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Distribution and evolution of uranium in the oceanic lithosphere

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Induced fission track techniques permit us to determine quantitatively the microscopic distribution of uranium in rocks, in their constituent minerals, and in percolating fluids. Both primary magmatic variations and secondary mobilization of uranium can be discerned.

Concentrations of uranium in phenocrysts and fresh glasses of oceanic basalts and gabbros are very low $(2-80 \text{ parts}/10^9)$ and are comparable to concentrations in the same minerals of the associated ultramafic rocks. Variations with depth in D.S.D.P. holes show several distinct cyclic variations of uranium, accompanied by parallel trends in some major and trace elements. In Hole 332B (mid-Atlantic ridge, 36 °N), uranium and other elements can be shown to fall into two distinct groupings, each group following its own characteristic fractionation trend, suggesting that two distinct magmas differentiated independently beneath the median valley, the two magmas alternating in their contribution to the formation of oceanic layer 2.

Earlier investigations of the uranium distribution in surface pillows and other dredged rocks exposed to sea water had shown that, owing to halmyrolysis, the uranium concentration increases systematically with distance from the axis of a midoceanic ridge. Subsequent investigations on rocks drilled from horizons deeper into oceanic layer 2 indicate that secondary enrichment or redistribution of uranium is confined to specific zones of altered basalt, near fractures, pillow and flow margins, and especially along horizontal planes of breccias and sediments in between massive flow where convective water circulation is thought to occur. Ultramafic rocks from the base of layer 3 and top of layer 4 are also enriched in uranium when hydrated by sea water during the process of serpentinization. A combination of these processes may double the uranium content of an oceanic lithospheric plate between the time of its formation and its eventual subduction.

Although uranium occurs only in trace concentrations in the oceanic crust, it is, by reason of its radioactivity and mobility, one of the crust's more important constituents. Our knowledge of the uranium distribution in the crust is critical to our understanding of the origin and generation of oceanic magmas, and to the estimation of the composition of the oceanic upper mantle, from which we can calculate its thermal energy potential. We also need to know the chemical changes which occur with time through interaction of oceanic rocks with sea water; these occur between basalts and cold sea water (halmyrolysis), and under more elevated temperatures, affecting both basalts and more deep seated rocks (hydrothermal); these reactions will control the eventual composition of 'old' oceanic crust and upper mantle before subduction. Finally we can use the uranium present to determine the geochronology of ocean floor spreading through radiometric dating.

Before 1974 our observations were limited to rocks accessible to the more primitive methods of ocean floor sampling, such as dredging, grabbing and coring. Obviously all rocks so collected had been in direct contact with sea water for extended periods of time, either from the instant of their formation, or since exposure through faulting and erosion. There appeared a regular

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pattern of alteration of surface rocks, dependent on time, and related to the extent of their oxidation and water absorption. Original whole rock uranium concentrations of the order of 100–200 parts/10⁹ increased at the rate of at least 1000 parts/10⁹ per 10 Ma, equivalent to 1000 parts/10⁹ for every 2% of sea water absorbed. Correlation coefficients of 0.8 were obtained for the U/(Fe₂O₃/FeO) relation, and of 0.6 for U/(H₂O total percentage), whereas K₂O appeared to behave somewhat differently, with a low correlation coefficient of 0.4 between U and K₂O (Aumento 1971).

At that time we questioned the validity of these results when applied to a finite thickness of the oceanic crust, since it was apparent that even with the larger specimens dredged the degree of alteration varied radially, decreasing from the margins of pillows to their centres. A typical 10 Ma old pillow might have 500 parts $U/10^9$ in its core and 4000 parts/10⁹ at its margin. At the same time one might find a 16 Ma old pillow sealed from contact with sea water by the rapid deposition of a thick ferromanganese crust, in which sideromelane with less than 100 parts/10⁹ uranium could be found, and in such a fresh condition as to be suitable for fission track dating.

At about that time major discrepancies appeared between theoretical heat flow calculations for cooling, accreting oceanic plates and the thermal profiles measured at sea, especially in the axial regions of mid-oceanic ridges. These data, together with other evidence, such as the discovery of the hot Red Sea brines, the investigation of the geothermal areas in Iceland, and the discovery of the occasional geothermal deposit on the deep ocean floor, pointed to the existence of another very efficient cooling mechanism at work beneath the solid ocean floor (Sclater & Francheteau 1970; LePichon & Langseth 1969). It was postulated that the oceanographers' heat probes were measuring conductive heat only, and that there might exist a system of convective sea water circulation, rapidly extracting heat from the more deeply buried, hot oceanic rocks (Lister 1972).

If these convective cells do exist, do they permeate the whole crust, or are they restricted to specific channels and zones? Are the channels horizontal, vertical, or do they simply follow faults and fractures? And what would be the state of alteration and resultant uranium content of the oceanic crust beneath the immediate surface? Would alteration decrease with depth, or would hydrothermal alteration take over where halmyrolytic waters were not capable of penetration? In time, would impervious sedimentary covers effectively seal the oceanic rocks from repeated water interaction, or would circulation continue unhindered? These were important questions to be resolved, since they had a bearing not only on the uranium and its heat generation properties, but on the overall geochemical state of the rocks and their physical and palaeomagnetic properties.

Fortunately, through the efforts of the Deep Sea Drilling Project, first in 1974 (Leg 37), and later on I.P.O.D. Legs, repeated penetrations of the upper parts of oceanic layer 2 have given us a much clearer picture of the state and structure of the crust down to the transition between oceanic layers 2A and 2B (Aumento *et al.* (eds) 1977). These holes have shown us the complexity, both horizontally and vertically, of the oceanic crust. Of interest to our water circulation questions was the discovery of massive intercalations of soft sediments and volcanic rubble, with interstitial voids, in between lava flows throughout oceanic layer 2A, and the realization that the seismic boundary between oceanic layers 2A and 2B is a diffuse horizon, below which the sedimentary intercalations cease to be of importance. This suggests that layer 2A could be a highly permeable volcanic-sedimentary sequence formed towards the end of massive

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submarine eruptions on the floor of the median valley, when sporadic volcanicity gave enough time for deep sea sediments and volcanic rubble to collect on top of the lava flows; these deposits were eventually buried by renewed volcanic outbursts, the process repeating itself a number of times.

At least the upper part of layer 2, therefore, is an ideal medium for the circulation of convective water cells. Detailed investigations of the uranium and other trace element distributions, of the oxidation states of the rocks, and of the individual minerals such as titanomagnetite (studied through measurement of their cation deficiency by paleomagnetic techniques), of the isotopes, and of secondary mineralogy, have permitted us to delineate more realistically the extent, the possible geometry, and the effect of the water cells. The ease with which uranium can be studied by induced fission track techniques, both in a bulk quantitative way and in its microscopic distribution patterns, has rendered the element particularly suited to these investigations. Summaries are presented below of both the primary and secondary distributions of uranium, and their relevant implications.

PRIMARY URANIUM IN BASALTS

The average whole rock uranium concentrations in basalts obtained by drilling have ranged from 100 to 290 parts/109 (Mitchell & Aumento 1977). Investigations of the constituents of fresh pillows show predictable uranium distributions. Olivine contains between 2.5 and 10.5 parts $U/10^9$ uranium, spinel between 3.0 and 5.0, plagioclase between 7.4 and 11.6, clinopyroxene between 10 and 42, fresh sideromelane between 19 and 83, and very fine grained groundmass between 78 and 230 parts/10⁹. Even the slightest palagonitization increases the uranium content of sideromelane to the range 150-1900 parts/109; since similar subtle alterations of the groundmass are not readily detectable, the question is raised whether the values reported for supposedly fresh groundmass are not those of groundmass which has already suffered some secondary uranium enrichment beyond that of the fresh sideromelane level, i.e. an increase from the range 19-83 parts/10⁹ range to the range 78-230 parts/10⁹ through the undetectable incipient palagonitization of minute sideromelane shards within the groundmass. A basat consisting of fresh glass and phenocrysts only would not have more than 80 ppb uranium: this may well be the level of the uranium concentration of ascending magmas before consolidation and incipient contamination.

One of the holes drilled into 16 Ma old Atlantic crust, site 335 at 36 °N (Mitchell & Aumento 1977), contains the surprisingly low 100 parts/109 whole rock concentrations expected from the foregoing arguments for extremely fresh basalts. The low average concentrations might have been explained if the pillow sequence had remained totally impervious to percolating fluids. However, cation deficiency measurements of titanomagnetite crystals (Hall & Ryall 1977), and evidence from other studies, indicate that these basalts have probably undergone more alteration than those of the other, younger drill sites; surprisingly, there was no associated secondary uranium enrichment. Furthermore, since there is no evidence for loss of uranium to sea water, one must conclude that the magma from which the pillowed sequence at site 335 was derived was originally depleted in uranium. Other investigators, including Bryan & Thompson (1976), Schilling et al. (1977), and Lambert & Holland (1977), noted the singular chemical nature of basalts at this site.

Whereas there is no uranium variation through the 100 m of drilled sequence of basalts at

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site 335, the uranium distribution pattern at the other 36 °N sites is quite marked. At site 332B, for example, several cycles of uranium concentration decreasing with depth through individual volcanic units can be recognized, with corresponding major element decreases in SiO_2 , Al_2O_3 , K_2O , TiO_2 and P_2O_5 , and an increase in MgO. In addition, U-MgO plots show two distinct diverging trends; in one there is the trend of decreasing uranium and increasing MgO for units exhibiting olivine settling: this will account for the increase in MgO, and since the olivine crystal lattice can accommodate only trace amounts of uranium (2.5–10.5 parts/10⁹), it will be accompanied by a lowering of the uranium concentration. The other trend, that of variable uranium at an average 7% MgO, is the result of clinopyroxene and plagioclase fractionation. That these two U-MgO trends are the result of two different magmas differentiating independently becomes more apparent on consideration of other trace element data, where rocks from the two trends produce two distinct groupings when plotting element pairs U/Sr, U/Zr, U/Y, and U/Ba.

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Thus from the uranium and other trace element data one can show that two distinct magmas were involved in the formation of one small section of the oceanic crust at site 332. The differentiation products of the two magmas occur in a complex interfingering relation through the 700 m of basalts penetrated in hole 332B. This rapid alternation of the two quite different magma types suggests that strong tectonic activity was taking place in the median valley of the mid-Atlantic ridge at the time of formation of the oceanic crust, permitting the differentiated magmas to be tapped repeatedly from two distinct sources beneath the Median Valley. The nature and relative positions of the two magma reservoirs is still in question: one might envisage two chambers side by side, or one beneath the other, or possibly different sites within one chamber; what is necessary, however, is for the two sources to be distinct, but in proximity, and differentiating independently.

PRIMARY URANIUM IN THE LOWER CRUST AND UPPER MANTLE

One of the more marked down-hole primary variations in uranium concentrations occurs in D.S.D.P. Hole 334, where an abrupt decrease is observed from an average of 180 parts/ 10^9 in the upper part of the hole to an average of 30 parts/ 10^9 below the 310 m sub-bottom depth. This marks the transition from the overlying basalts to gabbros and ultramafic rocks below. The underlying ultramafic sequence may be representative of an upthrusted section of oceanic layer 3 or layer 4, probably through the action of transform faults (Robinson *et al.* 1977).

As for similar rock types dredged previously from the Earth's oceans, this drilled sequence shows varying stages of serpentinization and amphibolitization. Measured whole rock uranium values, ranging from 6 to 330 parts/ 10^9 in the case of the drill cores, and from 19 to 700 parts/ 10^9 for the dredged equivalents, are hardly indicative of primary magmatic concentrations. Individual minerals may, however, still retain primary concentrations, although, as in the case of orthopyroxene, some anomalous results have been obtained (Aumento & Hyndman 1971).

Olivine, always partly serpentinized, contains between 2.5 and 3 parts $U/10^9$ in absolutely clear sections of crystals, the uranium increasing to 30 parts/10⁹ in areas showing even the slightest alteration. Plagioclase is usually low in uranium, around 6.8–8.2 parts/10⁹, with concentrations similar to those found in the plagioclase crystals of basalts. Concentrations in chrome spinels are highly variable, from 40 to 600 parts/10⁹. The pyroxenes are also very variable in their uranium content: though both clinopyroxene and orthopyroxene generally

yield values from 10 to 150 parts/10⁹, there have been occasional high values reported from certain oceanic areas: ultramafic rocks from the mid-Atlantic ridge at 45° N and 52° N (Aumento & Hyndman 1971) and from the Indian Ocean (Vinogradov *et al.* 1969) have yielded clinopyroxenes with 400 parts U/10⁹, and more surprisingly, orthopyroxenes with up to 1500 parts/10⁹. These high levels in orthopyroxene are suspect: in coexisting cumulate phases one would have expected uranium to have been preferentially held by the clinopyroxene phase relative to the orthopyroxene. And yet the crystals in question are apparently fresh: microscopic observations show no evidence whatsoever for secondary alteration, or of the homogeneous inclusion of particles which may be carrying uranium.

Not only does the slightest alteration enhance the uranium content of the primary minerals, but secondary phases themselves are invariably rich in uranium. The latter, being generally hydrated equivalents of the primary phases, indicate that the reactive fluids must themselves have been rich in uranium. Clear serpentine, with 130–560 parts U/10⁹, shows further enrichment to 480–600 parts/10⁹ when stained reddish brown by oxidation products. Haematitic veins range from 580 to 2300 parts/10⁹, and late stage chrysotile veins cutting across all other features have 440–800 parts U/10⁹.

If the values obtained from individual minerals represent primary uranium concentrations, then one can calculate the uranium concentrations of the original gabbroic/peridoitic rocks before serpentinization. One obtains two sets of ranges: most fresh ultramafic rocks may contain 10–30 parts/10⁹, whereas by using the anomalously high pyroxene values, there may also exist ultramafic rocks with 90–500 parts U/10⁹. The low range is in keeping with value expected for an ultramafic cumulate or the residual material from the extraction of basaltic magmas with somewhat higher uranium content. Although there is as yet no evidence for any oceanic basalt having direct petrogenetic affinities with the associated ultramafic rocks, it is improbable for the underlying ultramafic rocks to have higher primary uranium concentrations than the overlying basalts: hence the high uranium range (90–500 parts/10⁹) remains an inexplicable anomaly with interesting implications for the heat production properties of the oceanic upper mantle.

MOBILIZATION OF URANIUM

Uranium is a sensitive indicator of the alteration state of a basalt. When sideromelane is hydrated and oxidized by low temperature sea water, it produces, among other things, iron oxide and the zeolite phillipsite (their intimate admixture being termed rather loosely 'palagonite'), and is associated with an enrichment of uranium. Fission track maps show that even the thinnest palagonite rim surrounding sideromelane is enriched in uranium by an order of magnitude. This alteration is especially noticeable around phenocrysts, small fractures, and chilled contacts with sediments (see, for example, Mitchell & Aumento 1977).

Whole rock alteration of basalt increases with proximity to flow margins and fractures, but the effect occurs on a limited scale only. Thick, massive flows and large pillows are often highly altered near their margins, but their centres can be relatively fresh. The uranium-enriched zones are generally iron oxide stained and hence easily recognizable visually. Chemically one finds associated increases in Fe_2O_3 FeO, H_2O_{tot} , K_2O , CO_2 , Ba, Rb, Li and Sr, and the cation deficiency factor (Z) for titanomagnetite. These indicators do not show any systematic downhole variation; they do, however, show marked increases at specific horizons, as well as some trend of overall increase with time. Uranium concentrations at site 335, the oldest site drilled

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during Leg 37, contradicts this trend completely by giving the lowest uranium values on record; these, as explained previously, may be the result of a superimposed primary effect. The isotope ratios ${}^{87}\text{Sr}/{}^{86}\text{Sr}$, $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ have higher values in the upper levels of the core, suggesting greater interaction with sea water for rocks belonging to layer 2A than for those of layer 2B. Averages for the same ratios increase systematically with distance from the spreading centre, indicating that the passage of time, as well as the proximity to oceanic waters, has contributed to isotopic exchange with sea water.

It is difficult to explain why uranium does not correlate with these isotopes, and other elements mentioned, in their enrichment with time at depth, when it does so markedly in the surface rocks and along specific horizons. It does not appear to be a question of temperature control, since oxygen isotopes indicate that all the exchanges occurred at low ocean floor temperatures (Muehlenbachs 1977) even at depth. It seems more probable that uranium is deposited only where there is an active passage of sea water, rather than where thick sequences of rocks bathe in a more stagnant environment over prolonged periods. The detection of these zones of free passage of water, which have important implications in our understanding of the cooling mechanisms of the oceanic crust, may well be accomplished most effectively by detecting zones of anomalously high uranium concentrations.

Indeed, down Hole 332B, superimposed onto cyclic variations in uranium of a primary nature, one finds zones a metre or so thick in which whole rock concentrations increase by up to an order of magnitude. These abrupt increases occur at the junction of lithologic units, as for example between units IV and V at 465 m depth, VI and VII at 590 m, and IX and X at 639 m depth. Other high values occur in the upper sections of Holes 332A, 333 and 334. The boundary zones between these lithologically defined volcanic units are zones of sedimentary interbeds of nanno-fossil ooze and volcanic rubble, and represent time intervals between volcanic eruptions. Hence the upper parts of a previous volcanic outpouring lying exposed to the sea floor may have undergone incipient alteration by contact at first with sea water during its period of exposure, and later by the passage of percolating sea water through the overlying permeable zone.

HYDROTHERMAL CIRCULATION

Surface probe heat flow measurements in the proximity of the axes of mid-ocean ridges give values consistently below those predicted from theoretical calculations for an accreting, cooling plate. Measurements taken around sites 332 and 333 are no exception: with a mean value of 5.14 W cm^{-2} (1.23 cal cm⁻² s⁻¹; 1.23 h.f.u.) (Aumento *et al.* (eds) 1977), they are more representative of heat flow values expected from much older oceanic crusts (Le Pichon & Langseth 1969).

Subsequent down-hole measurements of temperature for Holes 332A, 332B, and 333 confirm the discrepancies: for example a temperature of 14.9 °C at 551 m sub-basement for 332B is extremely low considering the proximity of the site to the active spreading centre (35 km to the east); the resultant heat flow values, 0.70, 0.57, and 0.50 h.f.u. for the three holes respectively, are even lower than for surface measurements. They are indicative that the convective mechanisms responsible for cooling of the oceanic crust in that area are active to well below the 400–500 m level sub-basement. Similarly, the uniformity of heat flow across the sites 332 and 333, both of which lie within a small sediment filled valley, requires that convective cells be very large and regular, and stable over large horizontal and vertical distances. One can

envisage using the uranium distribution and heat flow evidence, the existence of localized downgoing cold water cells (probably following fault planes parallel to the axes of the midocean ridges), spreading sideways along specific rubbly horizons and planes of weakness in between massive flows and sills, eventually warming up and rising along fault planes. Upon their emergence into cold sea water, the upwelling waters will discharge heat and elements stripped from the underlying rocks. Given suitable conditions of salinity, topography, bottom currents and temperature, these elements may not be dispersed to the oceans, but may be deposited close to the ocean floor, forming small hydrothermal deposits.

Some of the horizontal limbs of the convective cells will travel along the many horizons demonstrated to occur in abundance within layer 2A; there will also be other, deeper horizons in layers 2B and 3, probably along similar flow or sheet contacts. Cooling is expected to be so efficient that temperatures should not exceed 75 °C at 5 km depths (Robinson *et al.* 1977). The downgoing cells, still rich in uranium, must on occasion, also find their way down the same planes up which partly serpentinized rocks from layer 4 are being upthrusted, resulting in their additional hydration, and the production of more uranium rich serpentine minerals.

URANIUM CONTENT OF AN OLD OCEANIC CRUST

We have seen that all oceanic basalts are altered to some degree by low temperature interaction with sea water (halmyrolysis), and while whole rock compositions do appear to change systematically more with time than with depth, corresponding uranium changes are more restricted. Uranium behaves differently: at least the upper part of oceanic layer 2 shows no overall increase in uranium with time, in contrast to specific horizons within and to rocks constantly exposed to sea water. Concentrations increase rapidly along any open passage that permits sea water to migrate through the lava pile. However, alteration and uranium concentrations of this kind do not extend far into the host rock: perhaps a few centimetres on either side of fractures, not more than a few metres from contacts and permeable horizons. Therefore, in contrast to localized order of magnitude changes in surface rocks and adjacent to passageways, one cannot expect the overall increase in uranium of the basaltic ocean crust to be more than twofold or threefold.

Quantitatively more important are the uranium levels in the overlying sediments $(100-300 \text{ parts}/10^9)$, since these are additions with time to an already established crust, and can range from a few tens of metres to several kilometres in thickness; the ferromanganese coatings on exposed rocks are very rich in uranium $(5-10 \text{ parts}/10^6)$, but being only a few centimetres thick they contribute little to any overall change. Extremely important may be the serpentinized ultramafic intrusives: at one time their existence was considered rare, but subsequently they have been found to be widespread and important constituents of oceanic layers 2 and 3. These ultramafic rocks, with uranium values increased by serpentinization from low tens of parts/10⁹ to 500 parts/10⁹ and more, might make up a significant fraction of the uranium content of an older oceanic crust; what we do not have at present is a good estimate of their abundance in a typical section of the oceanic crust.

Let us assume that an accreting oceanic plate is made up of 1500 m of basalts, which, when absolutely fresh, might contain 60 parts U/10⁹. These are underlain by 3000 m of diabases and gabbros with 20 parts U/10⁹, and some 100 km of ultramafic rocks with 10 parts U/10⁹. A simple indication of the total uranium held by a column of new oceanic plate is given by

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the sum of the uranium content in parts/ 10^9 times the thickness in metres of the constituent units; such an approximation gives 1.1×10^6 units of uranium for the plate at time zero. Upon re-examination of this same plate some tens of millions of years later one might expect to find it covered by a thick layer of sediments; let us say conservatively that 2000 m of sediments have accumulated, with an average content of 200 parts $U/10^9$. Beneath this, one may find 5 cm of ferromanganese crust with 10 parts $U/10^6$ encrusted onto the pillow lavas. The latter, still 1500 m thick, now contain 200 parts $U/10^6$. Within this level, or directly beneath it, there may be a 1000 m thick serpentinite intrusion (either as a single unit or as numerous dismembered bodies) with 500 parts $U/10^9$, underlain by 2500 m of gabbro (some of it having been serpentinized and now included with the serpentinite layer) with its original 20 parts U/10⁹, and the underlying massive 100 km of ultramafic rocks still retaining their 10 parts $U/10^9$. The total uranium content from such a section would be 2.2×10^6 units, or twice the original content of the new plate. We can readily increase or decrease this new value by changing the thickness of the sedimentary layer, a parameter we can measure readily for individual cases, or the importance of the serpentinite intrusions, or the serpentinization of the mafic/ ultramafic layers without specifying that they be intrusive. Studies of ultramafic rocks from ophiolites (Dostal et al. 1975), which are supposedly derived from the emplacement of oceanic layers 3 and 4 onto continental crust indicate that the uranium content of even the unmobilized horizons may have increased to the range 13-74 parts/10⁹. Hence the overall estimate of the final uranium content for the ultramafic rocks may be on the low side.

CONCLUSIONS

The newly formed oceanic lithosphere shows a systematic stratification of uranium concentration, decreasing with depth from 60 to 10 parts/10⁹, the decrease paralleling the change from basalt through to gabbro to ultramafics. During its residence period beneath the oceans, the upper part of the lithosphere undergoes both chemical and mechanical modification. Changes in uranium concentrations are localized: there will be a totally new occurrence formed by the deposition of the sedimentary oceanic layer 1; basaltic layer 2 will double its content through enrichment by convective sea water migration along surface boundaries and other fractures; the gabbroic layer 3 will remain largely unchanged; finally there will be a massive addition of uranium through sea water hydration of the lower part of layer 3 and the upper part of layer 4, the ultramafic layer, during serpentinization. The overall effect will be to increase the uranium content of the oceanic lithosphere, perhaps doubling it during its existence as an oceanic plate. Should such a plate be subducted, it will carry with it more uranium down back into the mantle, even if all sediments are scraped off, than was originally given up by the mantle in the process of producing the plate.

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