# U-Th Isotopes in Arc Magmas: Implications for Element Transfer from the Subducted Crust

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Uranium-thorium isotope results from island arc volcanic rocks were used to investigate the rates of transfer of fluids and sediments from the downgoing slab. Uranium, but not thorium, is readily mobilized in the fluid. A negative array between thorium/cerium and neodymium-143/neodymium-144 indicates that significant amounts of the thorium in arc rocks are derived from subducted sediments, although perhaps only about 30 percent of the thorium in subducted sediments is returned to the crust in this way. The transfer times for fluid through the mantle wedge are about 30,000 to 120,000 years, whereas those for sediment melts may be several million years. The low average uranium/ thorium ratios of bulk crust primarily reflect different crustal generation processes in the Archaean.

Destructive plate margins are major sites of differentiation in the evolution of the crustmantle system. The mantle is cooled by the subduction of cold oceanic crust, yet island arcs contain many of the most active and hazardous volcanoes on Earth. Partial melting occurs in response to the introduction of fluids from the subducted crust, and many arc magmas contain a significant additional contribution from subducted sediments (1-10). The magmatic flux constitutes a major component of new crust, and conversely the subduction of oceanic crust and sediment forms the principal mechanism for recycling of crustal materials into the upper mantle.

A major goal in the earth sciences is to understand the physical processes and element fluxes involved in the generation of arc magmas. A number of chemical tracers for recycled sediments have been developed, most notably <sup>10</sup>Be (4), but the balance of new and recycled material, the controls on magma composition, and the rates of movement of material from the subducted slab are not well known. Some elements are preferentially enriched, and others are relatively depleted in arc magmas, as compared with magmas generated along mid-ocean ridges and in intra-plate settings. Different elements behave differently; some are preferentially mobilized in fluids from the subducted slab, and the proportions of recycled and new mantle-derived material vary from element to element. Kay (11), for example, argued that  $\sim 90\%$  of the K in island arc magmas was recycled from preexisting continental crust, whereas many estimates for elements such as Ta, Nb, and Ti have suggested that they are derived largely from the upper mantle, with little or no contribution from subducted crust (8, 10).

The application of short-lived U-series isotopes has revolutionized the study of melt generation processes by providing information on the time scales of U-Th fractionation, and hence on the depths and rates of partial melting (12-14). Uranium and thorium appear to be readily fractionated in the processes responsible for island arc magmatism, and they are involved in radioactive decay schemes that may be used to investigate processes that operate on time scales ranging from a few tens of thousands of years to several billion years (15-17). Here we report <sup>238</sup>U-<sup>230</sup>Th isotope data on rocks from widely different island arc suites, chosen because their minor and trace element compositions may be modeled in terms of very different relative contributions from subducted sediments and fluids from the downgoing slab. The aim is to clarify the behavior of U and Th in the generation of arc magmas, to evaluate the fluxes and rates of transfer of different materials from the subducted slab, and to explore the implications for the generation of bulk continental crust and the recognition of recycled crust in oceanic basalts.

## Results

<sup>238</sup>U decays to stable <sup>206</sup>Pb via a chain of short-lived isotopes that includes <sup>230</sup>Th and <sup>226</sup>Ra. The half-life for <sup>238</sup>U is  $4.47 \times 10^9$ years, and so <sup>206</sup>Pb/<sup>204</sup>Pb variations in igneous rocks are widely used to evaluate differences in source components that may have evolved over hundreds of millions of years or more (18). In contrast, the half-lives of <sup>230</sup>Th and <sup>226</sup>Ra are 75,380 and 1662 years, so they provide unique insights into the timing and rates of processes that have fractionated U, Th, and Ra within the past 350,000 years. The approach adopted here is to evaluate the causes of the fractionation of U from Th in arc magmas, and then to use U and Th isotopes to investigate the rates, and hence the mechanisms, of transfer of material involved in the generation of arc magmas. Island arc suites were chosen for which comprehensive data sets were available and which encompassed a reasonable range of isotope and trace element compositions. Rocks from Central America have been omitted because the subducted sediments in that area have unusual high U/Th ratios (19), due to a high proportion of carbonate, and these result in regionally distinct mixing vectors on isotope and trace element diagrams (17, 20).

Results from representative rocks are presented in Table 1 and, for clarity, analvses from similar arcs are grouped together in the figures. Thus, those from the arcs that are more depleted in incompatible elements-the Marianas (21, 22), Tonga-Kermadecs (23), the South Sandwich Islands (24), and Vanuatu (25)-are considered together, as are those from the arcs that are more enriched in incompatible elements-the Aeolian Islands (26, 27), Indonesia (28-30), and the Philippines (31). Arcs with intermediate compositions include the Lesser Antilles (32, 33) and Japan-Kamchatka (34-36). We only considered data for basalts and andesites in order to lessen the effects of fractionation and crustal contamination on incompatible element and isotope ratios.

A distinctive feature of island arc rocks is that they have high ratios of large ion lithophile elements (LILEs) to high field strength elements (HFSEs) (37), and these ratios are highest in the more depleted, low-K rocks (5, 8, 10). The high LILE/ HFSE ratios are not commonly observed in other rock types, and so they are widely attributed to subduction processes and typically to the preferential mobilization of LILEs in hydrous fluids from the downgoing slab. Both U/Th and Ba/Th ratios are highest in rocks with low Th abundances (Fig. 1), so it is inferred that in arc magmas U behaves similarly to LILEs such as Ba, whereas Th is much less mobile. The arc rocks with higher Th contents have higher light rare earth element abundances and higher Ce/Yb ratios (5), and, as argued be-

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low, they contain a greater contribution from subducted sediments.

In a diagram of (<sup>230</sup>Th/<sup>232</sup>Th) against  $(^{238}U/^{232}Th)$ , rocks in which U and Th have remained unfractionated for more than 350,000 years will be in isotope equilibrium and therefore plot on the equiline on which  $(^{238}U/^{230}Th) = 1$ . Rocks in which U and Th have been fractionated within the past 350,000 years will be out of isotope equilibrium and will therefore plot away from the equiline. Many rocks from destructive plate margins, and particularly those with low  $(^{230}\text{Th})^{232}\text{Th})$  ratios, exhibit little or no  $^{238}\text{U-}^{230}\text{Th}$  disequilibrium (38). In contrast, the rocks that do exhibit <sup>238</sup>U-<sup>230</sup>Th disequilibrium tend to have high  $(^{230}\text{Th}/^{232}\text{Th})$  and U/Th ratios and lower abundances of Th and other incompatible elements (compare Fig. 1B and Fig. 2) (16, 17, 38). Island arc rocks exhibit a much greater range in Th isotope ratios than do mid-ocean ridge basalts (MORBs) or ocean island basalts (OIBs), which is consistent with their greater range in U/Th ratios (Fig. 1). MORBs and OIBs often have (238U/  $^{230}$ Th) <1 (they plot to the left of the equiline), and this disequilibrium is widely attributed to partial melting in the presence



**Fig. 1.** Plots of Ba/Th and U/Th ratios versus Th abundances for basalts and andesites from selected arc suites. For clarity, the data for the more incompatible element–depleted arcs of the Marianas (21, 22), South Sandwich Islands (24), Tonga-Kermadecs (23), and Vanuatu (25) have been grouped together (black circles), as have those from the more trace element–enriched arcs of the Aeolian Islands (26, 27), Indonesia (28–30), and the Philippines (31) (open triangles). Other, more intermediate data sets are from Japan-Karnchatka (34–36) and the Lesser Antilles (32, 33) (white diamonds). U/Th = 0.24 in the estimated GLOSS composition (19).

of residual garnet in their mantle source regions (39, 40). In contrast, many of the island arc rocks that are out of  $^{238}U^{-230}Th$  isotope equilibrium have ( $^{238}U/^{230}Th$ ) >1 (plot to the right of the equiline; Fig. 2).

The observation that <sup>238</sup>U-<sup>230</sup>Th isotope disequilibria are more common in arc rocks that are more depleted in incompatible elements indicates that the preservation of such short-lived disequilibria is linked to U and Th abundances. Thus, the controls on the development of  $^{238}\text{U-}^{230}\text{Th}$  disequilibria in arc rocks are different from those in MORBs and OIBs, and most authors have attributed them to the addition of material that was itself out of isotopic equilibrium (16, 17, 20, 32, 38, 41, 42). The depleted arc rocks have high U/Th ratios (Fig. 1), and many have high  $(^{238}U/^{230}Th)$ , which is consistent with the preferential mobilization of U in fluids from the subducted slab. Moreover, the overall data arrays in Fig. 1 and a plot of  $(^{238}U/^{230}Th)$  versus Th (38) can be modeled in terms of a broadly constant flux of U (and Ba) from the subducted slab, as that contribution is simply less significant in the rocks with higher U (and Ba) contents (17, 38).

More controversial is the cause of the high abundances of incompatible elements and lower Th isotope ratios in the rocks with lower  $(^{238}U/^{232}Th)$  (8, 17, 38). Interpretations are inextricably linked to the extent to which Th is mobilized from subducted oceanic crust and whether it is mobilized in the fluid or in some other component such as bulk sediment or partial melts of the subducted crust. The behavior of Th in this tectonic setting is illustrated by the variations of Sr/Th and Th/Ce ratios with variations in Sr and Nd isotopes (Fig. 3). Sr/Th ratios vary similarly to Ba/Th



**Fig. 2.** Plot of (<sup>230</sup>Th/<sup>232</sup>Th) versus (<sup>238</sup>U/<sup>232</sup>Th) for selected arc suites compared with fields for MORB and OIB taken from the literature. Symbols are as in Fig. 1, data are from (*16*, *17*, *21–23*, *30–33*, *67–69*), and the 30,000-year reference isochron (dashed line) is the minimum age consistent with the data from the Tonga-Kermadecs (*23*) and Marianas (*22*). Isotope ratios in parentheses are activity ratios, and activities are the decay rates of the radioactive isotopes.

ratios in island arc rocks (Fig. 1), and they may be used to constrain the mixing arrays for Sr and Th isotopes (Fig. 4). In a plot of Sr/Th ratios versus <sup>87</sup>Sr/<sup>86</sup>Sr, there is a striking hyperbola in which the more depleted arc rocks (those with low <sup>87</sup>Sr/<sup>86</sup>Sr) have high LILE/HFSE ratios, and the rocks with low Sr/Th and Ba/Th ratios and high Th contents (Fig. 1) tend to have high <sup>87</sup>Sr/ <sup>86</sup>Sr ratios. The observation that rocks with high LILE/HFSE ratios have low <sup>87</sup>Sr/<sup>86</sup>Sr ratios indicates that the fluid component also has relatively low <sup>87</sup>Sr/<sup>86</sup>Sr ratios of  $\sim$ 0.7035. This is surprising because the average Sr isotope values for altered oceanic crust sampled in ophiolites is  $\sim 0.7055$  (43) and on the ocean floor is  $\sim 0.7046$  (44). Subducted sediments, which should also be a fertile source of fluids from the subducted slab, typically have higher <sup>87</sup>Sr/<sup>86</sup>Sr [0.717 in the calculated Global Subducting Sediment composition (GLOSS) (19)]. The apparent discrepancy between the Sr isotope ratios of this inferred fluid component in arc rocks, and those in the likely sources of fluids in the downgoing slab, has led to suggestions that the LILE in the inferred fluid component may have exchanged with and in part been scavenged from the mantle wedge, which has low <sup>87</sup>Sr/<sup>86</sup>Sr (5, 45, 46).

The rocks with higher Th and other incompatible element abundances tend to have higher Sr isotope ratios (Fig. 3A), and in principle these compositions may be due to the presence of old trace element–enriched material in the mantle wedge or the



**Fig. 3.** (**A**) <sup>87</sup>Sr/<sup>86</sup>Sr versus Sr/Th and (**B**) <sup>143</sup>Nd/ <sup>144</sup>Nd versus Th/Ce for selected arc suites. Data sources and symbols are as in Fig. 1 and as discussed in the text; average MORB and OIB are from (*51*). PAAS, post-Archaean average shale (*52*); GLOSS, from (*19*).

addition of sediment from the subducted slab (3, 8-10, 47, 48). The plot of Th/Ce versus <sup>143</sup>Nd/<sup>144</sup>Nd highlights the behavior of Th in island arc rocks and constrains the nature of the enriched component (that is, with high Th and high 87Sr/86Sr). If Th were a relatively fluid mobile element in arcs, Th/Ce ratios should be highest in the rocks with high <sup>143</sup>Nd/<sup>144</sup>Nd ratios because, as outlined above, ratios of a more mobile element over a less mobile element (such as LILE/HFSE ratios) are higher in the rocks with the more depleted isotope ratios (low <sup>87</sup>Sr/<sup>86</sup>Sr and high <sup>143</sup>Nd/<sup>144</sup>Nd). However, Th/Ce ratios vary inversely with <sup>143</sup>Nd/ <sup>144</sup>Nd ratios, so that the rocks with high  $^{143}\ensuremath{\text{Nd}}\xspace{\ensuremath{\text{Nd}}\xspace}$  have low rather than high Th/Ce ratios (Fig. 3B). Thus, we conclude that Th is not preferentially mobilized in the fluids added to the mantle supplying arc magmas, which is consistent with the available experimental evidence (49, 50). Rocks with low <sup>143</sup>Nd/<sup>144</sup>Nd ratios also have high Th/Ce ratios, and whereas high Th/Ce ratios are rare in mantle-derived rocks [Th/Ce = 0.016 and 0.052 in average]MORB and OIB (51)], they are a feature of sediments with a substantial continental component [Th/Ce = 0.22 in average post-Archaean shale (52) and 0.12 in GLOSS (19)]. The low <sup>143</sup>Nd/<sup>144</sup>Nd rocks also have low U/Th ratios, and it is concluded that the shift to high Th/Ce and low U/Th ratios, high Th contents, and more enriched isotope ratios in these arc suites is primarily a result of contributions from subducted sediments [see also (9)] rather than an OIB component.

In summary, the isotope and trace element compositions of arc magmas require contributions from three components: (i) the upper mantle, (ii) (hydrous) fluids from the subducted crust, and (iii) subducted sediments (2, 7, 8-10). Uranium is preferentially mobilized in the fluid component, whereas Th appears only to be mobilized in the sediment component [high Th/Ce and low <sup>143</sup>Nd/<sup>144</sup>Nd (Fig. 3)]. This is consistent with the broad positive array between measured Th/U and <sup>208</sup>Pb\*/<sup>206</sup>Pb\*, which indicates that much of the Th/U variation observed in arc rocks is long lived (16, 17). Other evidence for contributions from subducted sediment comes from studies in which along-arc variations in the signatures of the volcanic rocks may be linked to those in sediments in the subducting plate (31, 47), from high <sup>10</sup>Be signatures in certain arc rocks (4, 20, 42), and from the correlations between fractionation-corrected incompatible element abundances in arc rocks and the amounts of sediment being subducted along different plate margins (9). A number of studies have used trace element ratios to argue that the sediment component is transferred as partial melts rather than bulk sediment (22, 23, 32, 53).

### Rates of Transfer

A robust feature of young sediments is that they contain <sup>10</sup>Be produced by spallation of oxygen and nitrogen in the atmosphere (54). Beryllium-10 is radioactive, with a half-life of 1.5 million years, and so the presence of <sup>10</sup>Be in island arc rocks is taken as compelling evidence for a contribution from subducted sediment (4, 55). The lack of <sup>10</sup>Be may reflect either the lack of a sediment contribution or the fact that the sediment was too old to retain any <sup>10</sup>Be.

Few studies have measured <sup>10</sup>Be and Th isotopes on the same samples, but in some arcs <sup>10</sup>Be abundances increase with (<sup>230</sup>Th/<sup>232</sup>Th) ratios (20, 41, 42). This relation suggests that the sediment signature as evidenced by <sup>10</sup>Be has high (<sup>230</sup>Th/<sup>232</sup>Th) ratios, and yet we have argued above that the sediment contribution in most arc rocks has low (<sup>230</sup>Th/<sup>232</sup>Th). However, the volcanic rocks that have high <sup>10</sup>Be contents also tend to have high B/Be ratios (4, 54): B is highly mobile, and so high B/Be ratios are

**Table 1.** Th, U, and  ${}^{230}$ Th/ ${}^{232}$ Th ratios determined on a Finnigan MAT262 with an RPQ-II. Total procedural blanks for U and Th were typically 100 and 50 pg, and the external reproducibility on U/Th and  ${}^{230}$ Th/ ${}^{232}$ Th ratios was  $\sim 1.0\%$  (2 $\sigma$ ). Repeat determinations of the ATHO Th standard yielded ( ${}^{230}$ Th/ ${}^{232}$ Th) = 1.017 ± 0.010 (2 $\sigma$ ). Sr and Nd isotope ratios for NBS 987 = 0.71022 ± 3 and J&M Nd = 0.511778 ± 24 (2 $\sigma$ ). For further details, see (32).

Sample	Locally	Age (years)	SiO <sub>2</sub>	Rb	Sr	Th	U	Nd	Sm	<sup>87</sup> Sr  <sup>86</sup> Sr	<sup>143</sup> Nd  <sup>144</sup> Nd	( <sup>230</sup> Th  <sup>232</sup> Th)	( <sup>238</sup> U  <sup>232</sup> Th)	( <sup>230</sup> Th  <sup>238</sup> U)
MLM-6	Merelava	<10.000	52.36	6	290	0.261	0.183	5.2	1.70	0.70415	0.51306	1.321	2.134	0.619
MLM-10a	Merelava	<10,000	51.38	5	126	0.280	0.171	3.9	1,40	0,70403	0.51305	1.359	1.857	0.732
AMB-26	Ambrym	<2,000	51.45	20	437	0.928	0.365	10.7	2.78	-	_	1.133	1.193	0.950
		,			Tonga	a-Kermade	ЭC							
T113cp	Tafahi	<10,000	52.84	1.3	145	0.175	0.097	1.25	0.46	0.70389	0.51293	1.200	1.686	0.718
TAF18/10	Tafahi	<10,000	52.35	1.8	132	0.097	0.052	1.72	0.55	0.70387	0.51295	1.297	1.643	0.796
26835	Tofua	1792–1959 A.D.	53.94	5.2	210	0.128	0.109	2.95	0.97	0.70345	0.51305	1.595	2.584	0.617
26837	Tofua	1792–1959 A.D.	53.54	6.1	226	0.143	0.134	3.70	1.35	0.70348	0.51305	1.677	2.842	0.595
26907	Tofua	1792–1959 A.D.	53.83	6.1	240	0.130	0.118	3.48	1.26	0.70340	0.51303	1.752	2.753	0.642
482-8-11	Ata	<10,000	50.40	9	243	0.318	0.154	5.18	1.41	0.70337	0.51304	1.310	1.470	0.898
482-8-12	Ata	<10,000	51.90	8	214	0.266	0.136	4.60	1.24	0.70344	0.51309	1.335	1.543	0.872
HHHTOP	Hunga Ha'apai	<2,000	55.35E	3.8	174	0.164	0.133	3.4	1.18	0.70368	0.51305	1.647	2.456	0.671
HHBTM	Hunga Ha'apai	<2,000	54.44	4.2	184	0.121	0.116	3.5	1.2	0.70371	0.51304	1.799	2.912	0.618
					Pł	nilippines								
B148	Bicol	<30,000	52.50	17	519	2.230	0.713	13.8	3.1	0.70385	0.51287	0.962	0.970	0.991
B107B	Bicol	<30,000	53.90	25	392	2.099	0.598	9.9	2.5	0.70375	0.51289	1.070	0.865	1.238
B220	Bicol	<30,000	58.30	60	602	10.795	2.766	31.0	6.2	0.70399	0.51294	0.793	0.777	1.020
1968	Bicol	1968 A.D.	54.74	21.1	713	1.768	0.572	19.6	4.3	0.70373	0.51287	1.003	0.982	1.022
1984	Bicol	1984 A.D.	54.70	19.9	714	2.033	0.603	19.5	4.3	0.70373	0.51291	0.887	0.900	0.985
				ŀ	Kamcha	atka-Aleut	ians							
A4-91	Avachinsky	1991 A.D.	55.90	21	359	0.639	0.349	10.7	2.97	0.70338	0.51305	1.262	1.659	0.767
TB-9-7-75	Tolbachik	1975 A.D.	49.70	25	296	0.527	0.336	12.1	3.32	0.70339	0.51311	1.877	1.948	0.972
T889	Tolbachik	1976 A.D.	50.40	39	314	1.134	0.707	20.2	4.96	0.70338	0.51308	1.816	1.891	0.968
J4662	Kizimen	1745 A.D.	55.30	20	349	1.312	0.763	12.3	3.25	0.70341	0.51305	1.739	1.765	0.994
Vil 4-92	Vilyuchinsky	2,000	58.90	20	462	1.646	0.746	16.3	3.8	0.70332	0.51297	1.236	1.375	0.907
C-11217	Ksudach	2,500	50.30	10	343	0.563	0.219	9.7	2.9	0.70336	0.51306	1.167	1.182	0.996
BAK32	Bakening	1495 A.D.	66.00	42.8	440	3.225	1.660	12.9	2.5	0.70338	0.51306	1.552	1.562	1.002
UM21	Umnak	1946 A.D.	52.24	16	360	1.174	0.610	10.9	2.9	0.70330	0.51299	1.286	1.576	0.823

evidently a feature of the fluid component that has high Ba/Th and U/Th ratios (56)(Fig. 1). One interpretation therefore is that the inferred slab-derived fluids, which have high LILE/HFSE ratios, may contain small amounts of <sup>10</sup>Be as well as high U/Th and  $(^{238}\text{U}/^{230}\text{Th})$  ratios. The implication is that the fluids contain a small contribution from subducted sediment, and they were transferred to the surface relatively rapidly because a number of young arc rocks still preserve U-Th isotope disequilibrium with high (<sup>238</sup>U/<sup>230</sup>Th) ratios (Fig. 2). In contrast, the component with low Th isotope and low U/Th ratios identified as sedimentderived on the basis of trace element and Sr and Nd isotope ratios, is characterized by low <sup>10</sup>Be/<sup>9</sup>Be ratios and <sup>238</sup>U-<sup>230</sup>Th equilibrium, and so it would appear to have taken substantially longer to have been transferred from the slab to the surface (several million years). An alternative explanation is that this component was derived from relatively old sediments in the subducted slab, and the rate of transfer of the sediment component is not constrained by the presence or absence of <sup>10</sup>Be.

As illustrated in Fig. 2, many arc rocks have higher  $(^{238}U/^{230}Th)$  ratios than those in MORBs and OIBs, and some exhibit <sup>238</sup>U-<sup>230</sup>Th isotope disequilibria, typically with  $(^{238}U/^{230}Th) > 1$ . The high U/Th ratios are presumed to be associated with subduction and to be due to the preferential mobilization of U in hydrous fluids from the subducted slab. If erupted magmas are in <sup>238</sup>U-<sup>230</sup>Th equilibrium, the time since the generation of their high U/Th ratios must be >350,000 years; but if they preserve <sup>238</sup>U-<sup>230</sup>Th disequilibrium, the timing can be more precisely determined. Such calculations require an estimate of the initial Th isotope ratios at the time of the develop-

**Fig. 4.** (<sup>230</sup>Th/<sup>232</sup>Th) versus <sup>87</sup>Sr/<sup>86</sup>Sr for the selected arc suites (*16*, *17*, *21–23*, *30–33*). Symbols are as in Fig. 1. Curve 1 is a mixing curve between MORB-type mantle wedge plus fluid and subducted sediment, as constrained from the hyperbolic relation between <sup>87</sup>Sr/<sup>86</sup>Sr and Sr/Th ratios in Fig. 3A. The wedge and fluid end-member is assumed to have (<sup>230</sup>Th/<sup>232</sup>Th) ratios similar to those of MORBs (in isotope equilibrium), and <sup>87</sup>Sr/<sup>86</sup>Sr = ~0.7034 from the array in Fig. 3A. The (<sup>230</sup>Th/<sup>232</sup>Th) of the sediment component is that at isotope equilibrium with average sediment U/Th (0.21 ± 0.05) and its <sup>87</sup>Sr/<sup>86</sup>Sr is taken to be 0.710, but that need not be precisely known in

ment of the relatively high U/Th ratios, and these can be constrained from (i) the variation in Sr and Th isotopes and (ii) linear arrays of bulk rock analyses on the <sup>238</sup>U-<sup>230</sup>Th equiline diagram.

Strontium/thorium ratios in arc rocks vary systematically with  $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$  ratios (Fig. 3A), and this trend can be used to evaluate the initial mixing arrays between Sr and Th isotopes. Data from arc rocks show no correlation between Sr and Th isotopes (Fig. 4), but that is to be expected because Th isotopes reflect changes in U/Th ratios associated with subduction on time scales of tens to hundreds of thousands of years, whereas the half-life for the decay of  ${}^{87}\text{Rb}$  to  ${}^{87}\text{Sr}$  is  $4.88 \times 10^{10}$  years. The data arrays in Figs. 1 and 2 suggest that at the time of U-Th fractionation, the Sr and Th isotope ratios primarily reflected mixing between the fluid, the mantle wedge, and subducted sediment. Both MORBs (and hence by inference depleted mantle in the wedge) and the fluid component have high Sr/Th ratios, albeit for different reasons. MORB-source mantle has high Sr/Th ratios  $(\sim 750)$  (51) because Sr is preferentially retained in clinopyroxene during melting, whereas the fluid component in arc rocks has high Sr/Th ratios because Sr is more mobile than Th in hydrous fluids (50) (Fig. 3A). A consequence is that the curvature of Sr-Th isotope mixing arrays between subducted sediments and contributions from the mantle wedge and the fluid are constrained irrespective of the relative contributions of the wedge and the fluid to the melt.

In detail, the relation between Th and Sr isotopes is complex, primarily because some arc rocks are in  $^{238}U^{-230}Th$  equilibrium and others are not, and because the U/Th and ( $^{230}Th/^{232}Th$ ) ratios of subducted



a high proportion of carbonate in the downgoing slab, such as Central America (19, 20). Nonetheless, most of the arc rocks plot between the curves of wedge plus fluid and sediment mixing (curve 1, Fig. 4) and their Th and Sr isotope ratios after 350,000 years (curve 2). Such mixing curves provide a minimum estimate of the Th isotope ratios at the time of U-Th fractionation, and for rocks with  $(^{238}\text{U}/^{230}\text{Th}) > 1$  they provide an upper limit on the age of that fractionation. The ages calculated are typically <120,000 years ago, and these are similar to those determined from suites of rocks from individual island arcs such as the Marianas (22) and Tonga-Kermadec (23), which are 30,000 to 50,000 years ago. Such results indicate that 30,000 to 120,000 years have elapsed since the fluid was released from the subducted slab and that average transport rates through the mantle wedge are  $\sim 1$  to 4 m/year. In practice, the release of hydrous fluids is likely to form amphibole peridotite, and so the rates of transfer are linked to the rates of convection in the mantle wedge (3). A recent study concluded that the fluid component would traverse just 1 to 2 km in 50,000 years, and that relatively hot mantle therefore lies close to the subducted slab at the depths of melt generation beneath island arcs (23, 57). Alternatively, fluid transfer may be by hydraulic fracture, and therefore much faster than that inferred from convection rates in the mantle wedge (58). Finally, some arc rocks preserve excess <sup>226</sup>Ra (16), but these cannot reflect the same processes as those responsible for the high (<sup>238</sup>U/<sup>230</sup>Th), because fractionated (<sup>226</sup>Ra/<sup>230</sup>Th) ratios return to isotope equilibrium in  $\sim$ 8000 years. Thus, the <sup>238</sup>U-<sup>230</sup>Th and <sup>226</sup>Ra-<sup>230</sup>Th disequilibria would appear to be decoupled in arc rocks, and the latter may reflect melt generation processes

sediments are likely to vary regionally. In

particular, carbonate-rich sediments have

high U/Th ratios, and so mixing arrays will

be different for arcs in areas where there is

### Implications and Conclusions

rather than the release of fluids from the

subducted slab (23).

The U and Th abundances and isotope ratios in arc rocks indicate that U, but not Th, is mobilized in fluids derived from the subducted slab. Variations in the Th contents of primary island arc magmas largely depend on the contribution from subducted sediments (Figs. 1 and 2) (9, 32), and so substantial amounts of Th and other HSFEs in arc rocks are derived from recycled crustal material in the sediments of the subducted slab. Th/Ta ratios appear to have been increased during partial melting of subducted sediments beneath some arcs

view of the shape of the Th-Sr mixing line. It is assumed that there is a constant fluid contribution of 0.14 ppm of U (*32*, *38*) and that that results in variable excess <sup>238</sup>U, depending on the proportion of sediment to fluid in each sample. The percentage of sediment is indicated along the lower curve, and these percentages will change if the low <sup>87</sup>Sr/<sup>86</sup>Sr wedge end member locally has higher incompatible element abundances than those in MORB-type mantle. With time, samples evolve vertically, and the upper curve (curve 2) illustrates the Th isotope ratios that develop from the resultant U/Th ratios after 350,000 years. The measured Th isotope ratio of each rock depends on the contribution from the subducted sediments and the value and age of their measured U/Th.

(22), but in general the distinctive high Th/Ta ratios of arc magmas are broadly similar to those in clay-rich sediments (19, 52). Thus, such high Th/Ta ratios may also reflect processes other than those associated with recent subduction-related magmatism, and high Th/Ta or Th/Nb ratios should provide more robust evidence for contributions from recycled sediments in OIBs than, for example, Nb/U ratios, which are more likely to have been modified during dehydration of the subducted slab (59, 60).

The fluid and sediment components in arc rocks, identified on the basis of trace element ratios, appear to be characterized by different transfer times through the mantle wedge. The fluid component contains U but little if any Th, and the resultant <sup>238</sup>U-<sup>230</sup>Th disequilibria indicate typical transfer times of 30,000 to 120,000 years (Figs. 2 and 4). In contrast, rocks with a greater contribution from subducted sediments [high Th/Ce and low <sup>143</sup>Nd/<sup>144</sup>Nd ratios (Fig. 3)] tend to have (<sup>238</sup>U/<sup>230</sup>Th) ratios ~1 and low <sup>10</sup>Be, which suggests transfer times of several million years.

The observation that the distinctive high LILE/HFSE ratios are best developed in the more depleted arc rocks is consistent with a similar flux of the more mobile elements in the fluid along different plate margins (5, 17). Condomines and Sigmarsson (38) estimated that for U such a flux might constitute 0.14  $\mu g/g$  of arc magma, and the intercepts of the straight line relations between fractionationcorrected trace element abundances in arc magmas and regional sediment fluxes (9) indicate that the size of the fluid contribution may be independent of the size of the sediment flux for U but not for Th. A recent estimate suggests that  $\sim$ 90% of the Th in an average arc composition is from subducted sediment, and perhaps only  $\sim$ 30% of the Th in subducted sediment is returned to the crust in arc magmas (61), which is consistent with previous estimates (53).

Bulk continental crust has a lower U/Th ratio (0.25) (62) than at least some estimates for the bulk Earth (0.26) (63) and the depleted upper mantle (0.39)(51). However, the island arc rocks with low U/Th ratios appear to have inherited those from subducted sediments, and arc rocks with a low sediment contribution have significantly higher U/Th ratios (average, 0.44; Fig. 1). Thus, the U/Th ratios of new crustal material generated along destructive plate margins are significantly higher than those of bulk continental crust. The low average U/Th ratios of bulk crust may primarily reflect different crustal generation processes in the Archaean,

when U would be less mobile because conditions were less oxidizing and when residual garnet may have had more of a role in crust generation processes (64, 65). The inferred fractionation of U and Th between the crust and the upper mantle requires the presence of residual garnet, as does the fractionation of Sm from Nd (66).

#### REFERENCES AND NOTES

- 1. T. Plank and C. Langmuir, *Earth Planet. Sci. Lett.* **90**, 349 (1988).
- R. M. Ellam and C. J. Hawkesworth, *Contrib. Mineral. Petrol.* 98, 72 (1988).
   J. H. Davies and D. J. Stevenson, *J. Geophys. Res.*
- J. T. Davies and D. J. Stevenson, J. Geophys. Res. 97, 2037 (1992).
   J. D. Morris, W. P. Leeman, F. Tera, *Nature* 344, 31
- 4. J. D. Morris, W. P. Leeman, F. Tera, *Nature* **344**, 3 (1990).
- C. J. Hawkesworth, K. Gallagher, J. M. Hergt, F. McDermott, *Philos. Trans. R. Soc. London Ser. A: Math. Phys. Sci.* 342, 179 (1993).
- M. T. McCulloch and J. Gamble, *Earth Planet Sci.* Lett. **102**, 358 (1991).
- D. M. Miller, S. L. Goldstein, C. H. Langmuir, *Nature* 368, 514 (1995).
- C. J. Hawkesworth, K. Gallagher, J. M. Hergt, F. McDermott, Annu. Rev. Earth Planet. Sci. 21, 175 (1993).
- 9. T. Plank and C. H. Langmuir, *Nature* **362**, 739 (1993).
- 10. J. A. Pearce and D. W. Peate, Annu. Rev. Earth Planet. Sci. 23, 251 (1995).
- 11. R. W. Kay, *J. Geol.* 88, 497 (1980).
- 12. D. McKenzie, Earth Planet Sci. Lett. 74, 81 (1985).
- 13. R. W. Williams and J. B. Gill, Geochim. Cosmochim.
- Acta **53**, 1607 (1989). 14. M. Spiegelman and T. Elliott, *Earth Planet. Sci. Lett*.
- 118, 1 (1993). 15. M. Condomines, C. Hemond, C. J. Allègre, *ibid.*, 90,
- 243 (1988). 16. J. B. Gill and R. W. Williams, *Geochim. Cosmochim.*
- Acta **54**, 1427 (1990). 17. F. McDermott and C. J. Hawkesworth, *Earth Planet.*
- *Sci. Lett.* **104**, 1 (1991).
- A. Zindler and S. Hart, Annu. Rev. Earth Planet. Sci. 14, 493 (1986).
- 19. T. Plank and C. H. Langmuir, Chem. Geol., in press.
- M. K. Reagan, J. D. Morris, E. A. Herrstrom, M. T. Murrell, *Geochim. Cosmochim. Acta*, **58**, 4199 (1994).
- D. W. Peate *et al.*, unpublished data; P. N. Lin, R. J. Stern, J. D. Morris, S. H. Bloomer, *Contrib. Mineral. Petrol.* **105**, 381 (1990).
- 22. T. Elliott, T. Plank, A. Zindler, W. White, B. Bourdon, J. Geophys. Res., in press.
- S. P. Turner et al., Trans. Am. Geophys. Union 76, F654 (1995); S. P. Turner et al., Geochim. Cosmochim. Acta, in preparation.
- J. A. Pearce, P. E. Baker, P. K. Harvey, I. W. Luff, J. Petrol. 36, 1073 (1995).
- 25. D. W. Peate et al., ibid., in press.
- 26. R. M. Ellam et al., Bull. Volcanol. 50, 386 (1988). 27. R. M. Ellam, C. J. Hawkesworth, M. A. Menzies, N.
- W. Rogers, J. Geophys. Res. 94, 4589 (1989).
   P. Z. Vroon, M. J. van Bergen, G. J. Klaver, W. M.
- P. Z. Vroon, M. J. van Bergen, G. J. Nave, W. M. White, *Geochim. Cosmochim. Acta* 59, 2573 (1995).
   P. Z. Vroon, M. J. van Bergen, W. M. White, J. G.
- Varekamp, *J. Geophys. Res.* **98**, B12, 22349 (1996). 30. S. P. Turner, unpublished data.
- 31. F. McDermott, M. J. Defant, C. J. Hawkesworth, R.
- C. Maury, *Contrib. Mineral. Petrol.* **113**, 9 (1993). 32. S. Turner *et al.*, *Earth Planet. Sci. Lett.* **142**, 191
- (1996).
- E. Heath, R. Macdonald, H. Belkin, C. J. Hawkesworth, H. Sigurdsson, in preparation; E. Heath, thesis, Lancaster University, Lancaster, UK, in preparation.

- A. Hunter and S. Blake, J. Petrol. 36, 1579 (1995).
   A. Hunter, thesis, The Open University, Milton
- Keynes, UK (1994).
  36. S. P. Turner, C. J. Hawkesworth, F. McDermott, P. Kepezhinskas, *Eos* 76, F537 (1995); S. P. Turner *et al.*, in preparation.
- J. Gill, Orogenic Andesites and Plate Tectonics (Springer-Verlag, Berlin, 1981).
- M. Condomines and O. Sigmarsson, Geochim. Cosmochim. Acta 57, 4491 (1993).
- 39. P. Beattie, Nature 363, 63 (1993).
- T. Z. LaTourrette, A. K. Kennedy, G. J. Wasserburg, Science 261, 739 (1993).
- O. Sigmarsson, M. Condomines, J. D. Morris, R. S. Harmon, *Nature* **346**, 163 (1990).
- 42. J. B. Gill, J. D. Morris, R. W. Johnson, *Geochim. Cosmochim. Acta* **57**, 4269 (1993).
- M. J. Bickle and D. A. H. Teagle, *Earth Planet. Sci.* Lett. **113**, 219 (1992).
- H. Staudigel, T. Plank, W. White, H.-U. Schminke, AGU Geophys. Monogr. 96, 19 (1996).
- 45. R. J. Arculus and R. Powell, *J. Geophys. Res.* **91**, 5913 (1986).
- E. Stolper and S. Newman, *Earth Planet. Sci. Lett.* 121, 293 (1994).
- 47. W. M. White and B. Dupré, *J. Geophys. Res.* 91, 5927 (1986).
- 48. J. D. Morris and S. R. Hart, *Geochim. Cosmochim. Acta* **47**, 2015 (1983).
- 49. E. H. Bailey and K. V. Ragnarsdottir, *Earth Planet*. *Sci. Lett.* **124**, 119 (1994).
- 50. H. Keppler, Nature 380, 237 (1996)
- 51. S. S. Sun and W. F. McDonough, *Geol. Soc. Spec. Publ.* **42**, 313 (1989).
- S. R. Taylor and S. M. McLennan, *The Continental Crust: Its Composition and Evolution* (Blackwell Science, Oxford 1985).
- 53. T. Plank and C. H. Langmuir, Eos 73, 637 (1992).
- 54. J. D. Morris, Annu. Rev. Earth Planet. Sci. 19, 313 (1991).
- J. G. Ryan and C. H. Langmuir, *Geochim. Cosmo-chim. Acta* 52, 237 (1988).
- 56. \_\_\_\_\_, *ibid.* **57**, 1489 (1993).
- 57. S. P. Turner *et al.*, *Geol. Soc. Aust. Abstr.* **45**, 95 (1997).
- 58. J. H. Davies and A. Rowland, ibid., p. 17.
- 59. A. W. Hofmann, K. P. Jochum, M. Senfert, W. M. White, *Earth Planet. Sci. Lett.* **79**, 33 (1986).
- 60. S. Turner, C. J. Hawkesworth, N. W. Rogers, P. King, Chem. Geol., in press.
- 61. C. J. Hawkesworth, S. Turner, D. Peate, F. McDermott, P. van Calsteren, *ibid.*, in press.
- R. L. Rudnick and D. M. Fountain, *Rev. Geophys.* 33, 267 (1995).
- C. J. Allègre, B. Dupré, E. Lewin, Chem. Geol. 56, 219 (1986).
- 64. M. T. McCulloch, *Earth Planet. Sci. Lett.* **115**, 89 (1993).
- R. M. Ellam, C. J. Hawkesworth, F. McDermott, *Chem. Geol.* 83, 165 (1990).
- R. K. O'Nions and D. P. McKenzie, *Earth Planet. Sci.* Lett. **90**, 449 (1988).
- 67. C. Hemond, thesis, University of Paris 7 (1986).
- G. Capaldi, M. Cortini, R. Pece, *Isotope Geosci.* 1, 39 (1983).
- K. H. Rubin *et al.*, *J. Volcanol. Geotherm. Res.* 38, 215 (1989).
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