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Iron isotopic evidence for growth of continental crust at convergent margins

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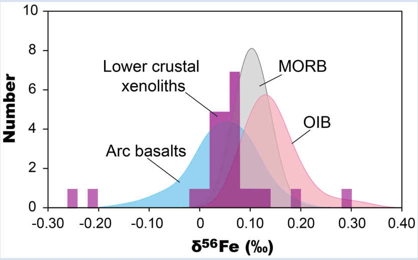
Abstract



Iron (Fe) isotopic compositions of 26 well characterised granulite-facies xenoliths from the Chudleigh and McBride volcanic provinces, North Queensland, Australia, exhibit large variations in δ^{56}Fe, from −0.248 ‰ to 0.287 ‰. The variations result from differences in protoliths and subsequent fractionation processes driven by Fe-Mg interdiffusion. The data yield a weighted mean δ^{56}Fe of 0.045 ± 0.007 ‰ (2 s.e.) for the lower continental crust, significantly lighter than the best estimate of δ^{56}Fe in average upper crust (0.109 ± 0.008 ‰, 2 s.e., compiled from published data). Using these data, the bulk continental crust has a mean Fe isotopic composition of ~0.060 ‰, which is lower than that of oceanic island basalts and Archean tonalite-trondhjemite-granodiorite rocks, but closely resembles that of arc basalts. These results suggest 90^{+13}_{-11}% of the continental crust formed in convergent margins, where arc basalts with light Fe are prevalent due to fO_2-buffered water-fluxed melting of the mantle wedge.

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Introduction

One of the defining characteristics of Earth is its differentiated continental crust, which provides the landmass where humans thrive. However, the origins of the continental crust are still debated, with hypotheses ranging from formation at convergent margins in subduction zones to forming via intraplate magmatism (*e.g.*, Rudnick, 1995; Albarède, 1998; Barth *et al.*, 2000; Foley *et al.*, 2002; Johnson *et al.*, 2017). The arc-like trace element signature of the continental crust suggests growth at convergent margins (Rudnick, 1995; Barth *et al.*, 2000). However, magmas with such geochemical signatures are not exclusively found at convergent margins and may also result from melting at the base of thick mafic crust, or melting of density-driven crustal drips in the upper mantle in intraplate settings (Willbold *et al.*, 2009; Johnson *et al.*, 2017). Recycling and reworking of pre-existing continental crust in various geological settings further complicate the effort to delineate the origins of the continental crust. Thus, the relative contributions of convergent margins *versus* intraplate magmatism to the formation of continental crust remain uncertain.

Iron isotope geochemistry offers a novel perspective regarding the origins of the continental crust. Iron isotopes fractionate based on the strength of Fe-O bonds, with heavier isotopes favoring Fe^{3+}-rich phases (Dauphas *et al.*, 2014). Arc basalts from convergent margins have systematically lighter Fe isotope compositions (expressed as per mil deviations from IRMM014, *i.e.* $\delta^{56}Fe = (^{56}Fe/^{54}Fe_{sample}/^{56}Fe/^{54}Fe_{IRMM014} - 1) \times 1000$) than oceanic island basalts (Fig. 1). This has been attributed to influx of fluids with low δ^{56}Fe, derived from slab serpentinites, into the mantle wedge (Chen *et al.*, 2023) and repeated, water-fluxed melting with fO_2-buffered by oxidised, slab-derived fluids (Foden *et al.*, 2018). The Fe isotopic composition of the continental crust, therefore, can provide insights into the proportions of magmatic crustal growth at convergent margins *versus* within plates. Furthermore, heavy Fe isotopes are commonly enriched in melts compared to their equilibrium residual/crystalline solids (*e.g.*, Telus *et al.*, 2012; Dauphas *et al.*, 2014; Xu *et al.*, 2017), and, thus, recycling and reworking of preexisting continental crust are expected to enrich heavy Fe isotopes in the crust, if the dense, isotopically light residua and crystalline cumulates have been (partially) removed from the base. The mean δ^{56}Fe of present day continental crust can, therefore, provide a minimum estimate of the relative contributions of convergent margin compared to intraplate magmas in its formation (Supplementary Information). Previous studies have estimated that the upper continental crust (UCC) has an average δ^{56}Fe value of approximately 0.10 ‰. This value derives from two independent estimates: a) the global granitoid δ^{56}Fe trend at $SiO_2 = 66.6$ wt. % (Foden *et al.*, 2015), and b) averaging δ^{56}Fe of Mesoarchean to Palaeozoic diamictite composite samples that were not impacted by iron formations (Liu *et al.*, 2022). By contrast, the Fe isotopic composition of the lower continental crust (LCC) is unknown.

To address this gap, we report high precision Fe isotopic data for 26 granulite xenoliths from the Chudleigh and McBride

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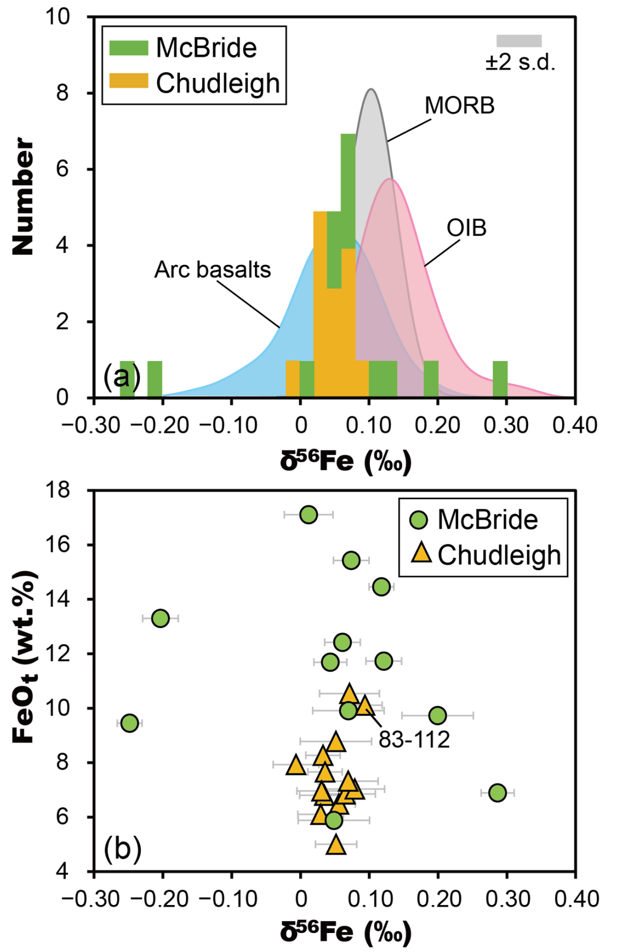

Figure 1 Fe isotopic compositions of granulite-facies xenoliths from the Chudleigh and McBride volcanic provinces, North Queensland, Australia **(a)**, and their variations with FeO_t. **(b)** OIBs, MORBs and arc basalts are from the compilation in Table S-3. Elemental data are from Rudnick *et al.* (1986) and Rudnick and Taylor (1987). Error bars are 2 sigma for δ^{56}Fe.

volcanic provinces, North Queensland, Australia. These samples are representative of the bulk LCC in terms of average elemental compositions (Rudnick *et al.*, 1986; Rudnick and Taylor, 1987; Rudnick, 1990). By averaging their Fe isotopes, and combining this with the above UCC estimate, the average δ^{56}Fe of the continental crust is estimated and, thus, the relative contributions from convergent margin and intraplate magmatism in generating continental crust are determined.

Sample Descriptions and Analytical Methods

Petrology, ultrasonic velocities and chemistry of the Chudleigh and McBride xenoliths have previously been reported (*e.g.*, Rudnick *et al.*, 1986; Rudnick and Taylor, 1987; Rudnick, 1990; Rudnick and Goldstein, 1990; Kempton and Harmon, 1992; Saal *et al.*, 1998; Teng *et al.*, 2013b). The Chudleigh xenoliths, carried in Plio-Pleistocene (<2 Ma) alkali basalts, range from 5 to 50 cm in size and are coarse-grained mafic cumulates (SiO_2 < 52 wt. %). They fall into three groups— pyroxene-rich, plagioclase-rich and transitional, reflecting chemical equilibration at depths of 20–40 km and temperatures of 700–1000 °C. The shallower xenoliths exhibit relict igneous textures and mineralogy, such as olivine (at the cores of orthopyroxene-spinel coronas) and lath-shaped plagioclase; their elemental and isotopic geochemistry (O, Sr, Nd, Pb and Os) suggest a cogenetic origin as crystal cumulates from mafic magmas that intruded and assimilated the preexisting LCC (Rudnick *et al.*, 1986; Rudnick, 1990; Kempton and Harmon, 1992; Saal *et al.*, 1998).

The McBride xenoliths are carried in a young (<3 Ma) basaltic cinder cone (Hill 32), located 180 km north of Chudleigh. They possess well equilibrated textures, and yield equilibration depths of 26–40 km and temperatures of 630–1070 °C (except for 85–107, which is likely derived from a shallower depth of *ca.* 18 km) (Rudnick and Taylor, 1987). Approximately 80 % of these xenoliths are mafic and likely originated from solidified mafic melts, crystal cumulates and residues left after partial melt extraction. The remainder are intermediate to felsic, with protoliths ranging from meta-sediments to crystallised felsic melts. Whole rock $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios suggest a mixing trend around 300 Ma, indicating large scale interaction between mantle-derived mafic melts and preexisting crust that mainly consists of a Palaeoproterozoic basement (Rudnick, 1990). Therefore, both sets of xenoliths represent the present day LCC (<3 Ma) beneath Queensland, and are interpreted to have formed from underplating by mantle-derived mafic magmas, their differentiation and mixing with pre-existing crust. While the McBride samples likely formed in the Late Palaeozoic in a continental margin subduction zone, the Chudleigh samples were interpreted to be cumulates of Cenozoic intraplate basaltic magmatism (Supplementary Information).

Iron isotopic analyses were performed at the Isotope Geochemistry Lab, China University of Geosciences, Beijing. Powdered samples (3–10 mg) were digested in concentrated HF-HNO_3-$HClO_4$, and then sequentially treated with aqua regia twice. Iron was separated using polypropylene columns filled with 1 ml AG1-X8 resin, with matrix elements eluted in 8 ml 6 N HCl and Fe collected in 9 ml 0.4N HCl. This process was repeated to ensure complete matrix elimination. The procedural blank resulted in minimal Fe (<10 ng), less than 0.01 % of the processed Fe, so no blank corrections were applied. Iron isotopic ratios were measured on a Thermo-Finnigan Neptune Plus MC-ICPMS at the "medium" resolution mode. Instrument mass bias was corrected using the standard-sample bracketing method. Standard-sample sequences were repeated nine times, with the reported compositions being the average of these analyses. Long term reproducibility and accuracy were within 0.03 ‰ for δ^{56}Fe, confirmed by analyses on international igneous rock standards (*e.g.*, JP-1, BHVO-2, W-2a, JB-2, AGV-2 and JA-1) (Table S-1).

Iron Isotopic Heterogeneity in the Lower Continental Crust

Granulite xenoliths from Chudleigh and McBride exhibit distinct Fe isotopic compositions (Fig. 1). The Chudleigh suite displays a narrow range in δ^{56}Fe from −0.006 ‰ to 0.094 ‰. The preservation of magmatic trends between soluble elements (*e.g.*, Rb, Ba and U) and Mg# (Rudnick *et al.*, 1986) suggests that granulite metamorphism has not significantly influenced the δ^{56}Fe values. The Chudleigh xenoliths are cumulates from magmas that experienced variable amounts of crustal contamination, indicated by variations in Sr and Nd isotopic compositions (*e.g.*, ε_{Nd} ranging from −6.1 to 9.6; Rudnick *et al.*, 1986). Nevertheless, δ^{56}Fe is constant in these samples and shows no correlation with either ε_{Nd} or Mg# (Fig. S-1). This leads us to conclude that crustal assimilation had a negligible effect on δ^{56}Fe of these samples, which is possible if the contaminant is felsic and relatively Fe-depleted;

the contaminant may also have had a similar δ^{56}Fe as the underplating magma. The Chudleigh granulites have δ^{56}Fe and Nb/La that are lower than those expected for intraplate basalts (Fig. 1), consistent with the accumulation of assemblages dominating by plagioclase and Fe^{2+}-rich pyroxenes with $D_{Nb}/D_{La} < 1$ (Supplementary Information). The positive correlation between δ^{56}Fe and Nb/La (Fig. S-1f) hints at a role for mineral modes influencing the bulk δ^{56}Fe (*e.g.*, light, low Nb/La pyroxenes *versus* heavy, high Nb/La Fe-Ti-oxides), but the effect is rather minimal. An abundance of Fe-Ti-oxides in sample 83-112, which are apparent in thin section and in the sample's high TiO_2/MgO ratio (Rudnick *et al.*, 1986), may have slightly elevated its δ^{56}Fe (~0.094 ‰) relative to the other cumulates (−0.006 ‰ to ~0.079 ‰) that do not show such large amounts of oxide enrichment.

The McBride xenoliths, with more diverse protoliths including crystallised mafic to felsic melts, crustal melting residues and siliclastic sediments, have highly heterogeneous δ^{56}Fe values (Fig. 1, Fig. 2 and Fig. S-2). Eight out of these twelve granulites have δ^{56}Fe ranging from 0.012 ‰ to 0.121 ‰, typical for siliclastic sediments and igneous rocks (Teng *et al.*, 2013a; Foden *et al.*, 2015; Foden *et al.*, 2018; Liu *et al.*, 2022). The two pyroxene mafic granulites that were interpreted to be crystallised melts have the lowest δ^{56}Fe values, ranging from −0.248 ‰ to −0.203 ‰, outside the range observed in arc and oceanic basalts (Fig. S-3; Teng *et al.*, 2013a; Foden *et al.*, 2018). Felsic xenolith 83-162 has the highest δ^{56}Fe value of 0.287 ‰, heavier than most previously reported felsic igneous samples at 66.9 wt. % SiO_2 (Fig. S-3). These unusually low and high δ^{56}Fe values do not correlate with SiO_2, Nb/La, δ^{18}O or protolith types, and cannot be explained by equilibrium melting, fractional crystallisation or protolith inheritance (Fig. S-2). The negative correlation between δ^{56}Fe and δ^{26}Mg (Fig. 2) suggests a role for Fe-Mg interdiffusion, which drives Fe and Mg to diffuse in opposite directions (Dauphas *et al.*, 2010). Samples with highly fractionated δ^{56}Fe and δ^{26}Mg values align with the predicted slope for Fe-Mg interdiffusion (Dauphas *et al.*, 2010), indicating that diffusion-driven fractionation is a plausible explanation for the observed variations in the most isotopically extreme McBride xenoliths (Teng *et al.*, 2013b; and this study). All McBride granulite xenoliths have lower Mg# than would be in equilibrium with the host basalt (Fig. S-2b), suggesting the diffusion of isotopically light Fe into the xenoliths. However, Fe-Mg exchange with the host magma cannot account for the heaviest xenoliths, suggesting that diffusion-driven isotope fractionation may have occurred prior to entrainment in the host, reflecting the inherent heterogeneity of the LCC. Such kinetic processes likely occurred at the boundary of diverse lithologies that were not in Fe-Mg exchange equilibrium, driven by heat input during magmatic underplating, as has been observed at intrusion boundaries (*e.g.*, Wu *et al.*, 2018).

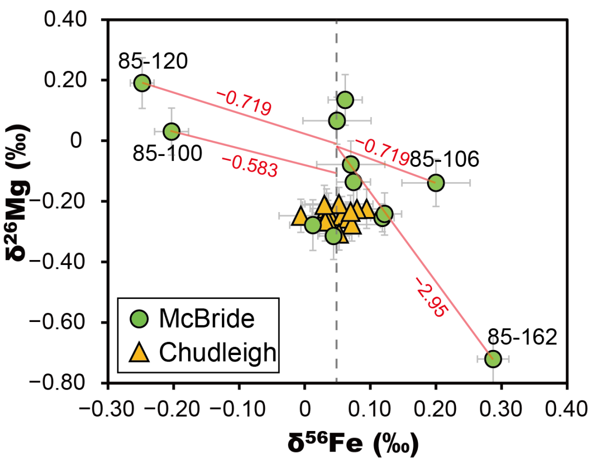

Figure 2 Plot of δ^{56}Fe *vs.* δ^{26}Mg for McBride and Chudleigh granulite xenoliths. Mg isotopic data are from Teng *et al.* (2013b). Symbols marked by sample numbers are the samples that likely have been affected by Fe-Mg interdiffusion. Red lines with labelled slopes represent the projection of Fe-Mg nterdiffusion following the equation ($\frac{\Delta i^{26}Mg}{\Delta i^{56}Fe} \approx -\frac{(\frac{26}{24}-1)}{(\frac{56}{54}-1)} \cdot \frac{Fe}{Mg}$, where Fe/Mg is a mole ratio) from Dauphas *et al.* (2010). Error bars are 2 sigma for δ^{56}Fe and 2 s.d. for δ^{26}Mg.

Iron Isotopic Composition of the Continental Crust

The mean δ^{56}Fe of the LCC beneath North Queensland, Australia, is 0.045 ± 0.007 ‰ (2 s.e.), which is derived from Monte Carlo simulations and weighted averaging of the δ^{56}Fe of the Chudleigh and McBride xenoliths (see Supplementary Information). This value is considered globally representative of the LCC for several reasons:

i) The Chudleigh and McBride xenoliths' average elemental compositions closely match the estimated composition of the bulk LCC (Rudnick *et al.*, 1986; Rudnick and Taylor, 1987; Rudnick, 1990).

ii) Despite their diverse protoliths and elemental compositions, both xenolith suites yield comparable weighted averages for δ^{56}Fe (0.051 ± 0.010 ‰ (2 s.e.) for Chudleigh and 0.041 ± 0.009 ‰ (2 s.e.) for McBride).

iii) The UCC has a mean δ^{56}Fe of ~0.10 ‰, based on a compilation of granitoid and clastic sedimentary rock data (Foden *et al.*, 2015; Liu *et al.*, 2022). With the inclusion of new granitoid data, the UCC estimate has been refined to be 0.109 ± 0.008 ‰ (2 s.e.) (Table S-2). The LCC is expected to have a δ^{56}Fe value lower than the UCC, likely around 0.03–0.07 ‰, assuming that the LCC is complementary to UCC through intracrustal differentiation with isotope fractionation estimated from migmatites (Telus *et al.*, 2012; Xu *et al.*, 2017; Supplementary Information).

Provided that the middle continental crust has a similar Fe isotope composition as the LCC, the mean δ^{56}Fe of the bulk continental crust is estimated to be 0.060 ± 0.004 ‰ (2 s.e.), by combining the average δ^{56}Fe, FeO_t content and weight proportions of the upper (0.109 ± 0.008 ‰, 5.04 wt. %, 0.317), middle (0.045 ± 0.007 ‰, 6.02 wt. %, 0.296), and lower crust (0.045 ± 0.007 ‰, 8.57 wt. %, 0.388) (Rudnick and Gao, 2014; Table S-2).

Growth of the Continental Crust Dominantly at Convergent Margins

Iron can be removed from the continental crust mainly through density foundering of mafic cumulates or residua at the base of the crust if such rocks convert to eclogite (*e.g.*, Kay and Kay, 1993), or through loss of these dense mafic rocks during continental subduction followed by relamination of the less dense portions of the subducted crust (Hacker *et al.*, 2011). During crustal melting, lighter Fe isotopes are concentrated in the residua and the melt becomes heavier (Telus *et al.*, 2012; Xu *et al.*, 2017). The mean δ^{56}Fe of the continental crust, thus, provides an upper limit for the average composition of its primitive building materials. These materials may be basaltic or more evolved slab melts (Rudnick, 1995). Tonalite-trondhjemite-granodiorite (TTG) series felsic igneous rocks constitute a significant fraction of the preserved Archean continental crust (Foley *et al.*, 2002; Moyen, 2011). They are proposed to be partial melts of subducted oceanic crust, and, accordingly, slab melting has been proposed as a key mechanism for continental growth during the Archean era (Martin, 1986; Foley *et al.*, 2002). Available data for Archean TTGs yield a mean δ^{56}Fe of 0.140 ± 0.016 ‰ (2 s.e., N = 53; Table S-5), similar to their younger counterparts (Fig. S-3). The lower mean δ^{56}Fe of ~0.060 ‰ for the continental crust suggests that such slab melts with intermediate to felsic compositions were not the dominant juvenile component contributing to continental growth. The higher geotherm in the Archean era promoted high degree partial melting of the mantle (Herzberg *et al.*, 2010; and references therein), resulting in komatiites with low δ^{56}Fe of ~0.04 ‰ (Dauphas *et al.*, 2010). Nevertheless, komatiites contribute minimally to both surface rocks and TTG sources (Foley *et al.*, 2002; Moyen, 2011). It is likely that most pre-3.0 Ga crust has been largely destroyed (Parman, 2015). Instead, the average Fe isotopic composition of the present day continental crust, which is close to the mean of arc basalts (0.048 ± 0.015 ‰, 2 s.e.; Table S-3) but substantially lower than that of oceanic island basalts (0.146 ± 0.012 ‰, 2 s.e.; Table S-3), indicates that its growth occurred predominantly at convergent margins. A binary mixing model suggests that arc basalts at convergent margins have contributed a minimum of 90^{+13}_{-11}% (95 % c.i.) to continental crust growth (Fig. 3), which is consistent with estimates based on the trace element budget (*e.g.*, 80–95 % estimated from Nb/La; Barth *et al.*, 2000; see also Fig. S-4). Analyses of additional samples of the deep continental crust from other localities would help refine the Fe isotope budget of the continental crust and the exact proportion of continental growth at convergent margins.

The robustness of our estimate is further tested by simulations with continental crust models considering plausible geological variability (Fig. S-5). Felsic magmas with arc-like trace element signatures generated by melting of basaltic crust in intraplate settings are expected to have δ^{56}Fe values higher than their parental sources, represented by OIB (Supplementary Information). The low δ^{56}Fe for bulk continental crust suggests that felsic intraplate magmas with arc-like trace element signatures were not important in continental growth.

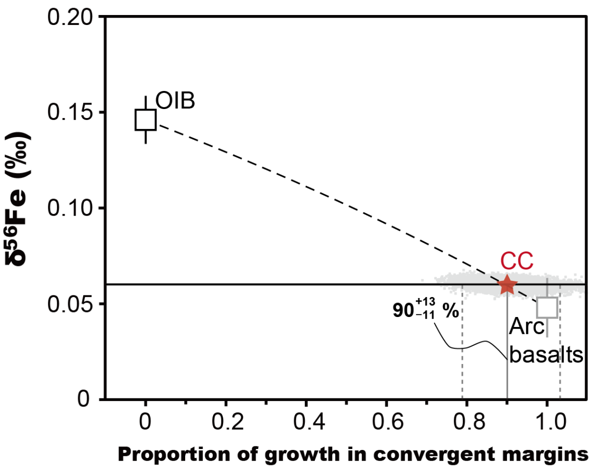

Figure 3 Proportion of continental crustal growth due to convergent margin *vs.* intraplate magmatism estimated from the Fe isotope budget of the continental crust. Mean compositions of arc basalts and ocean island basalts (OIBs) are from the compilation in Table S-3. The proportion was calculated based on binary mixing (*e.g.*, the black dashed line), with the uncertainty constrained by Monte Carlo simulations (Supplementary Information). Results of representative Monte Carlo runs (*i.e.* the first 10^4 runs) are plotted as grey dots.

Conclusions

We present Fe isotopic data for well characterised lower crustal granulite-facies xenoliths from the Chudleigh and McBride volcanic provinces, North Queensland, Australia. The δ^{56}Fe of these xenoliths range from −0.248 ‰ to 0.287 ‰, reflecting both heterogeneity in their protoliths and the influence of kinetic processes. These data yield a mean δ^{56}Fe of ~0.045 ‰ for the lower continental crust. When combined with literature data for the upper crust, the bulk continental crust has an estimated mean δ^{56}Fe of ~0.060 ‰, close to that of arc basalts. Our findings support the notion that growth of the continental crust has predominantly occurred at convergent margins and that arc basalts are the predominant juvenile component contributing to continental growth.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2542.

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