 1
		D16-17	13.03	44.84	4820	0.5 – 1
		D24-7	13.35	44.90	3011	0.5 – 1
		25-3.1	13.40	44.89	3857	0.5 – 1
		D26-3	13.66	44.97	3496	0.5 – 1

Table S1. E-MORB samples used in this study from the Northern Mid-Atlantic ridge, data for 13 °N is from Wilson et al. (2013).

Sample	BHVO-2	W-2A
Th (ng g^{-1})	1129	2112
Th 2sd	165	401
Th 2se	14	51
U (ng g^{-1})	383	469
U 2sd	57	85
U 2se	5	11
Th/U	2.9	4.5
(Th, U) N	147	63
Mo (ng g^{-1})	3645	400
Mo 2sd	734	104
Mo 2se	88	13
(Mo) N	69	63
Reference Th (ng g^{-1})	1114	2179
Reference U (ng g^{-1})	386	490
Reference Th/U	2.9	4.4
Reference Mo	4070	410

Table S2. List of reference materials used to check reproducibility and precision of measurements of Th, U, and Mo concentrations measured on an ICP-MS Element2. Reference values are from the USGS data sheets.

Sample	$\delta^{238}U$ (‰)	2sd	$\delta^{234}U$ (‰)	2sd	[U] (ng g^{-1})*	N.M	N.S
BHVO-2	–0.306	0.044	–0.1	1.8	419	145	25
	–0.314	*0.020*	*0.9*	*2.4*	*386*	*16*	*8*
BCR-2	–0.255	0.036	0.6	1.4	1687	27	4
	–0.297	*0.020*	*1.1*	*1.0*	*1671*	*1*	*1*
BIR	–0.277	0.189	–5.1	8.6	8	6	3
	–0.285	*0.020*	*–0.6*	*1.0*	*8*	*1*	*1*
CZ-1	–0.048	0.045	–0.2	1.8	7995	30	3
	–0.053	*0.030*	*0.1*	*2.7*		*15*	*2*
GUG-11	–0.460	0.050	0.3	2.0	185	10	2
	–0.419	*0.030*	*–0.4*	*2.0*	*143*	*1*	*1*
IT-3a	–0.289	0.063	0.1	2.6	70	26	10
	–0.296	*0.020*	*–0.3*	*1.7*	*62*	*3*	*1*
LP-45d	–0.319	0.049	–0.2	2.0	2356	83	3
	–0.300	*0.020*	*–0.3*	*4.5*	*2119*	*5*	*2*
Uraninite	–0.548	0.053	–1.5	2.1	11985	51	3
W-2A	–0.291	0.036	2.0	1.4	497	37	7

Table S3. List of reference materials used to check reproducibility and precision during analytical sessions. Data in blue and italics are from Andersen et al. (2015). * Concentrations of U determined from isotope dilution. N.M is the number of individual measurements and N.S is the number of individual samples dissolved and processed through column chromatography.

Sample	2sd	N	Variance × (N−1)
W-2a	0.05	35	0.0184
GUG-11	0.05	19	0.0108
CPI	0.04	56	0.0212
BHVO-2	0.05	20	0.0140
BCR-2	0.05	8	0.0038
JC24-80-23-AB	0.03	5	0.0010
JC24-80-23-C	0.08	5	0.0070
JC24-88-26-C	0.03	5	0.0008
JC24-88-26-AB	0.06	5	0.0032
JC24-89-13	0.08	5	0.0070
JC7-25-3.1-C	0.04	5	0.0019
JC7-25-3.1-B	0.02	5	0.0004
JC24-83-7-A	0.05	6	0.0035
JC24-83-7-B	0.02	6	0.0007
JC24-84-5-2-A	0.03	6	0.0015
JC24-84-5-2-B	0.04	6	0.0022
JC24-92-21-A	0.05	6	0.0036
JC24-92-21-B	0.07	6	0.0057
JC24-93-35-A	0.05	6	0.0033
JC24-93-35-B	0.03	6	0.0012
JC7-D15-1-A	0.06	6	0.0048
JC7-D15-1-C	0.04	5	0.0012
JC7-D24-7-A	0.01	5	0.0001
JC7-D24-7-B	0.06	5	0.0033
JC7-D26-3-A	0.02	5	0.0004
JC7-D26-3-B	0.03	5	0.0012
JC7-D16-17-A	0.05	6	0.0034
JC7-D16-17-B	0.06	6	0.0052
JC7-D16-17-C	0.06	6	0.0050
JC24-81-5-A	0.06	6	0.0044
JC24-81-5-B	0.06	6	0.0048
JC24-81-5-C	0.06	6	0.0044
JC24-79-22-A	0.04	6	0.0023

JC24-79-22-B	0.04	5	0.0018
JC24-79-22-C	0.02	5	0.0003
JC24-90-13-A	0.05	4	0.0018
JC24-90-13-B	0.04	5	0.0016
JC24-90-13-C	0.04	5	0.0012
JC24-82-21-B	0.04	8	0.0035
Sum		326	0.16
Pooled variance	0.0006		
Pooled 2sd	0.05		

Table S4. Homoscedastic approach for calculating external uncertainty on Mo isotopic compositions. Variance is calculated as Variance = $2sd^2/4$ and pooled variance as the sum of variance × (N-1) / sum of N-1. Pooled 2sd is calculated as pooled 2sd = 2 × √Pooled variance.

Sample	$\delta^{98/95}$Mo (‰)	2sd	2se	[Mo] ppb*	N.M	N.S
CPI	–0.27	0.05			56	
	–0.23	*0.04*			*42*	
BHVO-2	–0.06	0.05		4207	20	3
	–0.08	*0.04*		*3570*	*6*	
W-2A	–0.05	0.05		417	35	4
	–0.05	*0.05*		*460*	*9*	
GUG11	0.07	0.05		946	19	1
	0.049		*0.016*	*1003*		
BCR-2	0.00	0.05		259873	8	1
	–0.03	*0.04*		*236000*	*3*	

Table S5. List of reference materials used to check reproducibility and precision during analytical sessions. Data in blue and italics are from Freymuth et al. (2015): GUG-11, Willbold et al. (2016): CPI, Liang et al. (2017): BCR-2, and Chen et al. (2022): BHVO-2 and W-2A. * Concentrations of Mo determined from double spiking. N.M is the number of individual measurements and N.S is the number of individual samples dissolved and processed through column chromatography.

Sample		U (ng g^{-1})	Th/U	$\delta^{238}U$ (‰)	2se	$\delta^{234}U$ (‰)	2se	N	Mo (ng g^{-1})	Ce/Mo	$\delta^{98/95}Mo$ (‰)	2se	N	K_2O/TiO_2	$(La/Sm)_N$
45° N JC24	79-22	154	3.01	–0.302	0.021	0.1	0.8	3	328		–0.12	0.01	16	0.11	
	80-23	132	3.23	–0.263	0.028	0.6	1.1	4	289		–0.15	0.02	10	0.11	
	81-5	443	2.96	–0.318	0.009	0.3	0.4	12	967	28.4	–0.12	0.01	18	0.30	1.87
	82-21	111	3.35	–0.310	0.023	0.6	1.0	6	262	38.3	–0.14	0.02	8	0.13	0.96
	83-7	185	2.79	–0.331	0.019	0.1	0.7	4	420		–0.12	0.01	12	0.16	
	84-5-2	167	2.74	–0.304	0.019	0.6	0.7	4	382	32.3	–0.12	0.01	12	0.17	1.25
	88-26	141	3.15	–0.317	0.033	1.0	1.4	4	313	35.6	–0.13	0.02	10	0.12	0.97
	89-13	123	3.18	–0.316	0.036	2.5	1.5	2	282	36.8	–0.15	0.02	5	0.11	0.91
	90-13	252	3.04	–0.322	0.019	0.2	0.7	3	479	37.0	–0.11	0.01	14	0.27	1.79
	92-21	185	2.99	–0.313	0.023	0.3	0.9	3	407	36.1	–0.15	0.01	12	0.14	1.15
	93-35	153	3.06	–0.310	0.032	0.0	1.3	2	327	40.3	–0.12	0.01	12	0.12	1.05
13° N JC7	D15-1	342	3.09	–0.291	0.016	0.1	0.7	6	842	20.6	–0.16	0.01	11	0.35	1.27
	D16-17	213	2.80	–0.304	0.013	–0.2	0.5	6	546	39.2	–0.15	0.01	18	0.19	1.73
	D24-7	280	3.10	–0.305	0.019	0.3	0.8	4	733	39.5	–0.19	0.02	10	0.37	1.70
	25-3.1	252	3.14	–0.304	0.020	–0.1	0.8	6	672	47.8	–0.15	0.02	10	0.14	2.07
	D26-3	101	2.86	–0.288	0.027	0.4	1.1	2	189	36.7	–0.14	0.02	10	0.22	1.23
Atl. E-MORB				**–0.307**	**0.008**	**0.3**	**0.3**				**–0.14**	**0.01**			

Table S6. U and Mo concentration and isotopic data for E-MORB samples analysed in this work (mean compositions from multiple qualities of glass for each sample), along with select element ratios. Error is the 2se and N refers to the total number of repeat measurements. For the overall Atlantic E-MORB average, isotopic compositions are calculated as concentrated weighted averages, and the error calculated as the 2sd over the square root of total number of samples measured.

Sample		Mass picked (mg)	U (ng g^{-1})	δ^{238}U (‰)	2se	δ^{234}U (‰)	2se	N	Mo (ng g^{-1})	$\delta^{98/95}$Mo (‰)	2se	N
	79-22-A	544	154	–0.285	0.038	0.1	1.5	1	329	–0.12	0.02	6
	79-22-B	1056	154	–0.315	0.032	0.1	1.3	1	330	–0.12	0.02	5
	79-22-C	664	153	–0.306	0.039	0.2	1.6	1	325	–0.14	0.02	5
	79-22		**154**	**–0.302**	**0.021**	**0.1**	**0.8**	**3**	**328**	**–0.12**	**0.01**	**16**
	80-23-AB	814	133	–0.221	0.040	1.1	1.6	2	292	–0.14	0.02	5
	80-23-C	788	130	–0.305	0.040	0.2	1.6	2	286	–0.15	0.02	5
	80-23		**132**	**–0.263**	**0.028**	**0.6**	**1.1**	**4**	**289**	**–0.15**	**0.02**	**10**
	81-5-A	760	449	–0.325	0.019	0.0	0.7	3	976	–0.10	0.02	6
	81-5-B	1182	443	–0.312	0.014	0.3	0.6	5	964	–0.12	0.02	6
	81-5-C	1009	437	–0.319	0.016	0.4	0.6	4	960	–0.13	0.02	6
	81-5		**443**	**–0.318**	**0.009**	**0.3**	**0.4**	**12**	**967**	**–0.12**	**0.01**	**18**
	82-21-A	1042	112	–0.316	0.044	1.0	1.8	2				
	82-21-B	1154	111	–0.298	0.037	0.5	1.5	2	262	–0.14	0.02	8
	82-21-C	1055	111	–0.315	0.041	0.3	1.7	2				
	82-21		**111**	**–0.310**	**0.023**	**0.6**	**1.0**	**6**	**262**	**–0.14**	**0.02**	**8**
	83-7-A	991	185	–0.318	0.026	–0.1	1.0	2	421	–0.14	0.02	6
	83-7-B	1058	185	–0.344	0.026	0.3	1.0	2	420	–0.11	0.02	6
45° N JC24	**83-7**		**185**	**–0.331**	**0.019**	**0.1**	**0.7**	**4**	**420**	**–0.12**	**0.01**	**12**
	84-5-2-A	1030	168	–0.293	0.027	0.2	1.1	2	385	–0.13	0.02	6
	84-5-2-B	1069	166	–0.314	0.025	1.0	1.0	2	379	–0.12	0.02	6
	84-5-2		**167**	**–0.304**	**0.019**	**0.6**	**0.7**	**4**	**382**	**–0.12**	**0.01**	**12**
	88-26-C	1090	140	–0.304	0.039	1.6	1.6	2	315	–0.11	0.02	5
	88-26-AB	720	143	–0.330	0.059	0.5	2.5	2	310	–0.15	0.02	5
	88-26		**141**	**–0.317**	**0.033**	**1.0**	**1.4**	**4**	**313**	**–0.13**	**0.02**	**10**
	89-13	990	**123**	**–0.316**	**0.036**	**2.5**	**1.5**	**2**	**282**	**–0.15**	**0.02**	**5**
	90-13-A	519	253	–0.332	0.036	0.2	1.4	1	480	–0.10	0.02	4
	90-13-B	1054	252	–0.317	0.022	0.1	0.8	2	480	–0.12	0.02	5
	90-13-C	905							477	–0.10	0.02	5
	90-13		**252**	**–0.322**	**0.019**	**0.2**	**0.7**	**3**	**479**	**–0.11**	**0.01**	**14**
	92-21-A	524	184	–0.316	0.046	–0.1	1.8	1	407	–0.15	0.02	6
	92-21-B	1041	185	–0.311	0.026	0.5	1.0	2	407	–0.15	0.02	6
	92-21		**185**	**–0.313**	**0.023**	**0.3**	**0.9**	**3**	**407**	**–0.15**	**0.01**	**12**
	93-35-A	848	148	–0.300	0.038	–0.3	1.5	1	315	–0.12	0.02	6
	93-35-B	501	159	–0.320	0.056	0.3	2.3	1	338	–0.12	0.02	6
	93-35		**153**	**–0.310**	**0.032**	**0.0**	**1.3**	**2**	**327**	**–0.12**	**0.01**	**12**
	D15-1-A	828	343	–0.284	0.024	–0.1	1.0	3	845	–0.16	0.02	6
	D15-1-C	933	340	–0.297	0.023	0.3	0.9	3	838	–0.16	0.02	5
	D15-1		**342**	**–0.291**	**0.016**	**0.1**	**0.7**	**6**	**842**	**–0.16**	**0.01**	**11**
	D16-17-A	992	214	–0.301	0.022	–0.4	0.9	2	546	–0.14	0.02	6
	D16-17-B	1182	212	–0.300	0.022	–0.2	0.9	2	546	–0.16	0.02	6
	D16-17-C	1131	212	–0.311	0.022	–0.2	0.9	2	545	–0.14	0.02	6
	D16-17		**213**	**–0.304**	**0.013**	**–0.2**	**0.5**	**6**	**546**	**–0.15**	**0.01**	**18**
	D24-7-A	1090	282	–0.306	0.022	0.4	0.9	3	729	–0.18	0.02	5
13° N JC7	D24-7-B	492	275	–0.301	0.041	0.0	1.6	1	737	–0.21	0.02	5
	D24-7		**280**	**–0.305**	**0.019**	**0.3**	**0.8**	**4**	**733**	**–0.19**	**0.02**	**10**
	25-3.1-C	917	252	–0.296	0.030	0.0	1.2	3	675	–0.15	0.02	5
	25-3.1-B	822	253	–0.313	0.028	–0.2	1.1	3	670	–0.14	0.02	5
	25-3.1		**252**	**–0.304**	**0.020**	**–0.1**	**0.8**	**6**	**672**	**–0.15**	**0.02**	**10**
	D26-3-A	1031	101	–0.277	0.037	0.3	1.5	1	201	–0.14	0.02	5
	D26-3-B	1019	100	–0.299	0.039	0.5	1.5	1	178	–0.14	0.02	5
	D26-3		**101**	**–0.288**	**0.027**	**0.4**	**1.1**	**2**	**189**	**–0.14**	**0.02**	**10**

Table S7. Full table of amounts of MORB glass picked, U and Mo concentrations and isotopic compositions of MORB glass samples measured in this work split into each quality of MORB glass measured, mean compositions are shown in bold. For mean compositions the average is calculated as the average over all individual measurements.

	Pb (ug g^{-1})	Ce (ug g^{-1})	Ce/Pb	$^{207}Pb/^{204}Pb$
Igneous oceanic crust	0.48[a]	11.95[a]	24.9	15.53[b]
Recycled oceanic crust	0.0912[a]	5.8555[a]	64.21	15.53[b]
Mobility (%)	81[a]	51[a]		
GLOSS-II	21.2[c]	57.6[c]	2.7	15.694[c]
GLOSS-II melt	18.444[a]	44.352[a]	2.4	15.694[c]
Mobility Sediment-Melt (%)	13[a]	23[a]		
DMM	0.0232[d]	0.772[d]	33.3	15.515[e]

1873 Table S8. Compositions used in the mixing models shown in figure 5. a - Stracke et
1874 al. (2003). b – 801C average from Hauff et al. (2003). c – Plank (2014). d – Salters
1875 and Stracke (2004). e – Gale et al. (2013). The composition of recycled oceanic crust
1876 and GLOSS-II melt have been calculated using the element mobility % given in
1877 Stracke et al. (2003).

Phase	Olivine	Orthopyroxene	Clinopyroxene	Garnet
D_{La}	0.0005	0.004	0.015	0.0007
D_{Sm}	0.0011	0.02	0.1509	0.23
D_{Mo}	0.013	0.026	0.0053	0.017*
D_{Mo}^{4+}	0.5	0.7	0.3	
D_{Mo}^{6+}	0.006	0.009	0.001	
K'^{-1}	0.0000074	0.000012	0.0000015	
Modal %	0.52	0.22	0.16	0.1
Melt mode %	0.08	-0.19	0.81	0.3

DMM	Concentration (ug g^{-1})	Isotopic composition (‰)
La	0.234	
Sm	0.270	
Mo	0.025	–0.2 / –0.185

Force constant	Mineral	Melt
Mo	650	1041

1878 Table S9. Model parameters for non-modal batch melting calculation and Mo isotopic
1879 fractionation. Partition coefficients for La and Sm are from Salters and Stracke (2004). Partition
1880 coefficients for Mo and K'^{-1} are from Leitzke et al. (2017). Partition coefficient for Mo in garnet

is assumed to be ≈ to Ce and is taken from Salters and Stracke (2004). Modal % and Melt modes % are from Walter (1998). Depleted MORB Mantle (DMM) compositions are from Salters and Stracke (2004) and isotopic compositions are from Hin et al. (2022). Force constants for Mo in mineral and melt are from McCoy-West et al. (2019). Non-modal batch melting models and Mo isotopic fractionation models follow McCoy-West et al. (2019) and Chen et al. (2022).

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