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

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Most mid-ocean ridge basalts (MORB) are depleted in highly incompatible elements relative to 26 27 the primitive mantle and are termed normal (N)-MORB. Some MORB, erupted at ridge segments 28 distal from mantle hot-spots, are enriched in incompatible elements. The origin of these enriched 29 (E)-MORB is debated, although many studies have proposed that recycled oceanic crust shapes 30 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 δ^{238} U and 31 low $\delta^{98/95}$ Mo relative to the bulk silicate Earth (BSE). Here, we provide U and Mo isotopic data 32 on E-MORB samples from the northern mid-Atlantic ridge (13° & 45° N). We analysed hand-33 picked, leached MORB glass, yielding ²³⁴U/²³⁸U near secular equilibrium, therefore reflecting 34 samples unperturbed by surface processes. Samples have uniform δ^{238} U and $\delta^{98/95}$ Mo, with means 35 of -0.307 ± 0.032 ‰, 2sd, and -0.14 ± 0.04 ‰, 2sd, respectively, both within uncertainty of BSE, 36 and distinct from N-MORB. These data, as well as unremarkable Ce/Pb and radiogenic Pb isotopic 37 38 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 39 40 partial melting of the uppermost mantle that metasomatises the sub-oceanic lithosphere. Given 41 BSE-like U isotopic compositions of E-MORB, that are isotopically unfractionated during low 42 degree partial melting, we suggest that the initial melting event must have occurred prior to the 43 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 44 45 mantle preserve these ≥ 600 Ma U isotopic compositions, which are subducted and stirred back into 46 the convecting upper mantle, ultimately to be sampled at ridges as E-MORB. Molybdenum

47	isotopic compositions of E-MORB are in line with such a model but also reflect isotopic
48	fractionation to higher $\delta^{98/95}$ Mo during low degree partial melting of ≥ 600 Ma upper mantle, that
49	counter acts the lowering of $\delta^{98/95}$ Mo in the upper mantle by an on-going process of plate recycling.
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51	Keywords: U isotopes; Mo isotopes; Enriched MORB; Crustal recycling; low degree partial
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70 **1. Introduction**

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Mid-ocean ridge basalts (MORB), magmatic samples of the upper mantle, are chemically 72 73 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 74 75 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 76 primitive mantle, (La/Sm)_N <1) (Fig. 1) (McDonough and Sun, 1995). Rarer, enriched (E)-MORB 77 78 have incompatible element abundances markedly higher than N-MORB and are associated with 79 elevated ratios of more to less incompatible elements (e.g., $(La/Sm)_N \ge 1$) and distinctive isotopic signatures (e.g., radiogenic ⁸⁷Sr/⁸⁶Sr). While their presence is well documented, the exact 80 81 definition of E-MORB varies in different studies (Fig. 1).

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83 Some E-MORB locations are from topographically elevated MOR sections and are linked to 84 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 85 86 of the chemical enrichment is debated. It has been proposed that recycling of oceanic crust and/or 87 continental sediments into the upper mantle causes enrichment (e.g., Allègre and Turcotte, 1986; 88 Prinzhofer et al., 1989; Hémond et al., 2006; Waters et al., 2011; Ulrich et al., 2012; Yang et al., 89 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 90 91 material. Donnelly et al. (2004) argue for low degree partial melting of subducting crust that 92 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) propose a similar model but also argue for the addition of subducted sediment to the overlying 94 mantle that induces low degree partial melting to produce the E-MORB reservoir that is ultimately 95 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 upper mantle and sampled at ridges (e.g., Green, 1971; Kostopoulos and Murton, 1992; Halliday 99 et al., 1995; Niu et al., 2002; Chen et al., 2022; Guo et al., 2023). 100

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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 $\delta^{98/95}$ Mo_{NIST SRM3134} (the relative difference in 98 Mo/ 95 Mo between samples and standard reference 109 110 material NIST SRM3134, hereafter written $\delta^{98/95}$ Mo in the text), and U isotope ratios as δ^{238} U_{CRM145} (the relative difference in 238 U/ 235 U between samples and certified reference material 111 CRM-145, hereafter written δ^{238} U in the text). 112

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Slab dehydration during subduction releases oxidising fluids with high $\delta^{98/95}$ Mo and low δ^{238} U into the overlying mantle wedge, as inferred from the compositions of volcanic arc lavas (Andersen et

116 al., 2015; Freymuth et al., 2015, 2019; König et al., 2016; Gaschnig et al., 2017; Villalobos-117 Orchard et al., 2020) (Fig. 2). Exhumed eclogites and metasediments with low $\delta^{98/95}$ Mo also reflect 118 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 119 120 processing (Ahmad et al., 2021). The U isotopic system further reflects the importance of seawater 121 alteration of the oceanic crust (Fig. 2b). Seawater alteration of oceanic crust strongly increases its 122 U concentration (e.g., Staudigel et al., 1995) with the added U, on average, being isotopically 123 heavy (Andersen et al., 2015, 2024). Subducting slab dehydration and seawater alteration therefore 124 result in residual slabs with compositions that are isotopically light in Mo and isotopically heavy 125 in U. The recycling of Mo and U from this crustal material into the upper mantle is inferred from 126 compositions of N-MORB samples that are isotopically lighter in Mo and heavier in U than the 127 bulk silicate Earth (BSE) as defined by chondrites (Burkhardt et al., 2014; Andersen et al., 2015; 128 Hin et al., 2022) (Fig. 2). This can also be seen in higher Ce/Mo ratios and lower Th/U ratios of 129 N-MORB than the BSE. Cerium and Th are of similar incompatibility to Mo and U respectively 130 during mantle melting, but with different fluid mobility; Mo and U are aqueous fluid mobile, Ce and Th are not. 131

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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 ²³²Th/²³⁸U in MORB relative to the time integrated ²³²Th/²³⁸U 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 δ^{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).

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146 Molybdenum and U isotopes offer a way to investigate if recycled oceanic crust is mixed into the 147 E-MORB source and provide constraints on the timescales it takes to 'pollute' the upper mantle 148 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 149 150 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 δ^{238} U on the same samples. Herein we 151 152 combine Mo with U isotopic measurements on a set of E-MORB samples from the northern Mid-153 Atlantic ridge (MAR) as a diagnostic test for recycled crustal components in the E-MORB source. 154 155 156 157 158 159

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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).



Fig. 2. (a) $\delta^{98/95}$ Mo versus Ce/Mo and (b) δ^{238} U versus Th/U for mantle derived basalts, AOC, 207 208 subduction processed N-MORB-like eclogite and subducting sediment. Ocean island basalts 209 (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), 210 211 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 212 Pacific subducting sediment composition (red square with a plus sign) is from Ocean Drilling 213 214 Programme sites 800, 801, and 802 from Andersen et al. (2015) and Freymuth et al. (2015). We 215 use the global average subducting sediment Th/U ratio from Plank (2014) GLOSS-II. Average 216 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 217 218 average composition, \pm 1sd, weighted by Mo concentration from Ahmad et al. (2021). Average 219 composition, ± 1 sd, weighted by Mo concentration of a set of exhumed eclogites (pink square with 220 a plus sign) are from Chen et al. (2019) and Ahmad et al. (2021). Average composition, \pm 1sd, 221 weighted by Mo concentration of metasediments (brown square with a plus sign) is from Ahmad 222 et al. (2021). Mid-ocean ridge basalt data (yellow squares) are from Andersen et al. (2015), Bezard 223 et al. (2016), Chen et al. (2022), and Hin et al. (2022). For MORB Mo data we follow the filtering 224 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 225 reproduced and show markedly higher $\delta^{98/95}$ Mo compositions for MORB samples than Bezard et 226 227 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. 228

2. Geological location and samples

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232 We report U and Mo elemental and isotopic compositions for sixteen MAR E-MORB samples, 233 located at $\sim 13^{\circ}$ N 44° W (five samples) and 45° N 28° W (eleven samples) that span MgO contents 234 from ~ 10 to 7 wt. %. Our samples are chemically enriched, with either $(La/Sm)_N \ge 1$ and or $K_2O/TiO_2 \ge 0.11$. Enriched-MORB in the 13° N 44° W segment are common, and basalts have up 235 to forty times higher concentrations of highly incompatible elements than N-MORB from 236 237 segments nearby (Bougault et al., 1988). Samples from 13° N 44° W were collected by dredging 238 in the RSS James Cook JC007 cruise in March – April 2007 (Wilson et al., 2013) (Table S1). They 239 represent a group of basalts erupted before the formation of oceanic core complexes in the area, 240 and now sit off-axis, having been erupted at $\sim 0.5 - 1$ Ma. Samples from $\sim 45^{\circ}$ N 28° W are also 241 from a region where E-MORB commonly occurs (Bougault et al., 1988) and samples were 242 collected in situ during RSS James Cook cruise JC024 May – June 2008 and are all <3 Ma (Table 243 S1) (Searle et al., 2010). Major and trace element data for both sample sets were collected 244 following the methods in Wilson et al. (2013).

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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

252	the zone of influence of the Azores hot-spot, and geochemical enrichment is not linked to hot-spot-
253	ridge interaction (Bougault et al., 1988).
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- **3.** Methods
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277 Fresh MORB glass was crushed and processed to $\sim 600 \ \mu m$ size chips, using an agate pestle 278 and mortar. To avoid samples potentially affected by seawater alteration (e.g., Fe-Mn oxide 279 coatings), samples were hand-picked under a binocular microscope to ensure samples were 280 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 281 282 considering the quantities needed for isotopic analysis (e.g., >1 g), it is a rate limiting step. Hand 283 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 ²³⁴U/²³⁸U 284 activity ratios. If unaffected by recent seawater alteration, the ²³⁸U decay chain will be in secular 285 equilibrium, and so the activity ratio of $^{234}U/^{238}U$, typically expressed as ($^{234}U/^{238}U$), will be unity. 286 Seawater has $(^{234}U/^{238}U) \sim 1.14$ (e.g., Kipp et al., 2022) and elevated $(^{234}U/^{238}U)$ in samples may 287 288 indicate the addition of seawater U and other elements onto Fe-Mn oxide coatings (Siebert et al., 289 2003; Hin et al., 2022). We explored varyingly stringent picking strategies on samples. Up to three 290 different splits of glass of varying quality (A, B, and C in decreasing order of quality) were 291 prepared (classifications are detailed in Supplementary Material: Section 1) (Fig. S1). In some 292 cases, different splits were combined to ensure there was enough sample to measure.

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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.

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306 Uranium isotopic measurement methods followed Andersen et al. (2015), and Mo isotopic 307 measurement methods followed Willbold et al. (2016) and Hin et al. (2022), as detailed fully in 308 Supplementary Material: Section 2. Uranium and Mo isotope analyses were conducted in the 309 Bristol Isotope Group labs, University of Bristol. Approximately 0.5 to 1 g of MORB glass was 310 dissolved and after achieving full dissolution, a ~ 1 % fraction of samples was measured on an 311 Element2 ICP-MS for Th, U, and Mo concentration (Andersen et al., 2014). Measured reference 312 materials are in good agreement with literature values (Supplementary Material: Section 2) (Table 313 S2).

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Samples were spiked with the IRMM3636 ${}^{236}\text{U} - {}^{233}\text{U}$ 50:50 double spike (Richter et al., 2008) aiming for a ${}^{236}\text{U}/{}^{235}\text{U}$ ratio of 5. Samples were also spiked with an in-house ${}^{97}\text{Mo} - {}^{100}\text{Mo}$ double spike, with a ${}^{97}\text{Mo}/{}^{95}\text{Mo}$ ratio of 47.58 and ${}^{100}\text{Mo}/{}^{95}\text{Mo}$ ratio of 58.32, aiming for a natural Modouble 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 $100 - 300 \text{ ng g}^{-1}$) for isotopic analysis. Procedural blanks were <30 pg U, negligible compared to amount of U consumed per measurement, 30 - 80 ng.

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Uranium isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS (serial no. 1002) in low mass resolution ($M/\Delta M \sim 2000$, 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 (²³²Th), 233 (²³³U), 234 (²³⁴U), 235 (²³⁵U), 236 (²³⁶U), and 238 (²³⁸U) 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.

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

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

This results in an estimated external reproducibility of δ^{238} U and δ^{234} U from ± 0.09 to 0.03 ‰, 2sd, and ± 4 to 0.9 ‰, 2sd, for ²³⁸U intensities from 200 – 1000 pA ranges respectively (Fig. S4). Uranium isotopic measurements of international reference materials (BHVO-2, BCR-2, BIR, W-2A, and CZ-1) agree with literature values (Table S3).

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Collected Mo fractions from the TRU resin U chemistry were dried and dissolved for Mo chemistry using Eichrom AG 1-X8 anionic resin. Final Mo collections were dried and re-dissolved in 0.4 M HNO₃ – 0.4 M HF for a Mo concertation of 200 ng g⁻¹ for isotopic analysis. Procedural blanks were <400 pg Mo, negligible compared to the amount of Mo consumed per measurement, ~ 30 ng.

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354 Molybdenum isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS 355 (serial no. 1020) in low mass resolution (M/ Δ M ~ 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 356 357 a Cetac Aridus (1st generation) desolvating system. Masses 91 (91Zr), 92 (92Mo), 95 (95Mo), 96 (⁹⁶Mo), 97 (⁹⁷Mo), 98 (⁹⁸Mo), 99 (⁹⁹Ru), 100 (¹⁰⁰Mo), and 101 (¹⁰¹Ru) were measured 358 359 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 integration time. 361

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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

366	both ⁹⁹ Ru and ¹⁰¹ Ru respectively as monitors of Ru intensity; both corrections yielded the same
367	Mo isotope ratios within uncertainty of one another and uncorrected data. We take a homoscedastic
368	approach to determine our external reproducibility, pooled 2sd, on any single stable Mo isotopic
369	measurement (i.e., one standard-sample-standard measurement) (Table S4). Using this approach,
370	we define an external reproducibility of $\delta^{98/95}Mo$ \pm 0.05 ‰, 2sd, for a single measurement in a
371	given run. This pooled 2sd is then used to calculate the standard error for a given sample given the
372	number, n, of repeat measurements, typically 4 to 6 for unknown samples. This is identical to the
373	2sd, \pm 0.05 ‰, of 35 repeats of W-2A measured over 4 digestions across 4 measuring sessions,
374	and is similar to that reported in Chen et al. (2022) and Hin et al. (2022). Molybdenum isotopic
375	measurements of international reference materials (BHVO-2, BCR-2, and W-2A) agree with
376	literature data (Table S5).
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389 4. Results

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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).

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Values of δ^{238} U show little variation between -0.331 ± 0.019 ‰, 2se, and -0.263 ± 0.028 ‰, 2se, 398 with a concentration weighted average of -0.307 ± 0.032 %, 2sd, (Fig. 3c, table S6). The 399 400 variability is similar to our long-term external reproducibility of samples measured at similar conditions ~ \pm 0.03 ‰ and reflects a near uniform composition of our sample set. The $\delta^{238}U$ 401 402 compositions of the different qualities of glass picked and leached are all, bar one sample, within 403 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 δ^{234} U (Fig. S5b), also has a 404 similar δ^{238} U to other samples (Fig. S5a). Our E-MORB δ^{238} U average is lower than global N-405 406 MORB, which has a concentration weighted average of -0.259 ± 0.041 ‰, 2sd, (Andersen et al., 2015) (Fig. 3c). We also report δ^{238} U data for four N-MORB samples from the Indian ocean basin 407 408 (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 ²³⁴U/²³⁸U data. These data are provided in the supplementary 409 material and were collected following methods in Andersen et al. (2015). Our E-MORB δ^{238} U 410 average is indistinguishable from BSE, δ^{238} U -0.306 ± 0.026 ‰, 2se, (Andersen et al., 2015). 411

Molybdenum isotopic compositions show little variation with $\delta^{98/95}$ Mo ranging between $-0.11 \pm$ 413 414 0.01 ‰, 2se, and -0.19 ± 0.02 ‰, 2se, and are within analytical uncertainty of a concentration 415 weighted average of -0.14 ± 0.04 ‰, 2sd, (Fig. 3d, table S6). The variability, ± 0.04 ‰, 2sd, is 416 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 417 the value reported in Hin et al., (2022) for global E-MORB, -0.12 ± 0.03 ‰, 95 % c.i. The $\delta^{98/95}$ Mo 418 419 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 420 -0.19 ± 0.01 ‰, 95 % c.i. and indistinguishable from BSE, $\delta^{98/95}$ Mo -0.14 ± 0.02 ‰, 95 % c.i. 421 422 (Fig. 3d) (Hin et al., 2022).

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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).

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Fig. 3. (a) U and (b) Mo concentrations and (c) δ^{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).

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458 **5.** Discussion

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- 460 5.1 Potential alteration of U and Mo isotopes
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462 The reductive leaching process removed some U but little Mo, reflecting the presence of 463 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 464 and are all largely within uncertainty in δ^{238} U, $\delta^{98/95}$ Mo, and δ^{234} U (all near secular equilibrium) 465 (Fig. S3, S5). We therefore average all different splits measured for samples into overall δ^{238} U, 466 $\delta^{98/95}$ Mo, and δ^{234} U compositions for each sample (Supplementary material: section 1). We further 467 468 test that our hand-picked MORB glass samples reflect primary magmatic compositions by 469 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 δ^{234} U, and 470 have low δ^{238} U and $\delta^{98/95}$ Mo, ~ -0.69 ‰ and -0.92 ‰ respectively (Siebert et al., 2003; Goto et 471 472 al., 2014). In a binary mixing calculation between our average E-MORB composition and Fe-Mn crusts in δ^{238} U- δ^{234} U and $\delta^{98/95}$ Mo- δ^{234} U space, our samples do not form arrays towards the 473 composition of Fe-Mn crusts (Fig. 4). The minor variability in δ^{238} U and $\delta^{98/95}$ Mo appears 474 475 unrelated to δ^{234} U, and the samples with minor deviations in δ^{234} U from secular equilibrium do not show compositions systematically perturbed towards Fe-Mn crusts in either U or Mo isotopic 476 compositions (Fig. 4). We therefore infer that the δ^{238} U and $\delta^{98/95}$ Mo of the samples represent 477 primary values. 478

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481 5.2 Fractional crystallisation

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 δ^{234} U versus (a) δ^{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, δ^{238} U = -0.307 ‰, δ^{234} U = 0 ‰, [U] = 202 ng g⁻¹, $\delta^{98/95}$ Mo = -0.14 ‰, and [Mo] = 465 ng g⁻¹. Fe-Mn crust, δ^{238} U = -0.69 ‰, δ^{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).

527 5.3 Recycled crustal material in the E-MORB source

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Recent work from Yang et al. (2020) suggests, based on compatible element abundances, that E-529 530 MORB geochemistry is explained by the mixture of low degree partial melts of garnet-531 clinopyroxene pyroxenite (i.e., recycled oceanic crust), with depleted MORB like melts. Melting 532 of this recycled material and mixing with depleted MORB melts generates distinct compositions 533 of element ratios, such as lower Ge/Si, in E-MORB relative to depleted MORB (Yang et al., 2020). 534 The dehydration of subducting slabs during subduction zone processing strips oceanic crust of 535 fluid mobile elements, resulting in high fluid mobile/fluid immobile element ratios in arc lavas. 536 Complementary compositions should then be seen in E-MORB if they contain recycled crustal 537 components, but E-MORB are enriched in both fluid mobile and immobile elements, and have no 538 depletions in elements such as Nb, Tb, and Ti as seen in arc lavas (Niu et al., 2002, their figure 9). 539 To reproduce some of the such E-MORB characteristics in their mixing models, Yang et al. (2020) 540 suggest the recycling of upper continental crust material, that is enriched in elements such as Rb, 541 Ba, and Pb, along with recycled oceanic crust to explain high Rb/Sr, Ba/La, and low Zr/Pb ratios 542 of E-MORB.

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Subducted sediments, a proxy for upper continental crust material, are distinctly enriched in Pb and have low Ce/Pb ratios and high ²⁰⁷Pb/²⁰⁴Pb ratios relative to ²⁰⁶Pb/²⁰⁴Pb (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 increase the ²⁰⁷Pb/²⁰⁴Pb ratio of the upper mantle source to unobserved compositions (Fig. 5). 552 553 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 isotopically light in $\delta^{98/95}$ Mo (Freymuth et al., 2015; Ahmad et al., 2021) (Fig. 6, 7a). However, 556 557 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).

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Other studies also suggest a role for recycled crustal material in the formation of E-MORB sources. 563 564 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 Myr, to allow for radiogenic isotope ratio ingrowth, before being sampled again under MOR's in 567 568 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 EMORB 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).

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Some MORB samples with high "arc-like" $\delta^{98/95}$ Mo, low (La/Sm)_N, and other arc lava 578 geochemical signatures have been linked to mantle sources with substantial contributions from 579 580 fluid-modified mantle wedge components that formed during subduction processes such as 581 enrichments in U and Pb that result in lower Nb/U (33) and Ce/Pb (19) (Chen et al., 2025) than 582 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 583 584 upper mantle MORB sources with recycled crustal components (Chen et al., 2025). However, our 585 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). 586 587 Therefore, the high $\delta^{98/95}$ Mo our E-MORB samples cannot be explained by recycled fluid-modified 588 mantle components.

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590 The simplest interpretation of our new E-MORB data from the North Atlantic Ocean is that they 591 do not contain a recycled, subduction zone processed, crustal component (Fig. 6, 7). Therefore, we 592 argue for a model that does not invoke recycled crustal material to explain the Mo and U elemental 593 and isotopic compositions of E-MORB samples.

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Fig. 5. Mixing model (dashed black line) for ²⁰⁷Pb/²⁰⁴Pb versus Ce/Pb between the 607 depleted MORB mantle and subduction zone processed recycled oceanic crust mixed 608 609 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 610 611 crust and melted subducted sediment composition have been calculated from Stracke 612 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 613 614 obtained by ICP-MS methods).

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Fig. 6. δ²³⁶U and δ³⁶³⁵Mo 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 NMORB.



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.

5.4 Low degree partial melting and mantle metasomatism

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

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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 could exist as volumetrically minor lithologies of low degree melts dispersed as (frozen) dykes or 683 684 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 spots anomalously result in a surface expression of this process due to lithospheric fractures from 694 plate flexure during subduction allowing the melts to ascend (Hirano et al., 2006). 695

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Uranium isotopic data of E-MORB samples requires that the initial low degree melting event 697 happened ≥ 600 Ma, metasomatising the uppermost mantle and freezing in upper mantle 698 699 compositions with enriched trace element compositions and chondritic δ^{238} U. Thus, domains enriched in U are isolated from the evolution of ambient upper mantle to higher δ^{238} U by recycling 700 701 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).

712 Molybdenum isotope data of E-MORB are also compatible with such a model (Chen et al., 2022). 713 but unlike U, the Mo isotopic compositions of E-MORB may be fractionated from its source. In 714 pyroxene, the major upper mantle Mo host, Mo sits in the octahedral M1 site (Leitzke et al., 2017), 715 while in melt Mo⁶⁺ is coordinated tetrahedrally (Holzheid et al., 1994; O'Neill and Eggins, 2002; 716 Farges et al., 2006). While a minor species at modern ambient upper mantle oxygen fugacity, Mo⁴⁺is significantly less incompatible than Mo⁶⁺ in pyroxenes ($D_{Cpx/melt}$ Mo⁴⁺ = ~ 2) (Leitzke et 717 718 al., 2017) and is coordinated octahedrally in both mineral and melt (Farges et al., 2006). Heavier 719 isotopes are concentrated in phases with stiffer and stronger bonds, which form between ions with 720 lower co-ordination number and higher valence state (e.g., Schauble, 2004). Melts are therefore 721 predicted to become isotopically heavier in Mo than residues during partial melting (McCoy-West 722 et al., 2019). The fractionation between Mo isotopes becomes larger at smaller degrees of melting 723 and lower Mo⁶⁺/Mo_T (Mo total) ratios (i.e., more reduced compositions, unlikely for modern 724 oxidised mantle) (Fig. S7a). Chen et al. (2022), following the melting models of McCoy-West et 725 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, Mo⁶⁺/Mo_T = 0.99 (O'Neill and 726 Eggins, 2002), results in a melt composition, $\delta^{98/95}$ Mo = ~ -0.01 ‰ and (La/Sm)_N ~ 5.4, that is 727 728 sufficient to explain the high $\delta^{98/95}$ Mo values and chemical enrichments seen in global E-MORB 729 when mixed with a depleted MORB component (Fig. 9a).

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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
Mo⁶⁺/Mo_T ratios, ~ 0.999 (Fig. 9a). McCoy-West et al. (2019) used the 'ionic model' and literature

bond length data to derive a fractionation factor between ⁹⁸Mo and ⁹⁵Mo during mantle partial melting, $\alpha^{98/95}$ Mo_{Melt-Silicate}, ~ 0.99977. Such a value has not been directly, experimentally verified and notably predicts larger fractionation than for another redox sensitive system Cr (e.g., Jerram et al., 2022) for example. We note that our data imply a smaller melt-silicate fractionation of Mo isotopes, and we illustrate this using $\alpha^{98/95}$ Mo_{Melt-Silicate} of 0.9999 (Fig. S7b, 9b). Experimental work is required to assess if this empirical reassessment of the fractionation factor is justified.

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Our model fit to $\delta^{98/95}$ Mo data is further improved given inferences from δ^{238} U compositions of E-741 742 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. (2022) show that at least ~ 1 to 1.4 Gyr of oceanic crust recycling is needed to lower the $\delta^{98/95}$ Mo 745 746 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 various ages ≥ 600 Ma can help explain the variably elevated $\delta^{98/95}$ Mo of global E-MORB. We 751 752 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 member composition of $\delta^{98/95}$ Mo = ~ -0.1 ‰ and (La/Sm)_N ~ 5.4 at Mo⁶⁺/Mo_T = 0.99. 754

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


824	Fig. 9. Non-modal, batch melting modelling of $(La/Sm)_N$ and Mo isotopic composition of melt.
825	Mixing lines are shown between a depleted MORB (represented by 20 % melting of DMM) and
826	an enriched low degree melt end member generated by 0.2 % melting of DMM at different mantle
827	fO_2 (Mo ⁶⁺ /Mo _T = 0.999 to 0.95). Symbols and sources for data are the same as used in figure 2.
828	Model calculations follow McCoy-West et al. (2019) and Chen et al. (2022). Parameters used are
829	given in table S9. White diamonds indicate mixing proportions. (a) $\alpha^{98/95}$ Mo _{Melt-Silicate} ~ 0.99977,
830	Depleted MORB $\delta^{98/95}$ Mo = -0.2 ‰ (b) $\alpha^{98/95}$ Mo _{Melt-Silicate} ~ 0.9999, Depleted MORB $\delta^{98/95}$ Mo =
831	-0.185 ‰.
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A set of hand-picked E-MORB glasses from the Northern mid-Atlantic ridge picked to 849 850 different degrees of stringency all have ²³⁴U/²³⁸U ratios close to secular equilibrium, indicating 851 that samples likely reflect primary U and Mo isotopic composition. These E-MORB samples show a limited range of δ^{238} U and $\delta^{98/95}$ Mo from ~ -0.33 to -0.26 ‰ and -0.19 to -0.11 ‰ respectively 852 853 and are both, on average, indistinguishable from the bulk silicate Earth (chondritic) U and Mo 854 isotopic compositions. These E-MORB therefore have contrasting sources to modern N-MORB, 855 which are super-chondritic in U and sub-chondritic in Mo isotope ratios, thought to be due to the 856 recycling of modern day like subduction zone processed altered oceanic crust (Andersen et al., 857 2015; Hin et al., 2022). Our E-MORB data are inconsistent with models that explain their 858 enrichment with recycled oceanic crust and or recycled sediment (e.g., Allègre and Turcotte, 1986; 859 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 860 861 oceanic lithospheric mantle, metasomatised by small degree asthenosphere melts (Niu et al., 2002; 862 Chen et al., 2022; Guo et al., 2023). This process effectively isolates the composition of the upper 863 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 864 865 low degree partial melting) show that this initial low degree melting event occurred in an upper 866 mantle with a chondritic δ^{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 867 868 ocean oxygenation (Andersen et al., 2015), providing a minimum bound on the antiquity of the E-869 MORB source. Molybdenum isotopic compositions of global E-MORB are compatible with such

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

895	Joel B. Rodney: Data curation, formal analysis, investigation, methodology, validation,
896	visualisation, writing – original draft. Morten B. Andersen: Supervision, project administration,
897	funding acquisition, conceptualization, methodology, writing - review and editing. Bramley J.
898	Murton: Resources, writing - review and editing. Tim Elliott: Supervision, project
899	administration, funding acquisition, conceptualization, writing – review and editing.
900	
901	Declaration of competing interest
902	
903	The authors declare that they have no known competing financial interests or personal
904	relationships that could have appeared to influence the work reported in this paper.
905	
906	Data availability
907	
908	Data are available through Mendeley data at:
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910	
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924	Appendix A. Supplementary Material
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926	Supplementary Material related to this article can be found online at:
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1147	Constraints from uranium and molybdenum isotope ratios on the origin of enriched mid-
1148	ocean ridge basalts
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1177	Supplementary Information
1178	Section 1: MORB glass picking and leaching
1179	Section 2: Full detailed method for U and Mo isotopic analysis
1180	Section 3: Supplementary figures and tables
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1201 <u>Section 1:</u>

1202 MORB glass picking and leaching

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Hand specimen samples of E-MORB were crushed and processed to ~ 1000 μ m size glass chips. Glass chips were further crushed using an agate pestle and mortar, and sieved, aiming for a size of ~ 600 μ m. 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).

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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.

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A reliable check of sample alteration is given by $^{234}U/^{238}U$ activity ratios. If unaltered by any recent 1220 seawater alteration (that would likely affect U compositions), the ²³⁸U decay chain will be in 1221 secular equilibrium, and so the activity ratio of (²³⁴U/²³⁸U), will be at unity. Seawater has 1222 $(^{234}U/^{238}U) \sim 1.14$ (Kipp et al., 2022) and so elevated $(^{234}U/^{238}U)$ in glass samples indicates addition 1223 of seawater U onto Fe/Mn coatings, which could also indicate adsorption of isotopically distinctive 1224 1225 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. 1226 Samples of quality A is those that are most optically clear and devoid of all alteration and coatings 1227 1228 (Fig. S1). Samples of quality B is less optically clear, but still mostly devoid of alteration and 1229 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 1230 1231 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.

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1237 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 1238 and rinsed thrice with >18 M Ω .cm water (hereafter milli-Q water). Samples were shaken 1239 1240 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 1241 al., 2007). The leachate was collected, and samples rinsed thrice with 10 ml milli-Q water that was 1242 also added to the collected leachate solution. Collected leachate solutions were analysed in 0.3 M 1243 HNO₃ for elemental concentrations using an ICP-MS Element2, at the University of Bristol. As 1244 1245 noted by Andersen et al. (2015) and Hin et al. (2022) leaching can result in small amounts of glass 1246 dissolution, and thus U and Mo loss. Ratios of the concentrations of elements that absorb to Fe-1247 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. 1248

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1250 Uranium and Mo are removed during the leaching process (Fig. S2), and it is possible that the 1251 removed U and Mo is either from secondary coatings (Fe-Mn coatings) or from the dissolution of 1252 the MORB glass. Three leachates from MORB glass sample JC24-82-21, glass qualities A, B and 1253 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 % 1254 1255 or less lost in leaching steps 2 and 3 (Fig. S2). This was associated with systematic changes in 1256 U/Th of the leachates (Fig. S3). Comparing U to less mobile elements such as Th (which is only 1257 likely to be released during glass dissolution) shows the effects of the leaching procedure (Fig. 1258 S3). In the first leachate (sample JC24-82-21) U/Th are elevated, e.g., U/Th >4, and likely reflects 1259 the release of secondary coating hosted U (Fig. S3). In the second and third leachates, these ratios 1260 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 1261 1262 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.

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1267 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 1268 consistently <5 % and is relatively consistent between leaching steps (Fig. S2). This suggests very 1269 little secondary phase hosted Mo, which should be removed in the first leach (as for U), and 1270 1271 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 1272 procedure does not fractionate U or Mo isotopes of fresh glass, from comparing a leached and un-1273 leached BHVO glass sample. Therefore, even if some of U and Mo loss is from glass dissolution 1274 rather than the dissolution of secondary coatings, isotopic compositions should not be perturbed. 1275

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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).

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The δ^{234} U compositions of different qualities of glass picked and leached for each sample, A, B 1285 1286 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 δ^{238} U compositions of the different qualities 1287 of glass picked and leached are all, bar one sample, within analytical uncertainty (Fig. S5a) and 1288 bar JC24-89-13, are also within uncertainty of $\delta^{234}U = 0$ (Fig. S5b). Sample JC24-89-13, which is 1289 only elevated above 0 by 2.5 ‰ in δ^{234} U (Fig. S5b), also has a similar δ^{238} U to other samples (Fig. 1290 S5a). The $\delta^{98/95}$ Mo compositions of different qualities of MORB glass are also all within analytical 1291 uncertainty (Fig. S5c). For samples with multiple splits, the fact that different qualities of glass are 1292 1293 all within secular equilibrium and within uncertainty of one another has two important benefits:

1294	firstly, it allows us to assess the level of MORB glass quality needed to ensure minimal secondary
1295	alteration, i.e., quality C; secondly, we can average all the splits (A, B, and C) for individual
1296	samples for overall δ^{238} U, δ^{234} U, and $\delta^{98/95}$ Mo compositions (Table S6, S7). This approach is
1297	reasonable given that the majority of samples with different splits (A, B, and C) have δ^{238} U and
1298	$\delta^{98/95}$ Mo compositions largely indistinguishable from one another (Fig. S5a, c).
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- 1326 <u>Section 2:</u>
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1327 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).
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1333 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 1334 1335 ml of acid using a mixture of 5:1 15.6 M HNO₃ 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-1336 1337 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 1338 prior to dissolution. The double spike was added to samples according to their U concentrations 1339 aiming for a ${}^{236}\text{U}/{}^{235}\text{U}$ ratio of 5. Samples were also spiked with a ${}^{97}\text{Mo} - {}^{100}\text{Mo}$ double spike, 1340 prepared by the Bristol Isotope group, with a ⁹⁷Mo/⁹⁵Mo ratio of 47.58 and ¹⁰⁰Mo/⁹⁵Mo ratio of 1341 58.32, aiming for a natural Mo-double spike Mo proportion of 0.5. For measurement of Th, U, and 1342 1343 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 1344 1345 concentrations of U, Th, and Mo of reference materials measured on the Element2 are in good agreement with literature values (Table S2). 1346

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Samples were first processed for U isotopic analysis. Samples were loaded in 40 ml of 1.5 M 1348 HNO₃ onto 1 ml of TRU resin (100 – 150 mesh) in commercially available Bio-Rad Poly-Prep 1349 1350 columns. Matrix was eluted in 30 ml of 1.5 M HNO₃, 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 1351 chemistry. Collected U fractions were dried and fluxed in a 1 ml 50:50 15.6 M HNO3: 30 % Romil 1352 SpA H₂O₂ mixture to eliminate any organic material that may have leached off resin into samples. 1353 Samples were loaded in 5 ml 3 M HNO₃ onto 0.5 ml of UTEVA resin (100 – 150 mesh), for Th 1354 and U separation, with 10 ml of 3 M HNO₃ washed through to elute any residual matrix, before 1355 1356 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

1357 M HCl. Again, any potential organic material was eliminated from samples. Final U collections 1358 were then dried and re-dissolved in a requisite amount of 0.2 M HCl (aiming for U concentration 1359 of 100 - 300 ng g⁻¹) for isotopic analysis. This procedure achieved efficient removal of Th and 1360 Th/U ratios during isotopic analysis were typically <0.005.

1361

Uranium isotope compositions were measured on a ThermoFinnigan Neptune MC-ICP-MS (serial 1362 no. 1002) at the Bristol Isotope group in low mass resolution (M/ Δ M ~ 2000, 5 to 95 % peak height 1363 definition), using the setup detailed in Andersen et al. (2015). Samples were introduced to the 1364 plasma using a ~ 40 ul min⁻¹ micro-concentric PFA nebuliser connected to a Cetac Aridus (1st 1365 generation) desolvating system. A standard sample cone plus X-skimmer cone set up was used. 1366 Masses 232 (²³²Th), 233 (²³³U), 234 (²³⁴U), 235 (²³⁵U), 236 (²³⁶U), and 238 (²³⁸U) were collected 1367 in faraday cups, with most cups connected to feedback amplifiers with $10^{11} \Omega$ resistors, apart from 1368 234 which was connected to a $10^{13} \Omega$ resistor and 238 which was connected to a $10^{10} \Omega$ resistor. 1369 Before each session instrumental baselines were measured and amplifier gains intercalibrated. 1370 1371 Measurements consisted of 80 s of solution uptake to allow the ion beams to stabilise. Sample and 1372 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 1373 time. Individual sample and standard measurements consisted of 80 cycles each, with 4.194 s 1374 integration time. Each sample was preceded and followed by a measurement of the double-spiked 1375 1376 (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 1377 consumption between ~ 30 to 80 ng per measurement. Procedural blanks were <30 pg U, an 1378 insignificant amount compared to amount of U consumed per measurement. Ion beam intensities 1379 1380 were corrected for low mass tailings of ion beams and high mass plus hydride tailings of ion beams 1381 following Andersen et al. (2015). Each solution was corrected for solution blank and intensities 1382 re-calculated.

1383

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 ²³⁸U/²³⁵U and ²³⁴U/²³⁸U are reported in δ notation with $\delta^{238}U = [(^{238}U/^{235}U_{Sample} / ^{238}U/^{235}U_{CRM-145}) - 1]$ and $\delta^{234}U = [(^{234}U/^{238}U_{Sample} / (^{234}U/^{238}U_{CRM-145} / (1-0.0386)) - 1]$. 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).

1394

Long term external reproducibility at various measured U intensities has been estimated using 1395 aliquots of the well characterised reference material BHVO-2 measured during different analytical 1396 sessions. The external reproducibility of δ^{238} U and δ^{234} U for BHVO-2 at various intensities (e.g., 1397 238 U = 200 to 1000 pA) ranges from ± 0.09 to 0.03 ‰, 2sd, and ± 4 to 0.9 ‰, 2sd, respectively. 1398 1399 The external reproducibility of unknown samples has been determined from the long-term external 1400 reproducibility of BHVO-2 measured at various intensities. As samples were measured at varying intensities ($\sim ^{238}$ U = 200 to 1000 pA) depending on the U concentration, BHVO-2 was also ran at 1401 varying intensities. Repeat measurements of BHVO-2 were then used to construct error curves 1402 1403 using the two standard deviations of measurements in ranges of intensities. A power law was fit to the data for δ^{238} U and δ^{234} U at the different intensities and used to approximate errors for unknown 1404 1405 samples. This relationship was then used for samples of a given concentration, that corresponds to 1406 a given intensity, from which an approximate 2sd could be calculated and an external 2se 1407 calculated based on the number of repeats (Fig. S4).

1408

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.

1414

1415 Collected Mo fractions from the TRU resin U chemistry (40 ml of sample load and first 10 ml of
1416 1.5 M HNO₃ wash) were dried and dissolved for chemistry using Eichrom AG 1-X8 100 – 200
1417 mesh anionic resin, following Willbold et al. (2016). Samples were dissolved in 22.5 ml 3 M HCl
1418 and 1.25 ml 6 M HCl ready for column chemistry using 2 ml of Eichrom AG 1-X8 100 – 200 mesh

1419 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 Fe³⁺ 1420 to Fe²⁺ which is identified by a colour change of samples from yellow to green/colourless. Matrix 1421 was eluted using 6 ml 3 M HCl, 26 ml 0.5 M HCl- 0.5 % H₂O₂, 20 ml 1 M HF and 6 ml milli-Q 1422 1423 water, before collection of Mo in 24 ml of 1 M HCl. Collected Mo fractions were dried and fluxed 1424 in a 1 ml 50:50 15.6 M HNO₃: 30 % Romil SpA H₂O₂ mixture to eliminate any organic material 1425 that may have leached off resin into samples. Samples were dried and re-dissolved in the requisite amount of 0.4 M HNO₃ – 0.4 M HF for a Mo concertation of 200 ng g^{-1} for isotopic analysis. 1426

1427

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).

1433

1434 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/ Δ M ~ 1600, 5 to 95 % 1435 peak height definition). Samples were introduced to the plasma using a ~ 40 ul min⁻¹ micro-1436 concentric PFA nebuliser connected to a Cetac Aridus (1st generation) desolvating system. 1437 1438 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 (91Zr), 92 1439 (⁹²Mo), 95 (⁹⁵Mo), 96 (⁹⁶Mo), 97 (⁹⁷Mo), 98 (⁹⁸Mo), 99 (⁹⁹Ru), 100 (¹⁰⁰Mo), and 101(¹⁰¹Ru) were 1440 collected in faraday cups L4 to H4 respectively. All cups would ideally be connected to an 1441 amplifier with a $10^{11} \Omega$ resistor, however due to the limited number of $10^{11} \Omega$ resistors available 1442 on the Neptune, 91 Zr, 92 Mo, and 101 Ru were connected to $10^{10} \Omega$ resistors. This should not be an 1443 issue as ⁹¹Zr and ¹⁰¹Ru are interferences and removed in chemistry and ⁹²Mo is not used in the 1444 double spike inversion for the calculation of ⁹⁸Mo/⁹⁵Mo. Before each session instrumental 1445 1446 baselines were measured and amplifier gains intercalibrated. Measurements consisted of 80 s of 1447 solution uptake to allow the ion beams to stabilise. Sample and standard measurements were preceded by 90 s of washing with 0.4 M HNO₃ - 0.4 M HF, followed an on-peak baseline 1448 measurement of reagent, 0.4 M HNO₃ – 0.4 M HF, blank for 30 cycles of 4.194 s integration time. 1449

1450 Individual sample and standard measurements consisted of 30 cycles of 4.194 s integration time, 1451 consuming \sim 30 ng Mo per measurement for solutions measured at 200 ng g⁻¹. Each sample was 1452 preceded and followed by a measurement of the double-spiked (with a double spike proportion 1453 similar to samples) standard NIST SRM3134. Each solution was corrected for solution blank and 1454 intensities re-calculated.

1455

Measurements were internally normalised using a double spike inversion using ⁹⁵Mo, ⁹⁷Mo, ⁹⁸Mo, 1456 and ¹⁰⁰Mo, and ⁹⁸Mo/⁹⁵Mo ratios calculated. Samples were externally normalised to the bracketing 1457 standard and $\delta^{98/95}$ Mo calculated ($\delta^{98/95}$ Mo = [(98 Mo/ 95 Mo_{Sample}/ 98 Mo/ 95 Mo_{NISTSRM3134}) - 1]). 1458 Ruthenium has interferences with ⁹⁸Mo (⁹⁸Ru) and ¹⁰⁰Mo (¹⁰⁰Ru). Therefore, ⁹⁹Ru and ¹⁰¹Ru were 1459 1460 monitored to allow for corrections to be applied to ⁹⁸Mo and ¹⁰⁰Mo. Ruthenium doping experiments (Chen et al., 2019) show that corrections using ⁹⁹Ru can accurately correct Mo data. 1461 However, there is the potential for overcorrection due to species such as ⁶⁴Zn³⁵Cl and ⁴⁰Ar₂¹⁹F 1462 giving signals at mass 99 (Chen et al., 2019). Therefore, ¹⁰¹Ru is also monitored and can be used 1463 for correction. Data was corrected using both ⁹⁹Ru and ¹⁰¹Ru, and when compared, both methods 1464 give the same answer within uncertainty. If the total Ru correction was ever over 0.1 % in $\delta^{98/95}$ Mo 1465 the datum was rejected (no data in this study was rejected). 1466

1467

1468 We take a homoscedastic approach to determine our external reproducibility, pooled 2sd, on any 1469 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 ± 0.05 ‰, 2sd, for a 1470 1471 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. 1472 1473 This is identical to the 2sd, ± 0.05 ‰, of 35 repeats of W-2A measured over 4 digestions across 4 1474 measuring sessions, and is similar to that reported in Chen et al. (2022) and Hin et al. (2022). 1475 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 1476 1477 standards (CPI and GUG-11). A full list of data for reference materials is provided in table S5.

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- 1480



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. S4. Error curves for BHVO-2 constructed using the external 2sd of repeat measurements of BHVO-2 at various intensity brackets, $\geq 20 < 30$ V, $\geq 30 < 40$ V, ≥ 40 <50 V, $\geq 50 < 60$ V, $\geq 60 < 70$ V, $\geq 70 < 80$ V, $\geq 80 < 90$ 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 δ^{238} U measurements, (b) Error curve for δ^{234} U measurements.

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- 1572
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1658 Fig. S6. Variations of E-MORB samples in this study of (a) U concentration, (b) Mo concentration 1659 (c) δ^{238} U and (d) $\delta^{98/95}$ Mo with MgO as a tracer of magmatic differentiation. Linear variation in 1660 (e) the U and Th concentration and (f) Mo and Ce concentration of E-MORB samples, trend line 1661 is shown as a dashed line with its associated R² value.

- 1662
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- 1666



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 (Mo⁶⁺/Mo_T = 0.999 to 0.95) and start from DMM compositions of $\delta^{98/95}Mo = -0.2$ ‰ with $\alpha^{98/95}Mo_{Melt-Silicate}$ of (a) ~ 0.99977 and (b) 0.9999.



Fig. S8. (a, c, e) δ^{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).

Decien	Sample		Latitude	Longitude	Depth	Age
Region			(°N)	(°W)	(m)	(Ma)
		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
MAR 45 °N	JC24	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
		D15-1	13.07	44.09	4496	0.5 – 1
		D16-17	13.03	44.84	4820	0.5 - 1
MAR 13 °N	JC7	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

1727 Table S1. E-MORB samples used in this study from the Northern Mid-Atlantic ridge,

1728 data for 13 $^{\circ}N$ 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.

- 1745 Reference values are from the USGS data sheets.

1756	Sample	δ ²³⁸ U (‰)	2sd	δ ²³⁴ U (‰)	2sd	$[U] (ng g^{-1})^*$	N.M	N.S
1757		-0.306	0.044	-0.1	1.8	419	145	25
1758	BHVO-2	-0.314	0.020	0.9	2.4	386	16	8
1759		-0.255	0.036	0.6	1.4	1687	27	4
1760	BCR-2	-0.297	0.020	1.1	1.0	1671	1	1
1761	BIR	-0.277	0.189	-5.1	8.6	8	6	3
1762		-0.285	0.020	-0.6	1.0	8	1	1
1763		-0.048	0.045	-0.2	1.8	7995	30	3
1764	CZ-1	-0.053	0.030	0.1	2.7		15	2
1765		-0.460	0.050	0.3	2.0	185	10	2
1766	GUG-11	-0.419	0.030	-0.4	2.0	143	1	1
1767		-0.289	0.063	0.1	2.6	70	26	10
1768	IT-3a	-0.296	0.020	-0.3	1.7	62	3	1
1769		-0.319	0.049	-0.2	2.0	2356	83	3
1770	LP-45d	-0.300	0.020	-0.3	4.5	2119	5	2
1771	Uraninite	-0.548	0.053	_1.5	2.1	11985	51	3
1772	W-2A	_0 291	0.036	2.0	1.4	497	37	7
1773	··· 211	0.271	0.050	2.0	1.7		51	/

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 \times (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
ЈС24-83-7-В	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
ЈС24-93-35-В	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
ЈС24-79-22-С	0.02	5	0.0003
JC24-90-13-A	0.05	4	0.0018
JC24-90-13-B	0.04	5	0.0016
ЈС24-90-13-С	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		

1787Table S4. Homoscedastic approach for calculating external uncertainty on Mo1788isotopic compositions. Variance is calculated as Variance = $2sd^2/4$ and pooled1789variance as the sum of variance × (N-1) / sum of N-1. Pooled 2sd is calculated as1790pooled 2sd = $2 \times \sqrt{Pooled}$ variance.
1817							
1818	Sample	δ ^{98/95} Mo (‰)	2sd	2se	[Mo] ppb*	N.M	N.S
1819	CDI	-0.27	0.05			56	
1820	CPI	-0.23	0.04			42	
1821		-0.06	0.05		4207	20	3
1822	BHVO-2	-0.08	0.04		3570	6	
1823		-0.05	0.05		417	35	4
1824	W-2A	-0.05	0.05		460	9	
1825		0.07	0.05		946	19	1
1826	GUG11	0.049		0.016	1003		
1827		0.00	0.05		259873	8	1
1828	BCR-2	_0.03	0.04		236000	3	-
1829		0.00	0.07		200000	5	

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.

a 1		U		δ ²³⁸ U	2	$\delta^{234}U$	2		Mo	C M	δ ^{98/95} Mo			N O/TO	(1 (0)
Sample	Sample	$(ng g^{-1})$	Th/U	(‰)	2se	(‰)	2se	N	$(ng g^{-1})$	Ce/Mo	(‰)	2se	N	K ₂ O/11O ₂	(La/Sm) _N
	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
450 NI	83-7	185	2.79	-0.331	0.019	0.1	0.7	4	420		-0.12	0.01	12	0.16	
45° N JC24	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
	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
12º N	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
I3° N JC7	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			

1849Table S6. U and Mo concentration and isotopic data for E-MORB samples analysed in this work1850(mean compositions from multiple qualities of glass for each sample), along with select element1851ratios. Error is the 2se and N refers to the total number of repeat measurements. For the overall1852Atlantic E-MORB average, isotopic compositions are calculated as concentrated weighted1853averages, and the error calculated as the 2sd over the square root of total number of samples1854measured.

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Sample		Mass picked (mg)	U (ng g ⁻¹)	δ ²³⁸ U (‰)	2se	δ ²³⁴ U (‰)	2se	Ν	Mo (ng g ⁻¹)	δ ^{98/95} Mo (‰)	2se	Ν
	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	000	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
	D10-17-C	1151	212	-0.311	0.022	-0.2	0.9	2	545	-0.14	0.02	0
	D10-17	1000	213	-0.304	0.013	-0.2	0.5	2	720	-0.15	0.01	18
13° N JC7	D24-/-A	402	202	-0.500	0.022	0.4	1.4	3	129	-0.16	0.02	5
	D24-/-B	472	215	-0.501	0.041	0.0	1.0	1	733	-0.21	0.02	5 10
	25.2.1.0	017	200	0.204	0.019	0.0	1.0	*	675	-0.15	0.02	5
	23-3.1-C	21/ 822	252	-0.290	0.030	0.0	1.2	2	670	-0.13	0.02	5
	23-3.1-B	022	200 252	-0.515	0.028	-0.2	1.1	5 6	672	-0.14	0.02	5 10
	23-3.1 D26.2 A	1021	101	-0.304	0.020	-0.1	1.5	1	201	-0.13	0.02	5
	D26-3-A	1031	101	-0.277	0.03/	0.5	1.5	1	201	-0.14	0.02	5 5
	D20-3-B	1019	100	-0.299	0.039	0.5	1.5	1	1/0	-0.14	0.02	5 10
	D20-3		101	-0.268	0.027	0.4	1.1	4	109	-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	5.8555ª	64.21	15.53 ^b
Mobility (%)	81 ^a	51ª		
GLOSS-II	21.2°	57.6°	2.7	15.694°
GLOSS-II melt	18.444 ^a	44.352 ^a	2.4	15.694°
Mobility Sediment-Melt (%)	13 ^a	23 ^a		
DMM	0.0232 ^d	0.772 ^d	33.3	15.515 ^e

Table S8. Compositions used in the mixing models shown in figure 5. a - Stracke et al. (2003). b - 801C average from Hauff et al. (2003). c - Plank (2014). d - Salters and Stracke (2004). e - Gale et al. (2013). The composition of recycled oceanic crust and GLOSS-II melt have been calculated using the element mobility % given in 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	
$\mathrm{D}_{\mathrm{Mo}}^{6+}$	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		
Мо	650	1041		

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

1881	is assumed to be \approx to Ce and is taken from Salters and Stracke (2004). Modal % and Melt modes
1882	% are from Walter (1998). Depleted MORB Mantle (DMM) compositions are from Salters and
1883	Stracke (2004) and isotopic compositions are from Hin et al. (2022). Force constants for Mo in
1884	mineral and melt are from McCoy-West et al. (2019). Non-modal batch melting models and Mo
1885	isotopic fractionation models follow McCoy-West et al. (2019) and Chen et al. (2022).
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