PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Element Fluxes Associated with Subduction Related Magmatism

C. J. Hawkesworth, J. M. Hergt, R. M. Ellam and F. Mc Dermott

Phil. Trans. R. Soc. Lond. A 1991 **335**, 393-405
doi: 10.1098/rsta.1991.0054

Element fluxes associated with subduction related magmatism

By C. J. Hawkesworth[1], J. M. Hergt[1], R. M. Ellam[2] and F. McDermott[1]

[1] *Department of Earth Sciences, The Open University, Milton Keynes MK7 6AA, U.K.*
[2] *Scottish Universities Research and Reactor Centre, East Kilbride, Glasgow G75 0QU, U.K.*

Destructive plate margin magmas may be subdivided into two groups on the basis of their rare earth element (REE) ratios. Most island arc suites have low Ce/Yb, and remarkably restricted isotope ratios of $^{87}Sr/^{86}Sr = 0.7033$, $^{143}Nd/^{144}Nd = 0.51302$, $^{206}Pb/^{204}Pb = 18.76$, $^{207}Pb/^{204}Pb = 15.57$, and $^{208}Pb/^{204}Pb = 38.4$. However, they also have Rb/Sr (0.03), Th/U (2.2) and Ce/Yb (8.5) ratios which are significantly less than accepted estimates for the bulk continental crust. The high Ce/Yb suites have higher incompatible element contents, more restricted heavy REE, and much more variable isotope ratios. Such rocks are found in the Aeolian Islands, Grenada, Indonesia and Philippines, and their isotope and trace element features have been attributed both to contributions from subducted sediment, and/or old trace element enriched material in the mantle wedge. It is argued that for isotope and trace element models the slab component can usefully be taken to consist of subducted sediment and altered mid-ocean ridge basalts, since these may contain *ca.* 80 % of the water in the subducted slab, and the distinctive trace element features of arc magmas are generally attributed to the movement of material in hydrous fluids. The isotope data indicate that not more than 15 % of the Sr and Th in an average arc magma were derived from subducted material, and that the rest were derived from the mantle wedge. The fluxes of elements which cannot be characterized isotopically are more difficult to constrain, but for most minor and trace elements the slab derived contribution in arc magmas is too small to have a noticeable effect on the residual slab.

1. Introduction

Destructive plate margins are major sites of terrestrial magmatism which have long had a key role in most models for the generation of continental crust and the development of chemical heterogeneities in the upper mantle. However, several studies have recently emphasized that the net flux of material from the mantle to the crust above recent subduction zones is both more basaltic, and it has lower Rb/Sr and Th/U, than most estimates of the bulk composition of the continental crust (Kay & Kay 1986; Kushiro 1987; Ellam & Hawkesworth 1988; Ellam *et al.* 1990). Others have argued that the inferred fractionation in Sm/Nd between the mantle and the crust cannot be achieved in this tectonic setting, and that such fractionation primarily reflects the addition of small degree melts, generated in within plate environments, to the continental crust (O'Nions & McKenzie 1989). A high priority

Phil. Trans. R. Soc. Lond. A (1991) **335**, 393–405
Printed in Great Britain

is therefore to establish the size and nature of the element fluxes between the mantle and the crust in different tectonic settings, and destructive plate margins retain a unique position because that is where new continental crust is generated, and where crustal material is recycled into the upper mantle. This contribution reviews selected data on subduction related magmas, and what are early tentative steps towards evaluating the fluxes of different elements in magmas generated along destructive plate margins (after Kay 1980). Such fluxes in turn constrain the composition of material returned to the deep mantle, and hence the likely contribution of subducted crust in the generation of oceanic basalts.

Radiogenic isotopes are sensitive tracers for the different materials which may contribute in the generation of magmatic rocks, and in the last few years there have been a number of detailed geochemical and isotope studies on subduction related rocks. Several minor and trace element features of subduction related rocks remain consistently different from those of mid-ocean ridge basalts (MORB) and ocean-island basalts (OIB), and in particular they are characterized by high (large ion lithophile)/(high field strength elements) (LIL/HFS) element ratios, and relatively low Nb, Ta, and perhaps Ti abundances (Pearce 1982). In contrast, the majority of destructive plate margin rocks have Nd, Sr and Pb isotope ratios which are broadly similar to those of OIB (Morris & Hart 1983), and to those MORB which have more enriched isotope signatures (higher $^{87}Sr/^{86}Sr$ and lower $^{143}Nd/^{144}Nd$). None the less, some subduction related rocks exhibit steep arrays on Pb isotope diagrams (Kay *et al.* 1978; Woodhead & Fraser 1985; White & Dupré 1986), consistent with the introduction of radiogenic Pb from subducted sediments, and more recently it has been demonstrated that young arc rocks often exhibit high ^{10}Be which requires a contribution from young (less than 5 Ma) sedimentary material, presumably in the subducted slab (Tera *et al.* 1986; Morris *et al.* 1990). U/Th ratios are highly fractionated in both altered MORB and in sedimentary carbonates, and so contributions from each material in young arc rocks should also be readily detected by Th isotope studies (Gill & Williams 1990; McDermott & Hawkesworth 1991). In general, however, the isotope data on subduction related rocks indicate that the contributions from sediments and altered oceanic crust in the subducted slab are typically much less than estimates of the 'subduction component' calculated on the basis of minor and trace element variations (Pearce 1983). It is this discrepancy which lies at the heart of recent debates on the processes responsible for the isotope and trace element signatures of subduction related magmas, and which is critical in any attempt to estimate element fluxes in this tectonic environment.

2. Minor and trace element variations

(*a*) *Rare earth elements*

The rare earth elements (REE) are widely used in models of petrogenesis and earth evolution, because they are a geochemically coherent group and they include the radioactive decay scheme of ^{147}Sm to ^{143}Nd. Figure 1 summarizes Ce and Yb variations in destructive plate margin rocks, and the data broadly fall into two groups: one in which Ce and Yb vary together, and a second group characterized by much higher Ce contents at similar Yb. Such differences require major differences in either the REE profiles of the source rocks for the two groups, and/or in the bulk distribution coefficients and the degree of melting. Rocks in the high Ce/Yb group have consistently higher $^{87}Sr/^{86}Sr$ ratios (figure 1*b*), and so the simplest interpretation

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

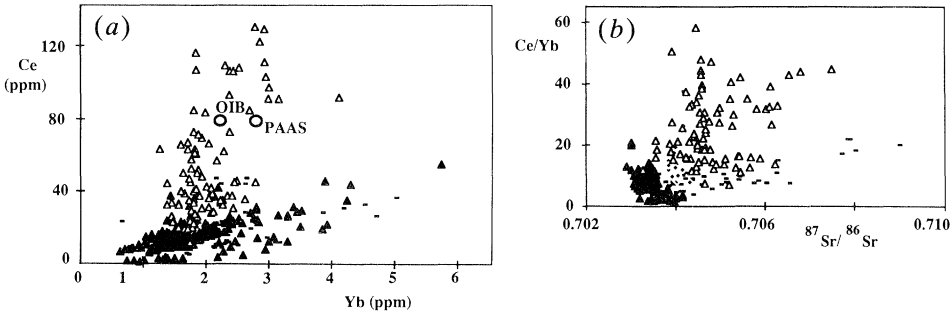

Figure 1. (*a*) Ce and Yb contents of destructive margin magmas with not more than 62 % SiO_2, compared with those for average ocean island basalt (OIB) from Sun & McDonough (1989), and post-Archaean average shale (PAAS, Taylor & McLennan 1989). The symbols illustrate that the arc rocks tend to fall into low and high Ce/Yb groups: △, Aeolian Islands, Grenada, Indonesia, and the Philippines; ▲, Aleutians, Manam Island, Marianas, New Britain, S. Andes, S. Sandwich Islands, Tonga–Kermadecs; –, Lesser Antilles, apart from Grenada. (*b*) Ce/Yb vs. $^{87}Sr/^{86}Sr$ for arc magmas, illustrating the restricted range in $^{87}Sr/^{86}Sr$ in the low Ce/Yb suites, and the much greater range in $^{87}Sr/^{86}Sr$ in the high Ce/Yb rocks. The symbols are as in (*a*).

is that they contain at least a contribution from material which was both old (i.e. old enough to have developed different isotope ratios), and light REE enriched. Such features have been attributed both to subducted sediments and to trace element enriched source regions in the mantle wedge.

The paradox of subduction related magmas is that the classic island arc suites, which appear to represent widespread and relatively simple products of destructive plate margin magmatism, do not have some of the key trace element features of the bulk continental crust. In the data base used here these suites are represented by rocks from Tonka-Kermadec, the Marianas, Manam Island, New Britain, the Aleutians, the South Sandwich Islands, and the Northern Lesser Antilles; they tend to have relatively restricted radiogenic isotope ratios (figures 1*b* and 3), and both Ce and Yb vary together (figure 1*a*). However, they have average Rb/Sr (0.03), Th/U (2.15), and Ce/Yb (8.5) which are all less than most estimates of the average continental crust (0.12, 3.8 and 15, respectively, Taylor & McLennan 1985). Note also that estimates of such element ratios in the bulk crust are consistent with the observed isotope variations in both crustal and mantle rocks widely used in Earth evolution models (Allègre 1982).

In general, the rocks with higher Ce/Yb in figure 1*a* also have higher Rb/Sr and Th/U ratios. Thus the destructive margin rocks which have trace element ratios more typical of the bulk crust are those which are slightly unusual in that they have initial isotope ratios indicating that they already contain trace element enriched material. If such material is subducted sediment, there is clearly some danger of concluding that subduction related magmas only have the trace element ratios of continental crust when they contain a significant contribution from pre-existing crust, in the form of subducted sediment. In some areas the high Ce/Yb ratios may reflect old trace element enriched source regions in the mantle wedge, and in that model the element ratios which are similar to those in the continental crust were set up by the introduction of small degree melts to the mantle wedge before the onset of subduction.

In summary, the low Ce/Yb group appears to be typical of the simpler island arc systems, and that is the one we focus on in seeking to evaluate the element fluxes in

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

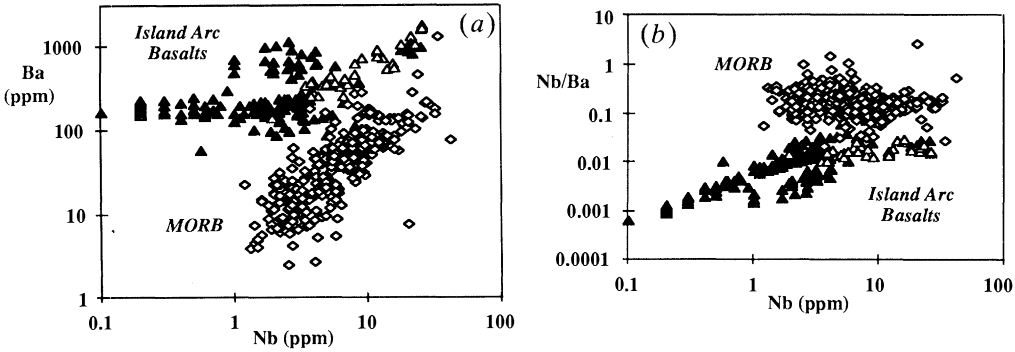

Figure 2. Ba, Nb, and Nb/Ba variations in destructive plate margin and MOR basalts. All samples have not more than 53% SiO_2: ▲, △, island arc rocks with Ce/Yb ⩽ 15 and ⩾ 15, respectively (see figure 1); ◇, samples of MORB.

subduction zone magmas. The higher Ce/Yb rocks are often developed in more complex tectonic areas, and their trace element signatures are considered elsewhere (Hawkesworth *et al.* 1991).

(b) LIL and HFS elements

Figure 2 contrasts the variation of a typical LIL (Ba) and HFS (Nb) element in MOR and subduction related basalts. Any discussion of LIL/HFS element fractionation processes in the upper mantle requires rocks which have been relatively little affected by low pressure fractionation of Ti oxides, and yet Ti oxides start to crystallize at significantly different SiO_2 contents in tholeiitic and calc alkaline suites. All the samples selected to evaluate LIL/HFSE ratios have less than 53% SiO_2, and consideration of the individual suites indicates that while some of them show the effects of Ti oxide fractionation the effects are small compared with the overall trends.

On the plot of Ba against Nb the MOR basalts define a positive array, consistent with suggestions that Ba and Nb have similar bulk distribution coefficients during partial melting in the upper mantle. The subduction related basalts are displaced to high Ba/Nb ratios, and their field has a much shallower slope than that for MORB (figure 2*a*). It is noticeable that Ba is relatively constant at low Nb contents, and this could only be ascribed to partial melting if the bulk D for Ba is *ca.* 1 during melting above subduction zones. This is regarded as unlikely, and so the preferred interpretation is that the almost constant Ba abundances reflect the addition of Ba-rich material associated with subduction. Moreover, the amount of Ba which has been added would appear to have been relatively constant at these different destructive plate margins.

A consequence of the relatively constant Ba contents in low Nb arc rocks is that their distinctive high LIL/HFS element ratios (in this case high Ba/Nb) are best developed in rocks with low HFS element abundances (figure 2*b*). Secondly, the proportions of an element from subducted material and the mantle wedge depend on how much its abundance varies in the wedge, as well on the size of the slab derived flux. Ba can be much more depleted in the wedge (see the MORB array in figure 2) than, for example, Sr, at least while clinopyroxene is present. Thus the ratio of subduction related Sr, to mantle wedge derived Sr, in arc rocks will tend to be less than the same ratio for Ba.

In summary, it is useful to identify three ways in which elements and isotopes appear to behave in subduction related rocks.

(i) Some elements are thought to be derived entirely from the mantle wedge, and therefore to reflect partial melting processes and the pre-subduction trace element contents of the mantle wedge (e.g. Nb, Ta, Ti).

(ii) There is then a group of predominantly LIL elements which exhibit relatively high abundances in subduction related rocks, but the resulting high LIL/HFSE ratios are best developed in rocks with low HFS element contents (i.e. Ba, K, Th, Sr).

(iii) There are also a number of tracers which appear to be so sensitive to the contribution from the subducted slab, that their abundances in arc rocks may be largely independent of the contribution from the mantle wedge, for example, ^{10}Be, ^{207}Pb, and perhaps highly incompatible trace elements, such as B (Morris *et al.* 1990; White & Dupré 1986).

A key point to emerge from such considerations of minor and trace elements in subduction related rocks is that the relative contributions of subducted material and the mantle wedge are very different for different elements. Thus in most models Nb and Ti, for example, are derived wholly from the mantle wedge, whereas Ba and B may be largely derived from the introduction of material which is related to subduction. In between there are a number of elements in which the inferred contribution from subducted material and the mantle wedge may vary significantly.

3. Radiogenic isotopes

Subduction related magmas may contain contributions from fresh and hydrothermally altered oceanic crust, subducted sediments, and variably enriched or depleted material in the mantle wedge. These tend to exhibit at least some distinctive parent/daughter trace element ratios, and so with time they will be characterized by distinctive radiogenic isotope signatures (table 1). Moreover, radiogenic isotopes are generally regarded as highly sensitive tracers in the identification of different source components in magmatic rocks. None the less, the general observation is that for Sr, Nd, Pb, and Th isotopes, the isotope ratios measured in low Ce/Yb subduction related rocks are surprisingly similar to those in ocean island basalts (Morris & Hart 1983).

(*a*) Sr, Nd *and* Pb *isotopes*

Figure 3 summarizes much of the available Nd and Sr isotope data on subduction related rocks, with different symbols for the high and low Ce/Yb suites identified in figure 1. The low Ce/Yb suites have remarkably restricted $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ with average values of 0.7033 ($\pm$0.0002) and 0.51302 ($\pm$0.00004) (one s.d.), and most of the observed isotope variations are in the high Ce/Yb rock suites of Grenada, the Aeolian Islands, Philippines and Indonesia. Subduction related rocks are characterized by high Sr/Nd ratios which suggests that material derived from the subducted oceanic crust has high Sr/Nd (Pearce 1983; Ellam & Hawkesworth 1988). Mixing between such high Sr/Nd material and a lower Sr/Nd end-member from, for example, the mantle wedge should result in curved mixing lines on figure 3, and in many suites these have not been observed. One exception is in the rocks from Tonga where $^{87}Sr/^{86}Sr$ does vary with LIL/HFS element ratios, and which suggests $^{87}Sr/^{86}Sr$ in the high Sr/Nd component in that area is 0.7042 (Ewart & Hawkesworth 1987).

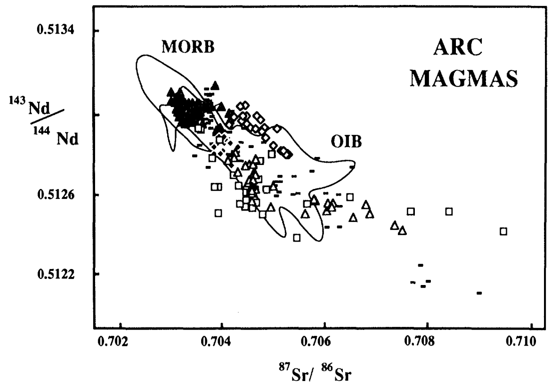

Figure 3. Nd and Sr isotope ratios in destructive margin rocks, compared with the fields for MORB and OIB, after Zindler & Hart (1986). The samples denoted by filled triangles, horizontal bars, and the filled diamonds are as for figure 1; and the open symbols are for the high Ce/Yb arc rocks: □, the Philippines; △, Aeolian Islands; ◇, Grenada.

Table 1.

	average arc basalt	melt from depleted mantle	PAWMS	PATS	altered MORB	altered MORB and sediment[b]	slab contribution[d]
Rb	9.7	0.34	3.6	160	9.04	10.4	13
Ba	183	3.79	1338	650	16.85	76	56
Th	0.78	0.07	0.23	14.6	0.072	0.22	0.08
U	0.36	0.03	0.05	3.1	0.321	0.338	0.09
Nb	1.4	1.4	1.25	19	2.8	2.9	0
La	5.1	1.5	25.8	38	3.3	4.5	1.7
Ce	11.9	4.5	9.6	80	12.7	13.3	5.0
Pb	2.0	0.18	10	20	0.3	0.89	0.62
Sr	407	54	500[a]	200	118	134	63
Nd	8.7	4.4	19.3	32	6.8	7.5	2.8
Zr	46	45	21.6	210	100	98	14
Y	18	17	46.3	27	42	42	12
Yb	1.5	1.8	5.6	2.8	3.8	3.8	0.9
H_2O		0.17	(6.3)	(6.3)	2.7		
$^{87}Sr/^{86}Sr$	0.7034	0.7028	0.710[a]	0.717	0.7046	0.7056	
$^{143}Nd/^{144}Nd$	0.51302	0.51300	0.5123	0.5123	0.51308	0.51303	
$^{206}Pb/^{204}Pb$	18.76	18.46	18.7	18.9	18.5	18.68	
$^{207}Pb/^{204}Pb$	15.57	15.49	15.7	15.7	15.5	15.64	
$^{208}Pb/^{204}Pb$	38.37	38.0	38.9	38.9		38.59	
$(^{230}Th/^{232}Th)$	1.3	1.3	0.66	0.6	13	4.4[c]	

[a] Values modified in the light of other sediment data (Ben Othman *et al.* 1989).
[b] Calculated as 1 part PATS, 4 parts PAWMS, and 95 parts altered MORB.
[c] Assuming U/Th fractionation does not occur until the depth of magma generation.
[d] Calculated contribution from subducted sediment and altered MORB, assuming that the relative mobilities depend on ionic radius/charge (after Tatsumi *et al.* 1986).

The Pb contents of oceanic sediments are much higher than those in basic magmas, and they tend to have relatively high $^{207}Pb/^{204}Pb$ (Ben Othman *et al.* 1989). Thus Pb isotopes have been widely used to evaluate contributions from subducted sediments in arc magmas (Kay *et al.* 1978; Woodhead & Fraser 1985; White & Dupré 1986).

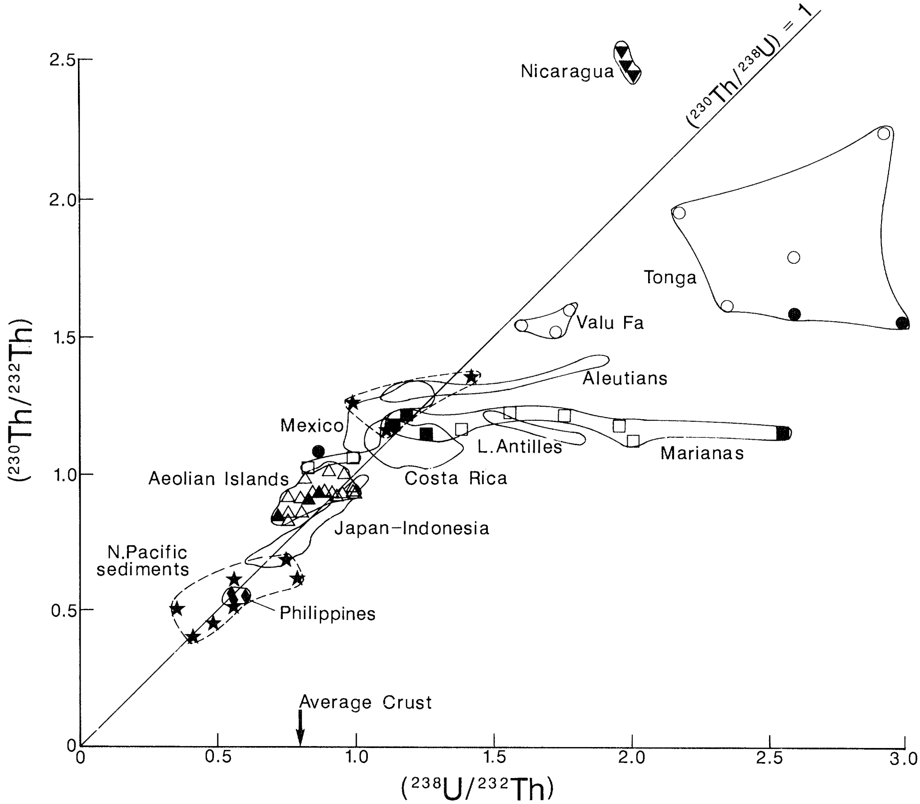

Figure 4. A $(^{230}Th/^{232}Th)$–$(^{238}U/^{232}Th)$ diagram summarizing the available results for subduction related magmas and N. Pacific sediments (after Gill & Williams 1990; McDermott & Hawkesworth 1991). The overall range in $(^{230}Th/^{232}Th)$ is similar to that in MORB and OIB, but destructive margin rocks are unusual in that some of them are displaced to relatively high $(^{238}U/^{230}Th)$. Note that the majority of arc rocks have $(^{238}U/^{232}Th)$ values which are significantly higher than most estimates of the bulk continental crust (Taylor & McLennan 1985). The dashed fields enclosing the 'star' symbols are N. Pacific sediments.

However, the marked Pb isotope variations are again largely confined to the high Ce/Yb suites, and the lower Ce/Yb rocks have a restricted range and an average Pb isotope composition of $^{206}Pb/^{204}Pb = 18.76$ (± 0.13), $^{207}Pb/^{204}Pb = 15.57$ (± 0.02), and $^{208}Pb/^{204}Pb = 38.4$ (± 0.18). Zindler & Hart (1986) noted the high frequency of Nd and Sr isotope ratios in intraplate oceanic basalts at about 0.5130 and 0.7033, and suggested that they reflected the existence of a mantle component which they termed 'prevalent mantle' (PREMA). Given the diversity of materials which may contribute to arc magmatism, it is surprising to note that the average of the low Ce/Yb arc rocks is indistinguishable from PREMA for Nd and Sr isotopes, and similar for Pb (PREMA has Pb isotope ratios of *ca.* 18.3, 15.46, and 37.9).

(*b*) Th *isotopes*

^{230}Th is generated within the natural decay chain from ^{238}U to ^{206}Pb, and it has a half-life of 75200 years. In secular equilibrium $(^{230}Th) = (^{238}U)$ (the parentheses denote activities), and equilibrium is restored within *ca.* 300000 years of any change in Th/U. The combination of Th and Pb isotopes therefore offers a powerful new

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

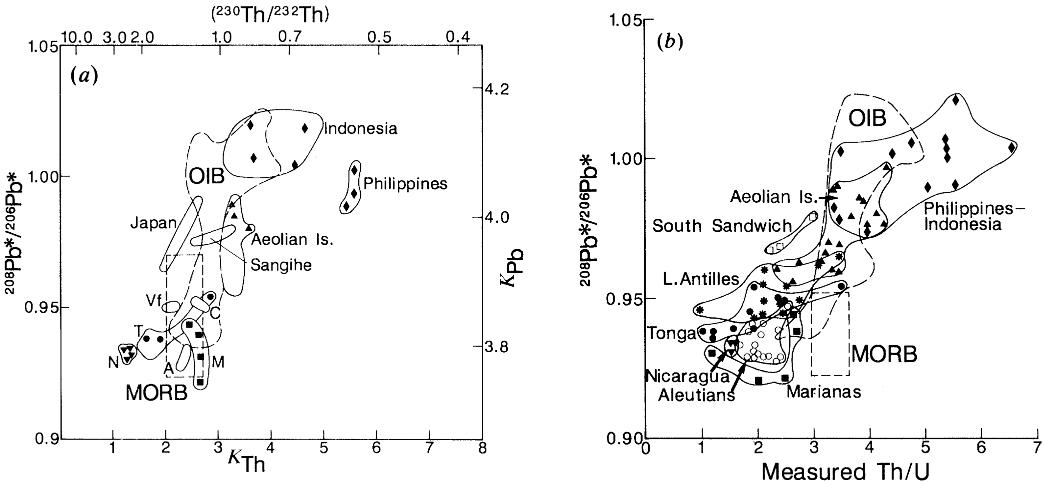

Figure 5. (*a*) $^{208}Pb^*/^{206}Pb^*$ against Th/U calculated from ($^{230}Th/^{232}Th$), which is taken to represent Th/U in the source of the magmas before any recent fractionation of Th/U. $^{208}Pb^*/^{206}Pb^*$ is the ratio of $^{208}Pb/^{204}Pb$ to $^{206}Pb/^{204}Pb$ relative to Canyon Diablo troilite (after Allègre *et al.* 1986), and so it reflects Th/U which have persisted for a minimum of several 100 Ma. Data symbols as in figure 4, N, Nicaragua; T, Tonga; A, Aleutians; M, Marianas; Vf. Valu fa; C, Costa Rica. (*b*) $^{208}Pb^*/^{206}Pb^*$ against measured Th/U ratios in the basalts. In both diagrams there is a striking positive correlation between $^{208}Pb^*/^{206}Pb^*$ and Th/U, and particularly in (*a*) the arc rocks plot in a similar array to MORB and OIB (McDermott & Hawkesworth 1991).

approach to investigate changes in Th/U over time scales ranging from tens of thousands to billions of years (Condomines *et al.* 1988; Gill & Williams 1990; McDermott & Hawkesworth 1991). Moreover, they are particularly suited to the study of arc magmas because Th/U ratios are highly fractionated in both altered MORB (Hart & Staudigel 1989) and marine carbonates (Chen *et al.* 1986) which are presumably present in the subducted oceanic crust.

On a ($^{230}Th/^{232}Th$)–($^{238}U/^{232}Th$) diagram samples in secular equilibrium plot on the equiline (figure 4). Many young MORB and OIB are displaced to the left of the equiline, indicating that U/Th in the liquid is less than that in the source during partial melting in the upper mantle (Condomines *et al.* 1988). Some island arc rocks are unusual in that they plot to the right of the equiline, and this has been regarded as a feature of subduction related magmatism (Allègre & Condomines 1982). In more detail it is clear that the high ($^{230}Th/^{232}Th$) values, and any significant displacement to high U/Th, occur in the low Ce/Yb arc suites (figure 1). The high Ce/Yb rocks have lower U/Th, similar to that of the bulk crust, and more of them are in secular equilibrium. The range in Th isotopes in arc rocks is similar to that in MORB and OIB (Gill & Williams 1990; McDermott & Hawkesworth 1991), and this contrasts with the very high values of ($^{230}Th/^{232}Th$) $\geqslant 10$ which may be inferred for altered MORB and marine carbonates from their measured U/Th ratios (Hart & Staudigel 1989).

Pb isotopes, expressed as $^{208}Pb^*/^{206}Pb^*$ because that reflects variations in Th/U on the time scale of 100 s of Ma (Allègre *et al.* 1986), correlate with both measured Th/U and κ_{Th} calculated from ($^{230}Th/^{232}Th$) (figure 5). Thus the major variation in Th/U in subduction related rocks is between different arc systems, it broadly correlates with Ce/Yb, and it has been present for long enough to affect the Pb isotope ratios. As indicated above, such relatively long-lived chemical differences may either be present in the mantle wedge, and/or reflect contributions from sediments with old source ages, but the key point is that these old variations in Th/U are both larger, and they appear to reflect different processes, than the displacement to low Th/U (high $^{238}U/^{232}Th$) observed in some arc rocks (figure 4).

Finally in this section, we note that, as with Nd and Sr isotopes (figure 3), the Th and Pb isotope ratios of many arc rocks plot within the mantle array defined by MORB and OIB (figure 5). This similarity between many of the isotope ratios of subduction related rocks, and those of MORB and OIB, appears to be a general feature, and it suggests either (i) the various components which might contribute to the generation of destructive margin magmas mix together in such proportions that the resulting isotope ratios are fortuitously similar to those in OIB; (ii) ocean island basalts contain a significant contribution from subducted oceanic crust recycled at subduction zones, and so it is to be expected that subduction related and ocean island basalts have similar isotope signatures; or (iii) the contribution of subducted materials in destructive plate margin magmas is much less than that implied by models in which the 'excess' LILE contents are derived from the subducted slab, and instead the ocean island-like isotope signature of many destructive margin magmas is a feature of the mantle wedge. The latter in turn implies that some of the isotope characteristics of ocean island basalts are also derived from relatively shallow levels in the Earth's mantle.

4. Discussion

Several studies have sought to estimate the relative contributions of the mantle wedge and subducted slab in the generation of new crust, and it is now accepted that those relative contributions are different for different elements. In his pioneering work Kay (1980) argued that the excess K responsible for the high K/REE ratios in arc magmas was derived from subducted material, and that in the case of K as much as 90% was probably of continental origin via the oceans. Pearce (1983) extended that approach and proposed that the excess LIL elements responsible for the high LIL/Nb ratios in arc rocks were from the subduction zone, and he estimated that 70–90% of the Sr, Th, Ba, Rb and K in a typical Chilean basalt could have been derived from subducted material. Such calculations are discussed further below (see figure 7), but the problem is very simply how these high values for the slab contribution can be reconciled with the radiogenic isotope data which for the most part is similar to those from many OIB (e.g. figures 3 and 5). One solution was to argue that the slab derived flux beneath different arcs was relatively constant, and that it was best estimated in areas where the mantle wedge was highly depleted (Hawkesworth & Ellam 1989). In such areas all the measured LILE contents of the arc magmas might be inferred to have been derived from the slab, and the relative contribution of such a flux in an estimate of average new crust was sufficiently small (not more than 25%) that it could be reconciled with the available isotope data.

The isotope ratios of different materials that are likely to be in the mantle wedge and the subducted slab are now sufficiently well known that in principle isotope-trace-element diagrams can be used to evaluate the sources of different elements in arc magmas. Subducted related magmas are characterized by high Sr/Nd and Ba/Nb ratios and, as illustrated for Ba–Nb (figure 2), the high ratios are best developed in low Nd and Nb rocks. A graph of $^{87}Sr/^{86}Sr$ against Nd/Sr (figure 6*a*) demonstrates that the distinctive high Sr/Nd component in arc magmas tends to have low $^{87}Sr/^{86}Sr$ of *ca.* 0.704, and that the high $^{87}Sr/^{86}Sr$ ratios of some arc rocks are associated with low Sr/Nd. Similarly, Pb isotope ratios expressed as Δ7/4 (Hart

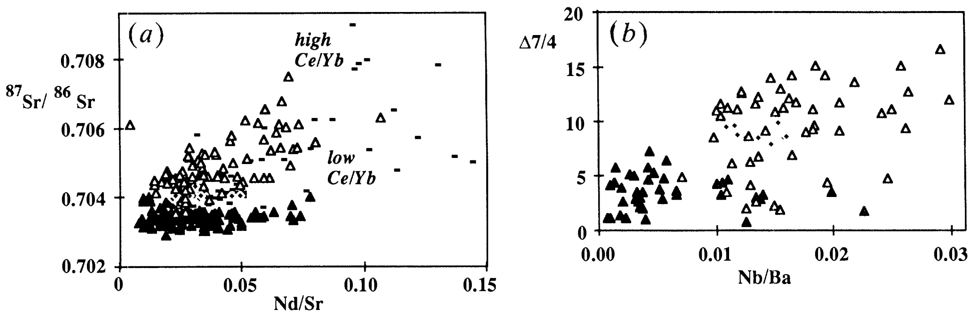

Figure 6. (*a*) $^{87}Sr/^{86}Sr$ against Nd/Sr, and (*b*) Δ7/4 vs. Nb/Ba, for arc rocks with not more than 62 % SiO_2. The symbols are as for figure 1, and Δ7/4 is the size of the displacement in $^{207}Pb/^{204}Pb$ above the Northern Hemisphere Reference Line for oceanic basalts, after Hart (1984). The rocks with the higher LIL/HFS element ratios (i.e. low Nd/Sr and Nb/Ba) have the less radiogenic Sr and Pb isotope ratios.

1984) have been widely regarded as sensitive indicators of contributions from subducted sediments, but the rocks with high Δ7/4 values which might suggest a larger sediment contribution tend to have low rather than high Ba/Nb (figure 6*b*). Thus in both these examples the more extreme isotope signatures are *not* associated with the distinctive high LIL/HFSE signature of the arc rocks, but instead the latter appears to have relatively unradiogenic Sr and Pb isotope ratios. For Pb isotopes this conclusion is particularly surprising, because the low Δ7/4 and Nb/Ba occur in the rocks with the lowest incompatible element contents, and they should therefore be most sensitive to any contribution from subducted sediment.

The proportion of different rock types in the subducted slab is difficult to constrain, but a reasonable estimate may be 20 % altered MORB, 1 % sediment, with the rest being unaltered MORB and gabbro. For isotope and trace element ratios the distinctive material is the altered MORB and sediments, and it is most unlikely that contributions from unaltered MORB in the subducted slab and MORB-type mantle in the overlying wedge can be readily distinguished. Unaltered MORB contains relatively little H_2O, and since the distinctive LIL/HFS element ratios of arc magmas appear to be linked to the release of hydrous fluids (Tatsumi *et al.* 1986), the slab contribution in arc magmas will be dominated by those from altered MORB and sediment, which contain *ca.* 80 % of the H_2O in the subducted oceanic crust (table 1). Thus, for the purposes of this discussion we shall simply investigate the contribution of altered MORB and subducted sediment in arc magmas.

Table 1 summarizes the isotope ratios and trace element abundances in altered MORB, subducted sediment, and an average arc basalt calculated for those rocks with $\leqslant 3$ p.p.m. Nb, and/or $\leqslant 65$ p.p.m. Zr. The estimated isotope ratios of the subduction component were used to calculate the maximum contribution from such material in the average arc magma, assuming that the isotope ratios of the arc magma reflect mixing between the subduction component and a depleted MORB-type magma. Some isotope ratios are more sensitive to the size of the slab contribution than others, because that depends on the differences between the isotope ratios and the element abundances in the slab component, the depleted mantle and the arc magma. Thus the slab contributions calculated from Nd and even Pb isotopes, are much less well constrained than those for Sr and Th isotopes, which are 15 and 5 % respectively.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

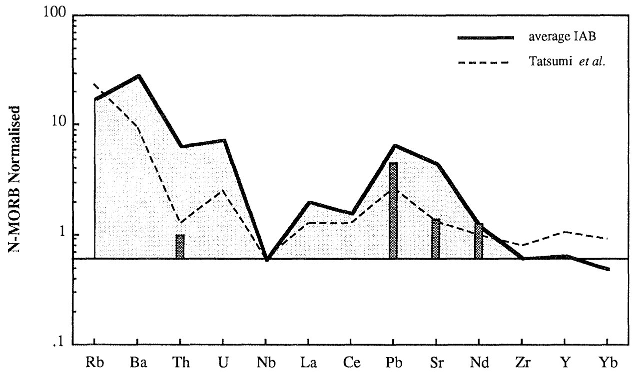

Figure 7. A MORB-normalized trace element diagram illustrating the composition of the average low Nb, low Ce/Yb arc basalt in table 1, and the maximum contribution of Th, Pb, Sr and Nd from altered MORB and sediment in the subducted slab, calculated from the isotope data. The shaded area is the slab contribution calculated by the method of Pearce (1983). The dashed line is the estimated slab contribution, from altered MORB and subducted sediment (table 1), which is consistent with both the isotope data and the relative mobilities of different trace elements during dehydration (Tatsumi *et al.* 1986).

Figure 7 presents the MORB-normalized trace element pattern of the average low Nb, low Ce/Yb arc magma in table 1. The trace element models suggests that the MORB-normalized element contents which are greater than that for Nb, Ta, and probably Zr and Ti, i.e. the shaded area in figure 7, are derived from subducted material (Pearce 1983). This indicates that 98% of the Ba, 90% of the Th, and Pb, 85% of the Sr, and 50% of the Nd in the average arc magma are derived from the slab, and yet for some elements these figures are much higher than the maximum slab contribution calculated from the isotope data (see also figure 7). One way to resolve the discrepancy might be to argue that the rest of the slab derived contribution inferred from the trace elements was from unaltered MORB, since this wasn't considered in the isotope calculations. However, for Th that would require unaltered MORB, which neither contains much water, nor presumably has any slab derived hydrous fluid fluxing through it, to be the source of *ca.* 90% of the Th in the arc magma.

Tatsumi *et al.* (1986) published some experimental results which indicate that the mobility of minor and trace elements during dehydration of serpentine depends largely on their ionic radios, although the conclusions drawn from our use of their data are not changed if we use the ratio of ionic radius to charge. Significantly, the differences in the ionic radii predict that Th will be less readily mobilized from the subducted slab during dehydration than Nd, and that Sr and Pb are more readily mobilized, in a way that is entirely consistent with the maximum slab contributions estimated from the isotope data (figure 7). Thus, the relationship between ionic radius/charge and mobility described by Tatsumi *et al.* (1986) can be used to calculate the possible contribution from subducted sediment and altered MORB (as in table 1) to the trace element budget of the arc magma, and that is illustrated in figure 7. The results are in reasonable agreement with the maximum slab contributions estimated from the isotope data, but the relative concentrations of Rb, Ba and K, for example, are much higher than those observed in the more depleted arcs, such as Tonga.

5. Summary

The radiogenic isotope ratios of the typical low Ce/Yb arc suites are very restricted relative to the range observed in oceanic basalts, and sediment and altered MORB in the subducted oceanic crust. It is argued that contributions from altered MORB and sediment will dominate the slab derived flux in arc magmas, and that the isotope data suggests that this represents not more than 15% of the Sr and the Th in an average arc magma. Such values are very much less than the estimates from trace element data alone, typically 85–90% respectively, but they are broadly consistent with earlier suggestions that the typical slab derived flux is best estimated in areas where the mantle wedge was highly depleted in incompatible trace elements before subduction (Hawkesworth & Ellam 1989). It is much more difficult to constrain the slab contribution for trace elements which cannot be characterized isotopically, and for the LIL elements significantly different results are obtained using the experimental data of Tatsumi *et al.* (1986), or observing the LIL element abundances in depleted arcs (Hawkesworth & Ellam 1989). None the less, both estimates require rather small proportions of the trace element inventory of the subducted lithosphere to be implicated in the production of the IAB trace element signature. Consequently, residual slab material subducted beyond the arc environment, and often invoked as OIB-source material, is relatively trace element rich and only marginally affected by removal of the slab flux, preserving more or less MORB-like incompatible trace element ratios.

We thank Julie Morris, Nick Rogers and Anthony Ware for their comments on earlier versions of the type, and many stimulating discussions on the problems of arc magmatism. The diagrams were prepared by Andrew Lloyd and the typescript by Janet Dryden.

References

Allègre, C. J. & Condomines, M. 1982 Basalt genesis and mantle structure studied through Th isotope geochemistry. *Nature* **299**, 21–24.

Allègre, C. J. 1982 Chemical geodynamics. *Tectonophysics* **81**, 109–132.

Allègre, C. J., Dupré, B. & Lewin, E. 1986 Thorium/uranium ratio of the Earth. *Chem. Geol.* **56**, 219–227.

Ben Othman, D., White, W. M. & Patchett, J. 1989 The geochemistry of marine sediments, island arc magma genesis, and crust-mantle recycling. *Earth planet. Sci. Lett.* **94**, 1–21.

Chen, J. N., Edwards, R. L. & Wasserburg, G. J. 1986 ^{238}U, ^{234}U and ^{232}Th in seawater. *Earth planet. Sci. Lett.* **80**, 241–251.

Condomines, M., Hemond, Ch. & Allègre, C. J. 1988 U–Th–Ra radioactive disequilibria and magmatic processes. *Earth planet. Sci. Lett.* **90**, 243–262.

Ellam, R. M. & Hawkesworth, C. J. 1988 Is average continental crust generated at subduction zones? *Geology* **16**, 314–317.

Ellam, R. M. & Hawkesworth, C. J. 1988 Elemental and isotope variations in subduction related basalts: evidence for a three component model. *Contrib. Mineral. Petrol.* **98**, 72–80.

Ellam, R. M., Hawkesworth, C. J. & McDermott, F. 1990 Pb isotope data from the Proterozoic subduction-related rocks: implications for crust-mantle evolution. *Chem. Geol.* **83**, 165–181.

Ewart, A. W. & Hawkesworth, C. J. 1987 The Pleistocene to Recent Tonga: Kermadec arc lavas: interpretation of new isotope and rare earth element data in terms of a depleted mantle source model. *J. Petrology* **28**, 495–530.

Gill, J. B. & Williams, R. W. 1990 Th isotopes and U-series studies of subduction-related volcanic rocks. *Geochim. cosmochim. Acta* **54**, 1427–1442.

Hart, S. R. & Staudigel, H. 1989 Isotope characterisation and identification of recycled components. In *Crust/mantle recycling at convergence zones* (ed. S. R. Hart & L. Gülen), pp. 15–28. Nato Workshop Volume, Kluwer Academic Publishers.

Hart, S. R. 1984 A large-scale isotope anomaly in the Southern Hemisphere mantle. *Nature* **309**, 753–757.

Hawkesworth, C. J. & Ellam, R. M. 1989 Chemical fluxes and wedge replacement rates along Recent destructive plate margins. *Geology* **17**, 46–49.

Hawkesworth, C. J., Hergt, J. M., McDermott, F. & Ellam, R. M. 1991 Destructive margin magmatism and contributions from the mantle wedge and subducted crust. *Spec. Publ. Geol. Soc. Austr.* (In the press.)

Hole, M. J., Saunders, A. D., Marriner, G. F. & Tarney, J. 1984 Subduction of pelagic sediments: implications for the origin of Ce-anomalous basalts from the Mariana Islands. *J. geol. Soc. Lond.* **141**, 453–472.

Kay, R. W. 1980 Volcanic arc magmas: implications of a melting–mixing model for element recycling in the crust–upper mantle system. *J. Geol.* **88**, 497–522.

Kay, R. W., Sun, S. S. & Lee-Hu, C. N. 1978 Pb and Sr isotopes in volcanic rocks from Aleutian Islands and Pribilof Islands, Alaska. *Geochim. cosmochim. Acta* **42**, 263–273.

Kay, R. W. & Kay, S. M. 1986 Petrology and geochemistry of the lower continental crust: an overview. In *The nature of the lower continental crust* (ed. J. B. Dawson, D. A. Carswell, J. Hall & K. H. Wedepohl), pp. 147–159. *Geol. Soc. Lond. Spec. Publ.* **24**.

Kushiro, I. 1987 A petrological model of the mantle wedge and lower crust in the Japanese island arcs. *Geochem. Soc. Spec. Pub.* **1**, 165–181.

McDermott, F. & Hawkesworth, C. J. 1991 Th, Pb and Sr isotope variations in young arc volcanics and oceanic sediments. *Earth planet. Sci. Lett.* (In the press.)

Morris, J. D. & Hart, S. R. 1983 Isotopic and incompatible trace element constraints on the genesis of island arc volcanics from Cold Bay and Amak Island, Aleutians, and implications for mantle structure. *Geochim. cosmochim. Acta* **47**, 2015–2030.

Morris, J. D., Leeman, W. P. & Tera, F. 1990 The subducted component in island arc lavas: constraints from Be isotopes and B–Be systematics. *Nature* **344**, 31–36.

O'Nions, R. K. & McKenzie, D. P. 1988 Melting and continent generation. *Earth planet. Sci. Lett.* **90**, 449–456.

Pearce, J. A. 1982 Trace element characteristics of lavas from destructive plate boundaries. In *Andesites: orogenic andesites and related rocks* (ed. R. S. Thorpe), pp. 525–548. New York: Wiley.

Pearce, J. A. 1983 Role of the sub-continental lithosphere in magma genesis at active continental margins. In *Continental basalts and mantle xenoliths* (ed. C. J. Hawkesworth & M. J. Norry), pp. 230–249. Nantwich, Cheshire: Shiva.

Sun, S. S. & McDonough, W. F. 1989 Chemical and isotopic systematics of oceanic basalts: implications for mantle composition and processes. In *Magmatism in ocean basins* (ed. A. D. Saunders & M. J. Norry), pp. 313–345. *Geol. Soc. Spec. Publ.* **42**.

Tatsumi, Y., Hamilton, D. L. & Nesbitt, R. W. 1986 Chemical characteristics of fluid phase released from a subducted lithosphere and origin of arc magmas: evidence from high-pressure experiments and natural rocks. *J. Volcanol. geotherm. Res.* **29**, 293–309.

Taylor, S. R. & McLennan, S. M. 1985 *The continental crust: its composition and evolution.* (312 pages.) Oxford: Blackwell.

Tera, F., Brown, L., Morris, J., Sacks, I. S., Klein, J. & Middleton, R. 1986 Sediment incorporation in island arc magmas: inferences from ^{10}Be. *Geochim. cosmochim. Acta* **50**, 535–550.

White, W. M. & Dupré, B. 1986 Sediments subduction and magma genesis in the Lesser Antilles: isotopic and trace element constraints. *J. geophys. Res.* **91**, B6, 5927–5941.

Woodhead, J. D. & Fraser, D. G. 1985 Pb, Sr and ^{10}Be isotopic studies of volcanic rocks from the northern Mariana Island. Implications for magma genesis and crustal recycling in the Western Pacific. *Geochim. cosmochim. Acta* **49**, 1925–1930.

Zindler, A. & Hart, S. 1986 Chemical geodynamics. *A. Rev. Earth planet. Sci.* **14**, 493–571.