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Chondritic osmium isotope composition of early Earth mantle

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Abstract

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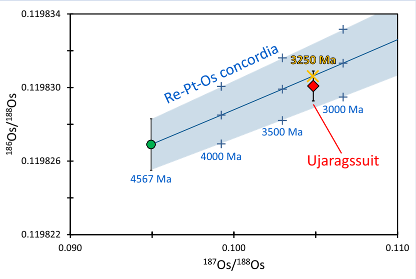

The Ujaragssuit Intrusion, North Atlantic Craton (NAC), Greenland, is thought to host the oldest chromitites (>3.8 Ga) on Earth, showing evidence of both Hadean mantle depletion events and nucleosynthetic isotopic heterogeneities. We set out to verify the age of the intrusion and identify the Os isotope composition of the Ujaragssuit mantle source. Here, we show that the only minimum age constraint is 2970 ± 8 Ma, provided by cross-cutting leucogranites. Concordant Re-Pt-Os isotope ages are consistent with formation of the intrusion from a chondritic primitive mantle source at 3246 ± 120 Ma; mean Pt-depletion ages of 3437 ± 587 Ma offer no direct evidence for Hadean mantle depletion. No nucleosynthetic Os isotopic anomalies could be identified, consistent with large scale Os homogeneity in the presolar nebular. The new 'young' age for Ujaragssuit means that nucleosynthetic anomalies occur repeatedly between ~3.8 and >3.0 Ga in the NAC, suggesting its unique mantle source was repeatedly tapped over ~600 Myr without significant mixing with the rest of Earth's mantle.

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Introduction

The Itsaq Gneiss Complex, North Atlantic Craton (NAC), Greenland, is among the largest and best preserved tracts of Eoarchaean crust on Earth (Nutman *et al.*, 1996). The Isua Supracrustal Belt and area immediately to the south has been of particular interest due to less intense Neoarchaean metamorphism (Friend and Nutman, 2019), the preservation of supracrustal sequences, and an abundance of mantle-derived lavas and ultramafic bodies. This has made the area an attractive target for studies of long lived (Bennett *et al.*, 1993, 2002; Coggon *et al.*, 2013; Waterton *et al.*, 2022), short lived (Bennett *et al.*, 2007; Willbold *et al.*, 2011), stable (Creech *et al.*, 2017; Xu *et al.*, 2023) and nucleosynthetic (Fischer-Gödde *et al.*, 2020) isotope systematics, to identify both ancient mantle differentiation events and primordial heterogeneities from Earth's accretion.

Among the ultramafic bodies in this region, the stratiform chromitite-bearing Ujaragssuit Nunât layered body (hereafter the Ujaragssuit Intrusion) was recognised as the oldest chromitite on Earth (Chadwick and Crewe, 1986), with a minimum age of >3.8 Ga indicated by its host orthogneisses (Nutman *et al.*, 1996). High concentrations of highly siderophile elements (HSEs) in the chromitites make them ideal for study using Re-Pt-Os (Bennett *et al.*, 2002; Rollinson *et al.*, 2002; Coggon *et al.*, 2013, 2015) and Ru isotopes (Fischer-Gödde *et al.*, 2020). These studies identified that the Ujaragssuit Intrusion records evidence of Hadean mantle depletion (Coggon *et al.*, 2013) and was derived from mantle deficient in chondrite relative to modern mantle (Fischer-Gödde *et al.*, 2020). In this study, we set out to verify the age of the Ujaragssuit Intrusion and to identify the Os isotope composition of the Ujaragssuit source.

Results

Field observations and U-Pb zircon dating. Detailed field mapping of the Ujaragssuit Intrusion was supplemented with aerial drone photography to produce a high resolution map (Fig. 1). The Ujaragssuit Intrusion is a boudinaged peridotite-dominated body, comprising major lenses connected by thin necks, reminiscent of pinch and swell structures in sheared mid-crustal domains. This, along with gneissic foliation that wraps the intrusion, suggests that the ultramafic rocks acted as a competent body around which the gneisses deformed (Gardner *et al.*, 2015). We found no evidence of large scale faulting within or around the intrusion. Chromitites are abundant in the northern portion of the intrusion, corresponding to its base (Rollinson *et al.*, 2002). The majority are stratiform chromitites with 1–20 cm thick layers. Six massive chromitite pods are also observed, ranging from ~0.5 × 1 m to ~3 × 7 m (Table S-1). Although all the chromitites have experienced alteration and metasomatism during high grade metamorphism, we divide these into 'fresh' and 'altered' chromitites based on field characteristics (Supplementary Information).

Zircon LA-ICP-MS U-Pb dating yields a poorly constrained crystallisation age of ~3.8 Ga for orthogneisses adjacent to the intrusion (Fig. 1; zircon U-Pb methods, data and age

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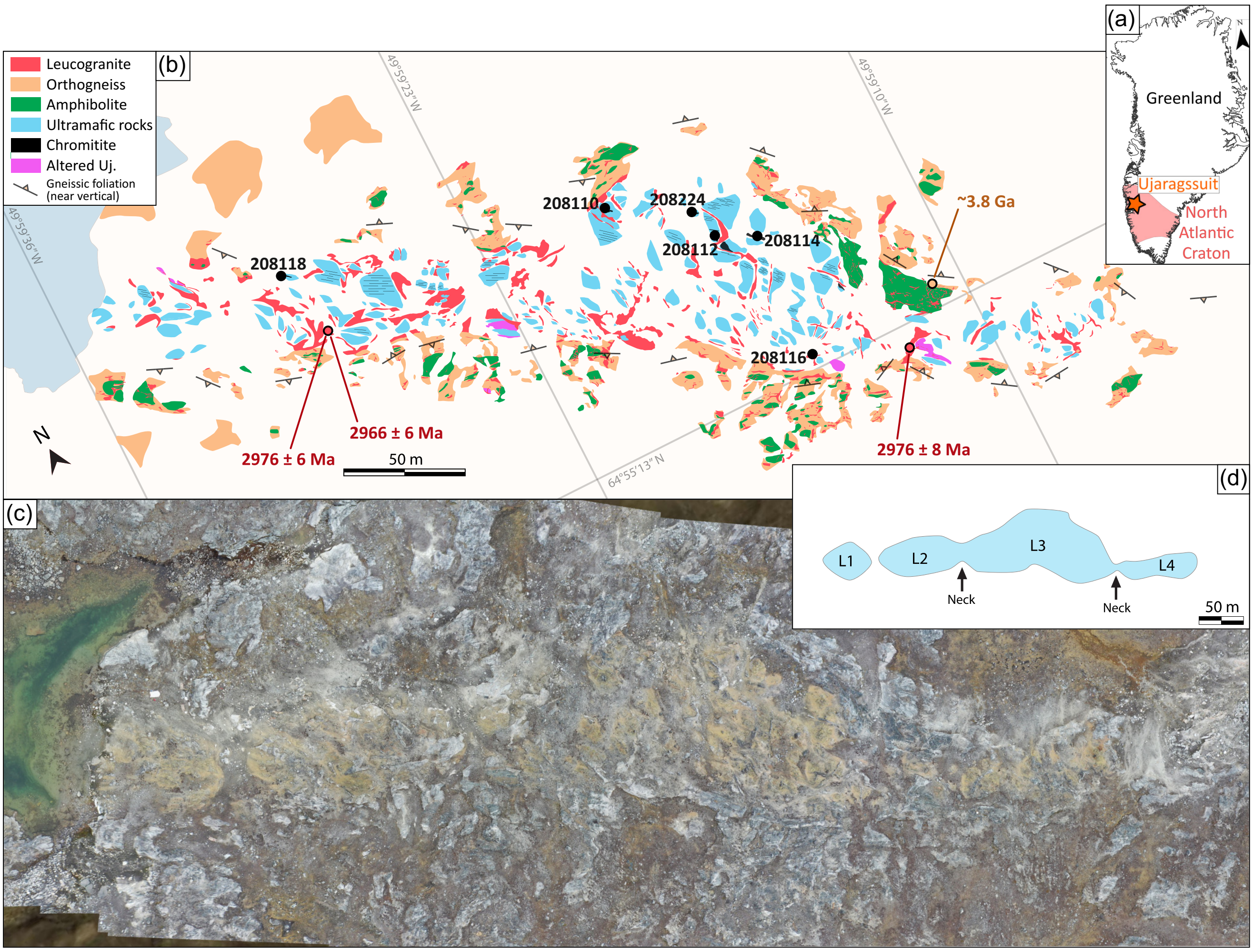

Figure 1 **(a)** Location of the Ujaragssuit Intrusion. **(b)** Detailed map of the Ujaragssuit Intrusion, showing locations of analysed chromitite and granitoids samples, including granitoid zircon U-Pb ages. 'Altered Uj.' refers to the most altered portions of the Ujaragssuit Intrusion, including corundum-bearing amphibolites and reaction zones. **(c)** High resolution aerial photograph of the Ujaragssuit Intrusion. **(d)** Outline of the Ujaragssuit Intrusion, showing major lenses connected by thin 'necks'.

interpretations are provided in Supplementary Information and Table S-2). Two orthogneiss samples collected ~4.5 km west of the intrusion yield more precise igneous ages of 3849 ± 6 Ma and 3842 ± 7 Ma. Though slightly older than previous orthogneiss ages near Ujaragssuit (Nutman *et al.*, 1996), our data confirms that the intrusion's host orthogneisses are >3.8 Ga. However, we could not find any exposed contacts between the orthogneisses and ultramafic rocks, so this does not provide a minimum age constraint on the Ujaragssuit Intrusion. Instead, the entire intrusion is 'sheathed' and cross-cut by anastomosing sheets of leucogranite. Three of these leucogranites yield much younger crystallisation ages of 2966 ± 6 Ma, 2976 ± 6 Ma, and 2966 ± 8 Ma, consistent with previously determined ages of ~2.97–2.95 Ga (Sawada *et al.*, 2023). These coincide with a population of metamorphic zircon present in all three orthogneiss samples at ~2.97 Ga, as well as growth of metamorphic zircon within the Ujaragssuit chromitites themselves (Sawada *et al.*, 2023). These ages reflect regional metamorphism in this part of the Itsaq Gneiss Complex (Friend and Nutman, 2019), driving formation of the leucogranites *via* intracrustal melting. We also identify ~3.00–2.99 Ga metamorphic zircon ages in the orthogneisses, which could suggest that the ~2.97 Ga regional metamorphic event was more protracted than previously recognised. A second event caused further growth of metamorphic zircon at 2693 ± 10 Ma in one of the leucogranites.

To summarise, the only robust age constraint on the Ujaragssuit Intrusion is that it is >2.97 Ga, the age of both cross-cutting leucogranites and metamorphic zircon within Ujaragssuit chromitites (Sawada *et al.*, 2023). The average crystallisation age of the leucogranites and of ~2.97 Ga metamorphic zircon yields a minimum age of 2970 ± 8 Ma for the Ujaragssuit Intrusion.

Highly siderophile element abundances and Re-Os isotopic data. All HSE and Re-Pt-Os isotope methods, data and calculations are given in Supplementary Information and Tables S-3 and S-4. Radiogenic Os isotope results are presented as Re- and Pt- model ages assuming chondritic evolution (Walker *et al.*, 2002) or primitive mantle models (Meisel *et al.*, 2001; Brandon *et al.*, 2006) based on chondrite; we address this assumption in the Discussion. The analysed massive chromitite samples have high Os contents (96–200 ppb), low Pt (0.72–4.5 ppb) and low Re (one altered sample has 82 ppt Re, the remaining analyses are below the 6.9 pg limit of detection). Their high Os concentrations reflect high modal chromite contents (Bennett *et al.*, 2002; Coggon *et al.*, 2015) and, combined with low $^{187}Re/^{188}Os$ ($<4.1 \times 10^{-3}$) and $^{190}Pt/^{188}Os$ (7.1×10^{-6} to 4.4×10^{-5}), make Re- (T_{RD}) and Pt-depletion (T_{DA}) model ages resistant to modification through crustal assimilation or metamorphic alteration.

High precision unspiked Os analyses of four fresh chromitite samples (Fig. 2) yield a tight range of weighted mean T_{RD} ages (relative to O-chondrite; Walker *et al.*, 2002), ranging from 3226.6 ± 1.6 Ma ($n = 4$; 95 % confidence limits) to 3244.2 ± 1.2 Ma ($n = 5$). Although the observed scatter in each is greater than the ~0.5 Myr (2*s*) scatter expected from analytical uncertainties, repeated analyses of the same sample ($n \geq 4$) never varied by more than 2.8 Myr. Conventional spiked analyses of the same samples yielded T_{RD} ages from 3232 ± 9 Ma to 3248 ± 9 Ma (2*s*; one analysis per sample) overlapping the high precision ages.

Two altered chromitites have younger and more highly variable model ages in repeated analyses of the same sample, with weighted average ages of 3046 ± 123 Ma (95 % confidence; T_{MA} age; the only sample with detectable Re) and 3181 ± 8 Ma (one spiked and two high precision analyses *per* sample). Taken together, our chromitite data show excellent agreement with previous high precision Re-Os analyses (Coggon *et al.*, 2013), which show maximum T_{RD} ages of 3246.4 ± 1.0 Ma ($n = 4$) and increasing scatter in samples that record younger T_{RD} (3209 ± 50 Ma; $n = 4$).

These 'young' T_{RD} ages relative to the age of the host orthogneisses and Pt-Os model ages have previously been explained by metamorphic resetting of the Re-Os system (Coggon *et al.*, 2015). However, this is difficult to reconcile with consistent T_{RD} ages in fresh chromitites from across the intrusion with variable but high Os concentrations. As igneous chromite has high Os and low Re/Os (Puchtel *et al.*, 2004) and the chromitites presently retain these HSE signatures, this would require that large quantities of Re or radiogenic Os were added to the chromitites during metamorphism, in proportion to the Os abundance in each chromitite. If Re was added, it would need to be removed after a long period of ^{187}Os ingrowth to produce the present day low Re/Os. Though some resetting of the Re-Os system is clearly possible given the younger T_{RD} ages in altered chromitites, this is associated with a significant increase in the scatter of these ages, both within and between samples. Instead, we interpret the Re-Os data to indicate an age of ~3246 ± 120 Ma for the Ujaragssuit Intrusion (oldest age from our data and Coggon *et al.*, 2013, including systematic mantle model uncertainties; Supplementary Information).

Pt-Os isotopic data. The T_{DA} (relative to primitive mantle; Brandon *et al.*, 2006) from all fresh chromitite samples form a single age population with a weighted mean age of 3382 ± 360 Ma (Fig. 2; $n = 17$; MSWD = 0.65; $p = 0.84$). Correcting for ^{190}Pt decay since chromitite formation has no significant effect, yielding a Pt-Os model age of 3398 ± 360 Ma. Though some altered chromitite analyses were indistinguishable from this weighted mean, we exclude these due to evidence of Os mobilisation in these samples from the Re-Os system. Use of alternative mantle models (Day *et al.*, 2017) yields younger ages.

All of these ages appear young compared to previously reported T_{DA} from Ujaragssuit, ranging up to 4.1 Ga (Coggon *et al.*, 2013). However, the uncertainties reported for both individual analyses and age groups in Coggon *et al.* (2013) are far smaller than the ~35 ppm precision they report for the Durham Romil Osmium Standard (DROsS) (Luguet *et al.*, 2008; equivalent to a ~1.5 Ga uncertainty in each T_{DA}). We therefore re-process these data by propagating the minimum analytical uncertainty indicated by DROsS and find: 1) the T_{DA} are also consistent with a single age population, with no statistically distinct older group; 2) the weighted average T_{DA} of 3558 ± 482 Ma (MSWD = 0.50; $p = 0.94$) overlaps our data within uncertainty. We combine all data from our fresh chromitite samples and Coggon *et al.* (2013) to estimate the initial ^{186}Os/^{188}Os of the Ujaragssuit Intrusion, which we believe to

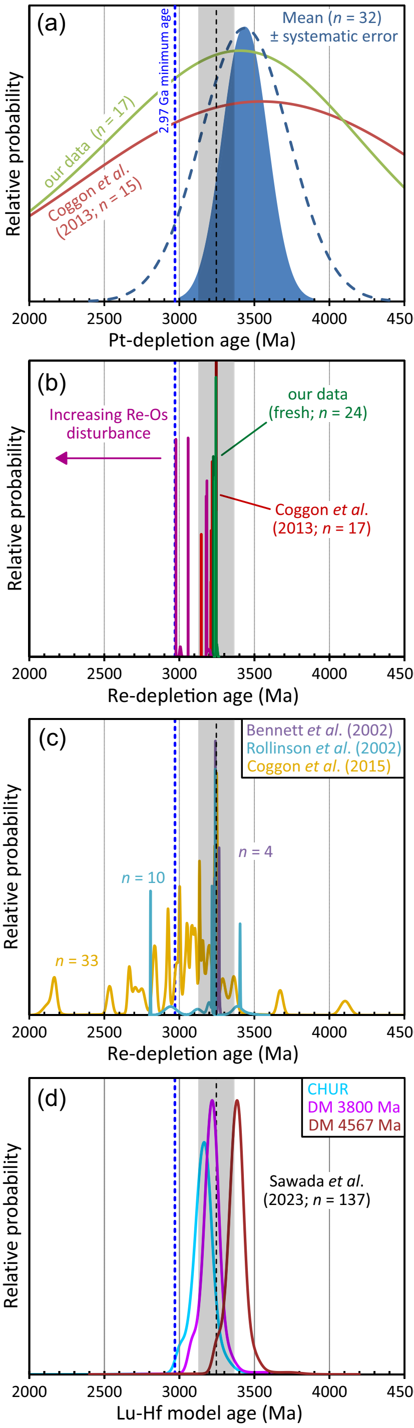

Figure 2 Geochronological data for the Ujaragssuit Intrusion. All plots show the 2970 Ma minimum age derived from cross-cutting leucogranites (blue dashed line) and the oldest high precision chromitite Re-depletion age, including systematic uncertainties (3246 ± 120 Ma; black dashed line and grey field). **(a)** Pt-depletion ages calculated from ^{186}Os analyses from this study and Coggon *et al.* (2013). **(b)** Re-depletion ages calculated from high precision analyses of massive chromitites from this study and Coggon *et al.* (2013). **(c)** Re-depletion ages calculated from other ^{187}Os/^{188}Os literature data (Bennett *et al.*, 2002; Rollinson *et al.*, 2002; Coggon *et al.*, 2015). **(d)** Lu-Hf model ages calculated from metamorphic zircons within the Ujaragssuit chromitites (Sawada *et al.*, 2023). Model ages are calculated relative to chondritic uniform reservoir (CHUR; Bouvier *et al.*, 2008), depleted mantle formed from CHUR at 3800 Ma (Fisher and Vervoort, 2018), and an end-member case where depleted mantle formed at 4567 Ma.

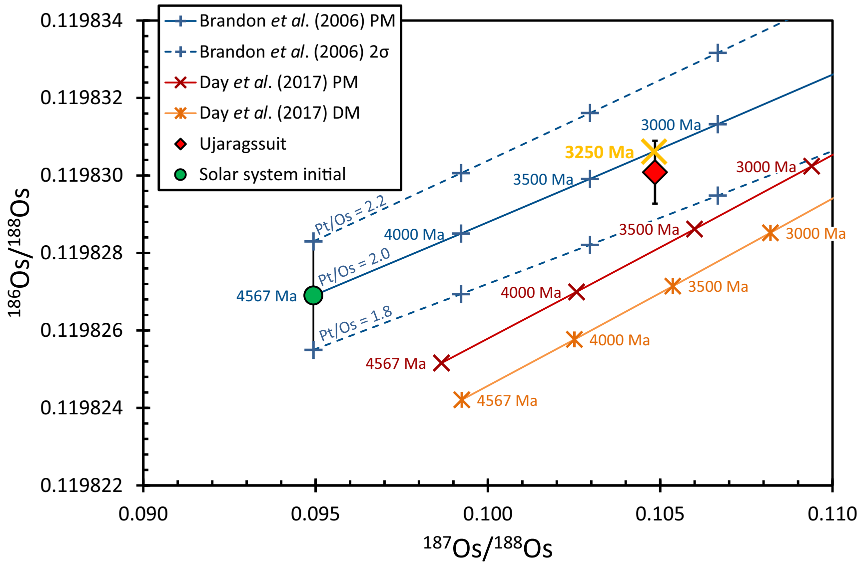

Figure 3 'Concordia' plot of Re- and Pt-depletion ages calculated for different Re-Pt-Os mantle evolution models. The Ujaragssuit ^{187}Os-^{186}Os data are consistent with a concordant ~3250 Ma Re-Pt depletion event from primitive mantle (PM) of Brandon *et al.* (2006) with Re-Os evolution of O-chondrite (Walker *et al.*, 2002). Uncertainties in mantle models from Day *et al.* (2017) are not shown for clarity. DM, depleted mantle.

be the most precise estimate of this ratio for a single terrestrial locality. This yields ^{186}Os/^{188}Os = 0.1198301 ± 0.0000008 (2 s.e.; $n = 32$; MSWD = 0.58; $p = 0.97$; normalised to UMd of Brandon *et al.*, 2006; Supplementary Information), corresponding to a T_{DA} of 3437 ± 288 Ma, or 3437 ± 587 Ma including the ~12 ppm uncertainty in the solar system initial (Brandon *et al.*, 2006).

Although this barely permits a 4.0 Ga age for Ujaragssuit, the mean age is much closer to the age of the intrusion estimated from Re-Os model ages, constituting a concordant Re-Pt-Os depletion age (Fig. 3). Furthermore, the ~3.25 Ga Re-Os model age overlaps with Hf model ages of metamorphic zircon in the chromitites, which requires special pleading if the Ujaragssuit Intrusion is indeed >3.8 Ga (Sawada *et al.*, 2023). The simplest explanation is therefore that the Ujaragssuit Intrusion formed at ~3.25 Ga and there is no evidence for Hadean mantle depletion. Given the lack of observed faults, the Ujaragssuit Intrusion likely has an intrusive rather than tectonic relationship with its host orthogneisses. Finally, we note that this ~3.25 Ga age may have been 'hiding in plain sight', with numerous older publications reporting identical ^{187}Os/^{188}Os to our data (Fig. 2).

Discussion

This study highlights the importance of careful field study and direct dating of ultramafic enclaves in Archaean cratons. While some ultramafic enclaves may be older than their host orthogneisses, others may represent much younger intrusions. In the case of Ujaragssuit, the age determined by direct dating is >550 Myr younger than if the ultramafic rocks are interpreted as an enclave intruded by the orthogneiss protoliths (Nutman *et al.*, 1996).

The precise initial ^{187}Os-^{186}Os systematics allow us to discriminate between potential Pt-Os mantle evolution models (Fig. 3) for the Ujaragssuit mantle source, which is depleted in chondritic components compared to modern mantle (Fischer-Gödde *et al.*, 2020). A mantle source that evolved with primitive mantle Pt-Os (Brandon *et al.*, 2006) and chondritic Re-Os (Walker *et al.*, 2002) yields a concordant age for coupled Re-Pt depletion occurring at ~3.25 Ga, consistent with Lu-Hf constraints (Sawada *et al.*, 2023). By contrast, use of either the primitive or depleted mantle models of Day *et al.* (2017) yields T_{DA} and T_{RD} that are inconsistent. Unfortunately, the large uncertainty on ^{186}Os/^{188}Os compared to its temporal variation means we cannot further distinguish between different Re-Os evolution models for the Ujaragssuit source; providing the Pt-Os evolution follows primitive mantle (Brandon *et al.*, 2006), then Re-Os evolution using any chondrite (Walker *et al.*, 2002) or primitive upper mantle (PUM; Meisel *et al.*, 2001) model yields concordant Re-Pt depletion ages within uncertainty. Given the presence of nucleosynthetic Ru anomalies at Ujaragssuit with no known cosmochemical analogue (Fischer-Gödde *et al.*, 2020), we cannot conclusively rule out a mantle source with exotic Re-Pt-Os isotope systematics. However, any such source would need to reach the Ujaragssuit initial ^{187}Os/^{188}Os and ^{186}Os/^{188}Os at ~3.2 Ga, within uncertainty of chondritic evolution (Fig. 3), to be consistent with zircon Lu-Hf model ages (Sawada *et al.*, 2023). Furthermore, this source requires a nucleosynthetic Os composition indistinguishable from bulk chondrite (see below). The most likely explanation is that the Ujaragssuit source evolved with chondritic Re-Pt-Os isotope systematics until formation of the intrusion at ~3.25 Ga.

We were unable to resolve nucleosynthetic Os anomalies beyond analytical uncertainty (Fig. 4); the slight anomaly in average ε^{184}Os appears to be an analytical artefact (Supplementary Information). This lack of resolvable Os isotopic anomalies reflects an absence of bulk Os isotopic anomalies in most major meteorite groups, including chondrites (Goderis *et al.*, 2017). Despite internal variations in Os composition within individual meteorites (Brandon *et al.*, 2005), Os was homogeneous at the planetesimal scale in the presolar nebula (Goderis *et al.*, 2017). Therefore, even though the Ujaragssuit mantle source was relatively depleted in late-accreted chondritic material (Fischer-Gödde *et al.*, 2020), this had no net effect on its Os isotopic composition.

Our new ~3.25 Ga age for the Ujaragssuit Intrusion sheds new light on previously identified isotopic anomalies, which indicate the mantle sources of NAC igneous rocks were depleted in chondritic components relative to bulk Earth (Willbold *et al.*, 2011; Creech *et al.*, 2017; Fischer-Gödde *et al.*, 2020; Xu *et al.*, 2023). In particular, a ~3.25 Ga age for the Ujaragssuit Intrusion means that nucleosynthetic Ru isotope anomalies

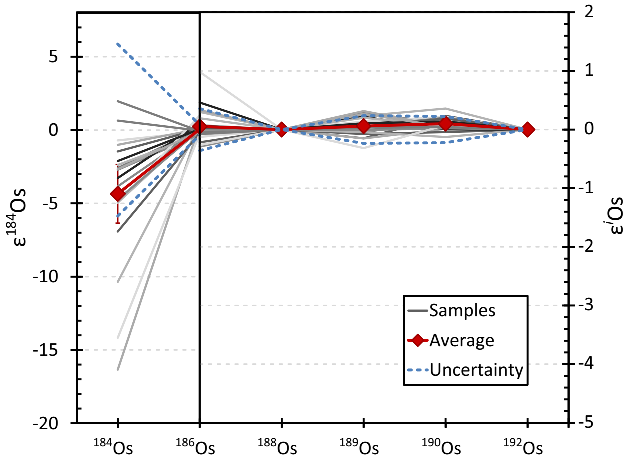

Figure 4 Nucleosynthetic osmium isotope composition of Ujaragssuit chromitites in epsilon units, relative to the DROsS reference material. ^{186}Os is reported relative to primitive mantle of Brandon *et al.* (2006) at 3250 Ma. Uncertainty envelope shows average propagated analytical uncertainty on an individual analysis. Note different scale for ^{184}Os.

(Fischer-Gödde *et al.*, 2020) are now known from NAC ultramafic intrusions formed in four major periods: at ~3.8 Ga in the Narssaq and South of Isua ultramafic bodies, at ~3.7 Ga in the Isua Supracrustal Belt, at ~3.25 Ga in the Ujaragssuit Intrusion, and at >3.0 Ga in the Fiskefjord peridotites. These anomalies occur across three different tectonostratigraphic terranes (Friend and Nutman, 2019), do not diminish over time and have not been identified in other cratons (Fischer-Gödde *et al.*, 2020), even where mantle depleted in late-accreted materials has been proposed (Maier *et al.*, 2009). Repeated tapping of this potentially unique chondrite-depleted mantle source across various NAC terranes over a period of 600 Myr is difficult to reconcile with a model in which the different terranes identified in the NAC were initially widely dispersed (Friend and Nutman, 2019). Instead, it appears to favour autochthonous formation of the various components of the NAC above a mantle source that was relatively isolated and poorly mixed with respect to the rest of the mantle. This most likely supports formation of the NAC within a non-uniformitarian tectonic regime (*e.g.*, Debaille *et al.*, 2013; Webb *et al.*, 2020), in which the crust was relatively immobile with respect to the underlying mantle sources that drove the formation of crustal ultramafic intrusions.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2424.

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