Evaluation of REE mobility in low-grade metabasalts using mass-balance calculations

JOHN M. BARTLEY

Some studies of rare-earth element (REE) patterns of altered basalts have suggested that REE may be mobile during alteration. When possible effects of total rock mass balance are considered, most published data suggest that (1) only La is required to have been mobile, (2) if other REE have been mobile, changes are within analytical error and do not correlate with mineralogical or major-element changes during alteration, and (3) at least some other 'immobile' elements, such as Ti, Y, and Zr, may have been mobile in the rocks considered here. REE, excluding La, may be among the most reliable geochemical indicators of the compositional affinities of altered basalts.

J. M. Bartley, Department of Geology, University of North Carolina, Chapel Hill, North Carolina 27514, USA. Present address: Department of Geology and Geophysics, University of Utah, Salt Lake City, Utah 84112, USA.

A major goal in analytical geochemistry in recent years has been to develop petrochemical techniques for discriminating between basalts erupted in different geologic and tectonic settings. The ancient rocks to which such petrochemical discriminants might be applied are commonly hydrothermally altered or otherwise metamorphosed. Thus, attempts have been made to identify those elements which are relatively immobile during hydrothermal metamorphism, and thus may preserve primary characteristics of the basalts (e.g., Pearce & Cann 1973, Floyd & Winchester 1975). In general, those elements deemed most immobile occur as small, highly charged ions, including Ti, Zr, Y, Hf, and the rare-earth elements (REE). While the reliability of some of these geochemical discriminants has been questioned (Floyd & Winchester 1975, Vallance 1975, Morrison 1978), they have been widely applied in the Scandinavian Caledonides and other orogens to both fresh and altered mafic volcanic rocks (e.g., Gale & Roberts 1974, Gale & Pearce 1980, Stephens 1981, Furnes et al. 1982, Boyd 1983).

The concept of an immobile element in metamorphism is problematic, because an ion may or may not appear to have moved depending on the reference frame used. In this paper, the reference frame is a rock volume defined by primary structures of the rock; an immobile element is one that does not move with respect to such things as vesicles, lithologic contacts, and so forth. Such an element may nonetheless change its concentration during metamorphism, because concentration reflects only the amount of an element relative to all others in the rock (Barth 1948, Gresens 1967). If significant amounts of major elements are added or subtracted during metamorphism, the effects of this upon concentrations of other elements must be evaluated before attempting either to evaluate whether a particular element has been immobile during metamorphism, or to interpret 'immobile' element concentrations in altered volcanic rocks.

A number of studies have inferred that REE may be mobile in basalts during hydrothermal metamorphism (Thompson 1973, Wood et al. 1976, Menzies et al. 1977, Hellman & Henderson 1977, Hellmann et al. 1979, Loeschke & Schock 1980), casting doubt on the use of REE in interpreting the genesis of altered basalts. These studies base their conclusions on changes of REE concentrations and on ratios of La to other elements, but did not explicitly evaluate effects of total-rock mass balance. The authors remarked on the striking coherence of the behavior of REE (Hellman et al. 1979, Walsh 1983), but ascribed this to similar chemical behavior of REE. However, this is also the predicted result if REE were immobile and their concentrations changed only because of a net loss or gain of other elements. Hydrothermal experiments on greenstones by Menzies et al. (1979) documented such an effect. Total REE concentrations varied slightly because varying amounts of major elements were pre-

cipitated from or dissolved by the hydrothermal solution, but no movement of REE was documented.

This paper examines possible mass-balance effects in some published REE data from altered basalts. The significance of the calculations is limited by not knowing actual volume changes during metamorphism (and not being able to estimate this precisely because rock densities were not measured), by the paucity of data from suites which include both genuinely fresh and altered basalts, and by analytical uncertainties of the REE concentrations. Within these limitations, it appears that of the REE only La is required to have been mobile. Concentration changes shown by other REE can be adequately explained as indirect effects of major-element mobility. While the data do not exclude the possibility that greater mobility has occurred, they do require that (1) any movement must have been remarkably uniform, and (2) changes in the concentrations of REE other than La do not correlate with major-element changes in the same rocks.

Data selection

Mass-balance calculations were made using published analyses selected to have the following characteristics:

a. Field data must allow reasonable certainty that primary compositions of variably altered rocks to be compared were essentially the same. The samples compared came either from a small volume (< 1 m^3) of a single flow, or from a single pillow. This still does not preclude the possibility of primary REE variations as a consequence of local crystal abundance or fractionation. However, common phenocryst phases in basalts all have very low partition coefficients for REE, and thus the bulk of the REE can be assumed to be in the matrix. Therefore, any primary REE differences from this source will amount to dilution of the matrix by the phenocrysts, and will be indistinguishable from changes in REE concentrations by metasomatic addition and subtraction of major elements. Such primary differences would therefore affect estimated volume changes during alteration, but should not affect the results of the test for evidence of mobility used here (see below).

b. Both major and trace element concentrations must have been determined on the same samples, so that complete analyses could be compared.

c. REE must have been determined using a reasonably precise technique. All concentrations used in this paper were determined by instrumental neutron activation analysis (INAA).

Two sets of published analyses that satisfied these criteria were used in the calculations: (1) a basalt flow metamorphosed in the prehnite-pumpellyite facies form the Cliefden outcrop in the Ordovician Walli Andesite of New South Wales, Australia (Smith 1968, Hellman et al. 1979); and (2) greenschist-facies metabasalt pillows from the Ordovician Bymark greenstones, which are assigned to the Støren Group (sensu lato) of south-central Norway (Loeschke 1976, Loeschke & Schock 1980, Furnes & Roberts 1980). Representative trace-element analyses for the Cliefden rocks (Smith & Smith 1976, Hellman et al. 1979) are shown in Table 1. Typical chondrite-normalized REE patterns from both suites are shown in Fig. 1.

The Cliefden outcrop is probably the most completely described metabasalt exposure in the world. Smith (1968) mapped the outcrop on a grid of 2-foot (61 cm) squares, defining seven different types of mineralogical and textural domains (characteristic domain width 30 cm). The domains reflect different degrees of alteration that produced complementary Na-enriched spilitic rocks (gray domains of Smith 1968) and Ca-enriched dimineralic rocks composed of either pumpellyite + quarz or epidote + quartz (yellow-green and green domains), which are locally amygdaloidal. Rocks transitional between these extremes were placed in gray-green domains. Smith (1968) determined major element concentrations in 61 samples from the outcrop, and Hellman et al. (1979) reported REE concentrations of 11 of the same samples. Smith & Smith (1976) analyzed selected minor and trace elements, including Ti, Zr and Y. Two groups of particularly closely spaced samples, one of five and the other of three, satisfied the criteria above. Analyses from these groups allow examination of REE behavior between five of Smith's domain types. For field and petrographic descriptions of the various domains, the reader is referred to Smith (1968).

The Bymark greenstone pillow lavas were metamorphosed at conditions transitional between the prehnite-pumpellyite and greenschist facies (Loeschke 1976). The pillows analyzed preserve clear primary textures (Fig. 2 in Loeschke 1976),
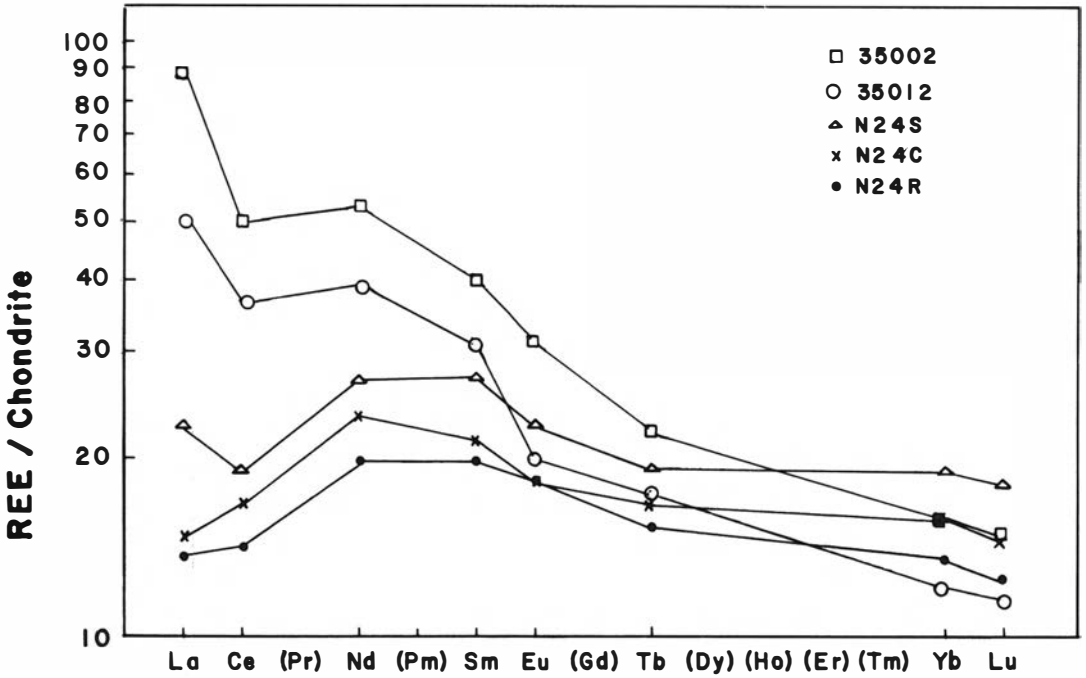
Fig. 1. Examples of chondrite-normalized rare-earth element patterns from variably altered metabasalts from the Cliefden outcrop (sample 35002: gray domain, sample 35012: green domain) and Bymark greenstones (N21C: pillow core, N21R: inner rim, N21S: pillow selvedge). Data were taken from Hellman et al. (1979) and Loeschke & Schock (1980).

which suggests that volume changes during alteration exceeding a few tens of percent are unlikely. Loeschke (1976) determined major-element concentrations in the massive pillow cores, epidote-rich inner rims, and chlorite-rich outer selvedges of four pillows. Loeschke & Schock (1980) determined REE in the same four pillows.

Neither of these suites includes genuinely fresh rocks. However, this lack of data comparing genuinely fresh to altered rocks does not affect one objective of this paper, which is to evaluate to what extent present data from natural rocks demonstrate mobility of REE during low-grade metamorphism in basalts.

Mass-balance calculations

Mass-balance calculations were done using the method of Gresens (1967), which considers the amount of each element or oxide that enters or departs from an initial rock volume. The basic approach requires no assumptions about the chemical behavior of any element, so that any particular hypothesis (i.e., immobility of REE) can be tested. The mass-balance equation for metasomatic changes from original composition A to final composition B is

$$\Delta x_i = W (K_v K_\varrho x_i^B - x_i^A)$$

where

Δx_i = grams of element i gained or lost from W grams of the initial rock
W = mass of initial rock volume
K_v = ratio of final to initial volume
K_ϱ = ratio of final to initial density
x_i^A = concentration of element i in rock A

For any concentration change, this equation gives the amount of any element added to or subtracted from the original rock if changes of volume and density are known. Densities are rarely determined as part of geochemical analysis. For the purpose of determining if a set of REE data requires that those elements were mobile during metasomatism, this can be avoided by normalizing the volume ratio to the density ratio ($\bar{K} = K_v K_\varrho$) so that $\Sigma x_i = (\bar{K}_v x_i^A)$.

The approach, dictated by a lack of specific

Table 1. Representative minor and trace-element analyses for Cliefden outcrop metabasalts, taken from Smith & Smith (1976), and Hellman et al. (1979). Sample 35002 is a Na-rich albitic spilite; 35002 is a Ca-enriched, pumpellyite metabasalt. Al concentrations are in ppm, except TiO_2, which is in weight percent.

	35002	35012
La	29.4	17.7
Ce	44.5	35.3
Nd	32.1	24.5
Sm	7.26	5.62
Eu	2.16	1.41
Tb	1.03	0.78
Yb	3.13	2.29
Lu	0.50	0.38
TiO_2 (wt.%)	0.88	0.71
Zr	79	70
Y	32	24

gravity measurements, is less precise, but the real volume change is virtually always unconstrained anyway. Even with known densities, the Gresens mass-balance equation does not determine volume change, but only gives the amounts of elements added to or substracted from a rock volume given an assumed volume change, or the volume change required if a particular element is assumed to have been immobile. Only a large density change with alteration would have a quantitatively important effect on results of the mass-balance calculations. In the present case, spilites tend to decrease in density by up to about 5% with increasing alteration (e.g., Vallance 1975, Table 1). This means that estimated values of $\bar{K}_v$ in this paper may be as much as 5% less than the actual value of K_v for conditions assumed in the calculation. Preservation of primary textures without great modification of their typical size in fresh lavas probably restricts acceptable volume changes to a maximum of 20–30%.

The hypothesis that element j is immobile can be tested by determining $\bar{K}_v$ for $\Delta x_j = 0$, then using that value of $\bar{K}_v$ to calculate Δx_i for all elements in the rock. One must then decide if the calculated volume-composition relationships are geologically and geochemically acceptable.

The calculations reported here estimated $\bar{K}_v$ by computing the geometric mean of the $\bar{K}_v$ values required for perfect isochemical behavior of each of the REE. Values of Δx_i were then computed for the elements assumed immobile using this estimate of $\bar{K}_v$. If the differences between calculated and observed Δx_i consistently exceeded

analytical error for an element, it was deleted from the group of elements assumed immobile and the estimated $\bar{K}_v$ was recalculated. Values of Δx_i were then computed for all elements using this estimate of $\bar{K}_v$. Results were then evaluated by considering whether the volume and chemical changes thus inferred are consistent with field and petrographic observations.

Results

Results of calculations for the Cliefden outcrop and the Bymark greenstones are shown in Tables 2 and 3, respectively.

Cliefden outcrop

Spilitic gray domains were used as reference compositions because they best preserve primary textures and mineralogy. However, the resulting computed metasomatic changes cannot be interpreted to reflect directly the actual metasomatic changes in the rocks. This would imply that the Na-rich spilites had been converted by metasomatism into the yellow-green and green Ca-rich domains. This is probably incorrect; Smith (1968) argues convincingly from petrologic evidence
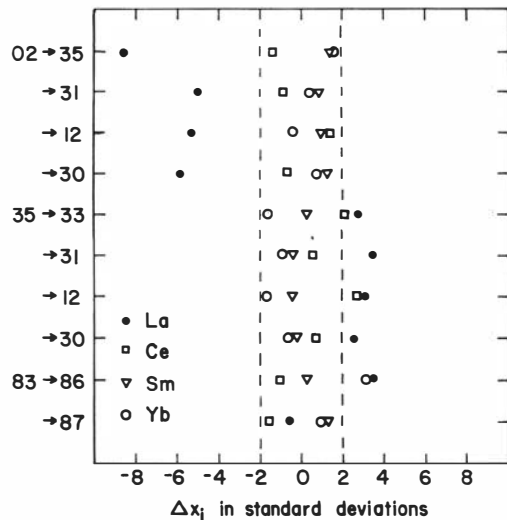

Fig. 2. Computed net changes between samples from the Cliefden outcrop of selected REE (see Table 1). Changes are expressed in number of standard deviations, as determined for each element by Hellman et al. (1979) by 20 replicate analyses of BCR-1. All changes are calculated passing from spilitic gray domains to Ca-enriched gray-green, yellow-green, green and amygdaloidal domains.

	GROUP 1								GROUP 2		
	02→35	02→31	02→12	02→30	35→33	35→31	35→12	35→30	83→86	83→87	SD
$\bar{K}_v$	1.04	1.22	1.34	1.36	0.93	1.18	1.30	1.31	1.08	1.06	
La	-30.9	-18.0	-19.2	-21.2	10.3	12.5	11.5	9.4	-12.8	-1.7	3.6
Ce	-6.4	-3.7	6.5	-2.9	10.1	2.7	12.6	3.4	-4.6	-7.0	4.6
Nd	-2.9	3.9	2.5	-0.4	5.2	6.6	5.3	2.4	9.0	8.9	6.4
Sm	5.3	3.4	3.9	4.9	1.1	-1.7	-1.2	0.4	1.1	5.3	3.7
Eu	-7.4	-7.6	-12.4	-14.0	-0.0	-0.1	-4.6	-6.4	-3.7	-2.1	6.4
Tb	3.6	6.3	1.7	6.6	-6.9	2.8	-1.7	2.8	-9.9	-3.1	6.7
Yb	6.2	1.9	-1.8	3.5	-7.3	-4.1	-7.6	-2.7	15.0	5.0	4.5
Lu	3.6	-2.6	2.0	5.7	-1.3	-5.9	-1.3	2.0	-2.7	-4.3	5.8
Ti	28.3	0.9	-5.4	3.2	-20.1	-26.3	-32.4	-24.3	17.7	-23.3	
Zr	28.5	10.9	19.0	-50.2	-14.4	-16.9	-9.0	-76.0	-11.8	-30.0	
Y	-12.6	-20.1	0.7	-15.3	3.0	-7.2	13.0	-2.6	3.0	11.4	

Table 2. Estimated percent changes of REE and Ti, Zr, and Y contents at the Cliefden outcrop, New South Wales, Australia, calculated passing from spilitic gray domains (samples 02 and 35 in Group 1; sample 83 in Group 2) to Ca-enriched grey-green (33, 87), Yellow-green (31), green (12, 30, 36), and amygdaloidal (87) domains. Data were taken from Smith & Smith (1976) and Hellman et al. (1979). Complete sample numbers are obtained by adding the prefix 350- to the number shown in table, i.e., 02 is sample 35002 (cf. Hellman et al. 1979). Normalized volume change ($\bar{K}_v$) is the geometric mean of volume changes required for each individual REE (excluding La) to have been perfectly immobile. Underlined REE changes differ from zero for this $\bar{K}_v$ by more than two standard deviations (SD) of the analytical method.

	N21		N24		N46		N61	
	C→R	C→S	C→R	C→S	C→R	C→S	C→R	C→S
$\bar{K}_v$	1.18	1.09	1.11	0.86	0.95	0.77	0.97	0.95
La	4.0	45.5	1.8	28.3	-18.9	-0.4	11.0	-0.8
Ce	7.7	20.4	-4.5	-2.0	-6.5	1.7	9.6	-3.0
Nd	-19.5	-17.7	-7.2	-2.9	7.5	19.5	-10.0	-4.2
Sm	0.2	14.3	2.3	9.3	-6.7	3.3	3.7	6.5
Eu	9.5	-1.8	12.6	6.8	0.4	-4.4	4.6	-0.7
Tb	13.7	9.9	-0.4	-1.9	-5.1	-4.4	-0.6	-2.0
Dy	-17.9	-11.7	-2.5	-13.9	23.8	-4.4	-4.5	-8.5
Yb	10.7	2.6	0.3	1.4	-1.8	-5.3	7.5	4.7
Lu	8.9	-4.2	1.8	7.2	-5.1	-1.7	-6.8	9.8
Ti	-2.7	14.9	10.8	20.6	2.2	12.7	-2.9	10.4
Sc	-5.2	17.0	6.0	23.7	-6.1	17.3	0.5	7.9
Hf	4.6	16.9	-2.0	2.7	-5.1	4.0	0.6	8.2

Table 3. Estimated present changes of REE, Ti, Sc, and Hf passing from core (C) to inner rim (R) and selvedge (S) of each of four metabasalt pillows from the Bymark greenstones, Trøndelag, Norway (data from Loeschke 1976, Loeschke & Schock 1980).

that the Na-rich and Ca-rich domains represent complementary diverging trends away from an original, unpreserved basalt. Thus, the trajectory in composition space described by the calculations joins two metasomatic compositions. The primary rock composition probably lies somewhere between these extremes and near this trajectory, but need not lie precisely along it because some elements probably moved on a scale larger than the outcrop (e.g., potassium). However, immobile elements will not change in amount from one rock to another. Therefore, the present purpose is adequately served by using any composition as the initial one in the computations.

The percent standard deviations for REE analyses of BCR-1 reported with the Cliefden outcrop data by Hellman et al. (1979) were used as an estimate of analytical precision of the REE analyses for the Cliefden outcrop. If all REE were assumed to be immobile, required changes in La consistently exceeded analytical uncertainty. Therefore, La was deleted from the estimate of the normalized relative volume change, $\bar{K}_v$ (i.e., La was assumed to have been mobile). As Table 2 shows, a nearly perfect fit of the remaining REE data is thus achieved. Of the computed values of Δx_i for the REE, only La consistently must change in amount from one domain to another by more than two standard deviations of the analytical error. In two sample pairs (samples 12 and 33 compared with sample 35), Ce increases by slightly more than two standard deviations (Fig. 2), suggesting possible limited mobility of Ce as well. Other than one Eu and one Yb value, all other REE can be interpreted as isochemical in these rocks. The chemical data are consistent with the interpretation that, of the REE, only La was mobile enough during alteration for the changes to be statistically resolvable.

The relative volume changes required for individual pairs of Cliefden rocks range from −7% to +36%, with a mean of +18.2%. These are quite reasonable in view of field and petrographic relationships of the Cliefden outcrop (Smith 1968), which suggest that major elements, especially Ca, were leached from the spilitic gray domains and deposited in the Ca-rich domains. Such a process could reasonably be accompained by a small volume loss in the gray domains and volume increase in the Ca-enriched domains. The $\bar{K}_v$ calculated here represents the combination of these two effects.

In fact, REE concentrations vary more from one gray-domain sample to another than they do from gray domains to Ca-enriched domains. This is reflected in the fact that the calculated differences vary systematically with the reference gray domain chosen for a calculation (02 or 35), but not between Ca-rich domains (Fig. 2). REE differences thus do not mimic changes in mineralogy and major-element geochemistry that occurred during alteration. REE relationships are thus difficult to relate to the hydrothermal processes that controlled major-element behavior.

Estimated values of Δx_i for Ti,Y, and Zr, elements that are commonly considered to be relatively immobile and used for basalt classification (Pearce & Cann 1973), range up to about 30% and are not consistently similar between the three elements (particularly calculated changes from 02 and 35 to 12, and 83 to 86). The large changes in Zr associated with sample 30 are suspicious and may indicate a problem with that analysis, which shows only 29 ppm Zr (compare Table 1). However, for several other samples, there is no choice of $\bar{K}_v$ that would allow all three of these elements to have been immobile unless uncertainties of the analyses are greater than 10% (Smith & Smith, 1976, did not report analytical uncertainties). Such a large error is probable for Y, but is unlikely for Ti and Zr analyses by X-ray fluorescence.

A possible interpretation is that one of these elements (e.g., Ti) was immobile, while all of the others, including REE, were mobile. In fact, this is *always* a possible explanation of the variations in a set of analyses, unless the absolute volume change can be determined independently in some way. The present objective is to evaluate published data that have been used as evidence for mobility of groups of elements that have been widely assumed to be immobile and therefore used to classify altered basalts. Smith & Smith (1976) interpreted the limited scatter of the Cliefden samples on a Ti-Y-Zr plot (Pearce & Cann 1973) to indicate that these elements had been largely immobile during alteration. The analysis presented here allows the degree of mobility to be evaluated quantitatively. These elements may sometimes be more mobile during alteration than has been hoped, although the magnitude of the changes shown here in most cases probably would not be large enough to cause misclassification of a suite.

Bymark greenstones

Pillow cores, being least altered in appearance, were used as the reference compositions; these again do not represent primary compositions (Loeschke 1976). The values in Table 3 were also computed assuming that REE except La were immobile. Loeschke & Schock (1980) unfortunately did not give numerical values for analytical uncertainties, but their graphical representation of errors (Loeschke & Schock 1980, Fig. 2) indicates similar precision to that reported by Hellman et al. (1979), except for Nd and Dy for which the uncertainties are quite large (> 30%). Consistent with the latter is that the absolute values of apparent Nd and Dy changes are larger than other elements, but not consistently positive or negative going from core to rim to selvedge. Given this, the Bymark greenstone REE data resemble those from the Cliefden outcrop, with only La consistently reflecting significant mobility. Unlike the Cliefden outcrop, other small, highly charged cations appear to have been relatively immobile (Ti, Sc, and Hf in Table 3).

Comparison of pillow cores to selvedges shows larger compositional differences than between cores and inner rims. Because the pillow selvedge originates as glassy material in direct contact with seawater, the selvedge is probably the most reactive material in the system and is located in the most favorable site for alteration. If REE were mobile in the pillow selvedges (see also Loeschke & Schock 1980 and Walsh 1983), these data indicate that mobility is largely limited to selvedges and fails to affect the remainder of the pillow strongly. This means that routine petrochemical studies are unlikely to be affected, because in collecting for bulk-chemical analysis one would ordinarily avoid pillow selvedges.

Conclusions

Previous studies suggesting REE mobility during alteration have suggested that light REE are most likely to have been mobile, particularly La and Ce (Thompson 1973, Frey et al. 1974, Wood et al. 1976). Mass-balance calculations for the Cliefdon outcrop and Bymark greenstone pillows indicate that La probably was mobile during hydrothermal metamorphism, but movement of Ce is not required in most cases. Hellman et al. (1979) suggested that the persistent negative Ce anomaly in the Cliefden outcrop (Fig. 1) might reflect low-temperature leaching of Ce, citing a report of Ce and Mn depletion in dredge samples from the Shatsky Rise that was attributed to low-temperature alteration (Masuda & Nagasawa 1975). However, the Ce anomaly at Cliefden is smaller than those in the Shatsky Rise samples, and may in some cases be exaggerated by La enrichment. Some fresh basalts with REE patterns broadly similar to the Cliefden rocks have a smal negative Ce anomaly (e.g., Hellman & Henderson 1977, Fig. 2b). A negative Ce anomaly could be a primary magmatic feature of some basalts and perhaps need not result from low-temperature redistribution of Ce. While a petrogenetic cause of a primary Ce anomaly is not obvious, it could perhaps relate to the fact that Ce is one of the REE that exists in more than one oxidation state, and could behave somewhat independently of the other REE under oxidizing conditions.

Baker & de Groot (1983) used the Gresens mass-balance approach to show substantial REE mobility in shear zones in felsic metavolcanic rocks. While their assumption of isovolumetric metamorphism in ductile shear zones seems unwarranted to this author, even if substantial volume changes occurred their data seem to require important REE mobility. A critical difference may lie in the mineralogical differences between mafic and felsic rocks. In particular, the felsic rocks studied by Baker & de Groot (1983) lack epidote minerals and contain rutile rather than sphene as the TiO_2-bearing phase. Thus, chlorite, which apparently fractionated the REE, may have been the only acceptable host for REE. This seems less likely to occur in rocks with a mafic bulk composition.

Variations of REE concentrations in the altered mafic rocks considered here thus can be explained if only La is mobile enough for changes to exceed analytical resolution. Other REE concentration changes can be attributed to indirect effects of net addition or loss of major elements. This means that shapes of REE patterns, excluding La, probably reflect primary igneous patterns and may be a useful guide to interpretation of altered basalts. If total-rock mass-balance effects can be constrained, primary REE patterns might be precisely constructed. On the other hand, elements such as Ti, Y, and Zr have not in all cases changed concentration coherently, and may not be as reliable for classifying basalt types. For all elements, concentration changes due to net loss or gain of major elements may be significant. If these effects cannot be evaluated quantitatively,

only discriminants using ratios of elements should be used, even for genuinely immobile elements. In any case, future studies that address the problem of the mobility of REE should be made with the intention of carrying out this type of study, including the determination of densities of the samples to be analyzed.

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