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Lead isotope and trace element variation in Tenerife pumices: evidence for recycling within an ocean island volcano

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Abstract

Three voluminous Quaternary phonolitic pumice fall deposits erupted from the compositionally-zoned Tenerife magma chamber exhibit variability in Sr and Pb isotope ratios. It has been previously argued that the Sr isotope variations are due to syn-eruptive interaction between magma and hydrothermal fluids (Palacz and Wolff, 1989). Pb compositions are not correlated with Sr, and are believed to reflect magmatic values. Pb isotope ratios exhibit regular variation with degree of fractionation, and one zoned deposit is heterogeneous in Pb. The highest values seem to characterize the most fractionated upper parts of the zoned system. This is unlikely to be a consequence of magmatic recharge. Isotopic and trace element behaviour is instead consistent with combined assimilation and fractional crystallization, involving the recycling of material containing relatively radiogenic Pb, from within the volcanic edifice. Assimilation of sediment intercalated within the submarine portion of the pile is ruled out by the isotopic data. The most probable contaminant is a felsic igneous rock. Early trachytes reported by Sun (1980) have the required Pb isotope compositions and may approximately represent the assimilant.

KEYWORDS: lead isotopes, trace elements, pumice, Tenerife, Canary Islands, recycling.

Introduction

A diverse array of mantle and crustal components may be present within the magmas of a single volcanic province, centre, or eruption, and progress towards the identification of these components on the basis of their isotopic and trace element signatures is one of the success stories of modern igneous geochemistry. The paths by which individual components become incorporated or blended into a final batch of magma are usually difficult to define. Particularly intractable is the extent of recycling via assimilation of material within an individual volcanic edifice into successive bodies of magma that pass through the edifice on their way to the surface, since assimilation and/or combined assimilation and fractional crystallization (AFC) within a volcano's own products does not leave a diagnostic isotopic imprint that can be distinguished from first-cycle mixtures of contributing components. For example, consider a lava from a continental intraplate volcano with isotopic and trace element abundances that can be modelled as a binary mixture of mantle and continental crust components. Does this lava represent a 'fresh' blend of mantle-derived liquid with crustal melt, or interaction of mantle-derived melt with older, crustally-contaminated lavas or intrusions within the body of the volcano?

For comagmatic samples not related by different degrees or styles of partial melting, non-zero intercepts of trends on incompatible trace element

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plots indicate the operation of some process(es) other than, or in addition to, crystal fractionation. Potentially, a well-correlated trend of this type is a powerful diagnostic of AFC within a volcano (Powell, 1984), although contaminants cannot be closely characterized without some estimate of the ratio of mass assimilation rate to mass fractionation rate (*r*; DePaolo, 1981).

This paper examines trace element and Pb isotopic variations in phonolitic pumices from three plinian deposits on Tenerife, Canary Islands. Although the Tenerife centre is known to be periodically recharged with mafic magma (Wolff, 1985*a*), a significant role for assimilation has not been recognized. Wolff and Storey (1984) ascribed trace-element variations *within* individual deposits to fractional crystallization. Here we show that the three deposits are comagmatic, that AFC is a more plausible explanation of trace-element trends than closed-system fractionation, and that some role for assimilation is required by Pb isotope variations.

Summary of previous work

General. Tenerife is a large, mature alkalic oceanic island with an extensive phonolitic pyroclastic series erupted from the centrally located, late Quaternary Las Cañadas caldera. Earlier publications deal with sampling techniques, mineralogy, petrology, geochemistry, and Sr isotope relations (Wolff, 1983, 1984, 1985*a*,*b*, 1987; Wolff and Storey, 1983, 1984; Palacz and Wolff, 1989); aspects of specific relevance to this study are reviewed in this section.

Tenerife consists of a late Tertiary, dominantly mafic shield, capped by younger differentiated volcanics and later basalts. Within this complex is developed the Las Cañadas caldera, a multiple structure of two or three overlapping collapse features modified by late northward landslip to give a horseshoe-shaped depression (Borley, 1974; Wolff, 1983; B. Booth and G. P. L. Walker, unpublished data); within the caldera stand the twin stratovolcanoes of Teide and Viejo. Lavas of the early shield-building phase are Miocene to Pliocene in age and are exposed in the extreme northwest and northeast of the island, as paleotopographic highs on the flanks (see geologic map given by Wolff, 1987), and at the base of the Las Cañadas caldera scarp. They are mostly mafic with subordinate flows and minor intrusions of trachyte and phonolite (Abdel-Monem *et al.*, 1972; Sun, 1980).

A late Quaternary pumice series, dominantly of phonolitic composition, is well exposed in the south and east of the island. Booth and Walker (unpublished) distinguish the products of some forty individual eruptions from vents located within Las Cañadas. The deposits are mostly nonwelded plinian pumice falls and ignimbrites, interbedded with closely related reworked and epiclastic material. At least twelve primary deposits have volumes greater than 0.5 km^3. The total volume is approximately 70 km^3 of magma. Booth and Walker divided the succession into a lower Granadilla Series and an upper Fasnia Series. Based on soil thicknesses, the Granadilla Series is considered to have accumulated fairly rapidly, perhaps over as little as 10 000 years. Although some individual members of the Granadilla Series are compositionally zoned, evidence for recharge of the system by basaltic magma is confined to the topmost unit, the Tajao ignimbrite (Wolff, 1985*a*), which post-dates the deposits of interest here.

The most voluminous members of the Granadilla Series are three units designated (in ascending stratigraphic order) D, E and G by Booth and Walker. G is the Granadilla Pumice (Booth, 1973), the most prominent member of the Series.

Trace element behaviour. Trace element variations (and major element variations where significant) within the Granadilla Series are broadly consistent with those predicted to result from fractionation of observed phenocryst phases. *MREE*, Ba, and Sr behave as compatible elements, due to fractionation of titanite and alkali feldspar. U, Th, Zr, and Rb are all strongly incompatible. Although bulk distribution coefficients cannot be closely constrained, the relative degrees of enrichment or depletion of trace elements within the suite can be modelled by simple Rayleigh fractionation using published coefficients (Wolff and Storey, 1984).

Sr-isotope behaviour. The Tenerife suite, including mafic rocks, exhibits little variation in $^{87}Sr/^{86}Sr$, with values clustering around 0.7032 ± 0.0001; $^{143}Nd/^{144}Nd$ is also virtually constant (Z. A. Palacz, unpublished data). Palacz and Wolff (1989) found that whole-pumice powders from D, E and G are invariably more radiogenic in Sr ($^{87}Sr/^{86}Sr = 0.70356–0.70668$), and that Sr isotope variation was coupled to major and trace-element abundances, but decoupled from Nd and Pb isotope ratios. They also found that cleaned feldspar separates from G yielded an effectively constant $^{87}Sr/^{86}Sr$ of 0.70316 ± 0.000025, and ascribed the anomalous behaviour of Sr to late-stage—probably syn-eruptive—contamination of the Sr-poor magma by hydrothermal fluids.

Table 1. Major and trace element data for pumice fall D. Samples DF1 - DF4 are in ascending stratigraphic order through the deposit. Major elements normalized to 100% volatile-free.

Sample	DF1	DF2	DF3	DF4
SiO_2	62.43	62.53	61.92	62.66
TiO_2	0.54	0.53	0.52	0.53
Al_2O_3	18.68	18.83	19.02	18.66
Fe_2O_3t	3.35	3.34	3.50	3.36
MnO	0.24	0.24	0.25	0.22
MgO	0.38	0.43	0.55	0.68
CaO	0.54	0.54	0.63	0.55
Na_2O	7.32	7.08	7.03	6.96
K_2O	6.47	6.43	6.54	6.34
P_2O_5	0.06	0.05	0.04	0.05
Rb	170	169	166	166
Sr	4	6	4	6
Zn	131	131	129	128
Y	52	54	52	57
La	137	135	129	126
Ce	254	277	278	282
Nd	62	62	61	62
Sm	8.5	8.8	8.5	9.2
Eu	1.4	1.4	1.5	1.6
Tb	1.2	1.3	1.4	1.1
Yb	4.5	4.4	4.2	4.2
Zr	1130	1130	1097	1130
Th	27.3	26.1	30.3	31.4
U	6.9	7.7	6.5	7.0
Nb	340	342	327	344
Ta	21.2	22.6	22.3	23.5

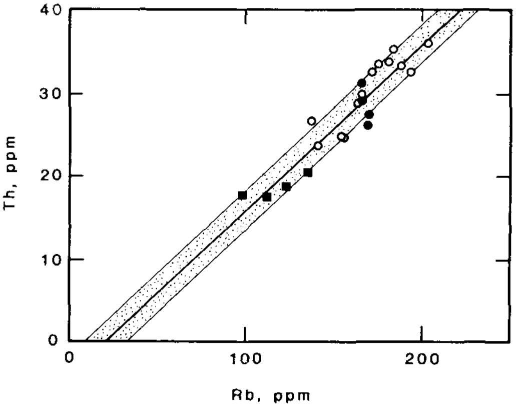

FIG. 1. Th concentration *vs.* Rb concentration in pumice from deposits D (filled circles), E (filled squares) and G (open circles). Heavy line is least-squares linear regression through data; stippled envelope around regression line is analytical error, from Wolff (1983).

Deposits D, E, and G: field aspects

Erupted volumes are estimated at 14 and 22 km^3 respectively for D and E (calculated by Wolff, 1983, from data of Booth and Walker), and 45 km^3 for G (Walker, 1981). Assuming a minimum pumice density of 0.5 g cm^{-3}, these convert to minimum values of 3.6, 5.7 and 11.7 km^3 of magma respectively. All three are widespread plinian pumice deposits. E and G are capped by non-welded ignimbrites produced during the closing phases of their eruptions.

Plinian pumice fall D possesses a fine basal ash but is otherwise even-grained and homogeneous with very slight reverse grading. The light grey pumice is almost aphyric, with alkali feldspar as the dominant phenocryst, accompanied by rare biotite, titanite, and opaques. At most exposures D pumices are visibly altered, and the unit was perforce sampled at a 1.7 m thick exposure lying on the 3 m isopach (Wolff, 1983), and thus the upper half of the deposit is missing. The sampled portion is mineralogically, geochemically and isotopically homogeneous (Tables 1 and 2).

E is a very widespread, coarse-grained, poorly-sorted plinian deposit locally overlain by an ignimbrite. It is the most widely dispersed of the three deposits, and must be the result of an unusually violent eruption with a very high discharge rate of magma feeding a high eruption column. Although the extent of exposure precludes a rigorous treatment, analysis of Booth and Walker's lithic clast dispersal data by the method of Carey and Sparks (1986) suggests a column in excess of 40 km high, assuming a vent position somewhere within Las Cañadas. This corresponds to a very high volumetric discharge rate at the vent, in excess of 10^5 m^{3}s^{-1} (Sparks, 1986). The capping ignimbrite contains clasts of mafic phonolitic pumice in addition to the dominant pale grey variety, and banded pumices with both varieties are common. The mafic phonolite is somewhat less evolved than the pale-coloured glass (Wolff, 1983), and the unit therefore possesses an overall chemical zonation. The fall deposit is itself compositionally zoned, especially in trace-element abundances (Wolff and Storey, 1984). The zonation is reversed below 30% thickness. The pale pumice is porphyritic with anorthoclase + diopside + biotite + titanite + Fe–Ti oxides. Major and trace element data for E are given by Wolff and Storey (1984); isotopic data are in Table 2. All samples of E have lower abundances of incompatible elements than D and G (Figs 1 and 2). The appearance of mafic pumice in the last-erupted portion may indicate near-exhaustion of the uppermost, more fractionated magma during the progressive removal of magma from a zoned chamber, or it may be a drawdown effect related to the unusually high discharge rate (Blake and Ivey, 1986).

G consists of four plinian fall units capped by an ignimbrite. Major, trace element, and isotopic data are given by Wolff and Storey (1984), and Palacz and Wolff (1989). G is compositionally

Table 2. Pb isotopic data for pumice deposits D and E

Sample	DF1	DF3	EF1
$^{206}Pb/^{204}Pb$	19.561	19.540	19.358
$^{207}Pb/^{204}Pb$	15.574	15.496	15.60
$^{208}Pb/^{204}Pb$	39.385	39.31	39.24

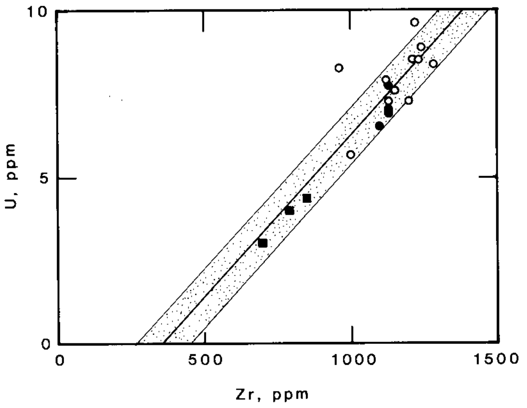

FIG. 2. U concentration *vs.* Zr concentration in pumice from deposits D, E, and G. Notation as Fig. 1.

zoned with a basal reversely zoned portion, and is isotopically heterogeneous in Sr (Palacz and Wolff, 1989) and Pb (this paper). Some samples of G have been visibly contaminated by partly disaggregated fragments of mafic country rock (Wolff and Storey, 1984); those samples are excluded from consideration in this paper.

Evidence for comagmatic nature of D, E, and G

On the basis of field and geochemical data, D, E, and G are considered to be comagmatic in the sense that all are derived from the same high-level zoned magma chamber, share a common parent (ne-normative trachyandesite or mafic phonolite), and followed closely similar crystallization paths.

(i) The deposits are the most voluminous in the Granadilla Series; Booth and Walker consider it unlikely that any others have vesiculated volumes in excess of 1 km^3. D, E, and G are therefore bigger than associated deposits by over an order of magnitude, and provide the most complete sampling of the magmatic system. Very much smaller deposits are locally observed separating D from E and E from G, but cannot be regarded as major events in the life of the system.

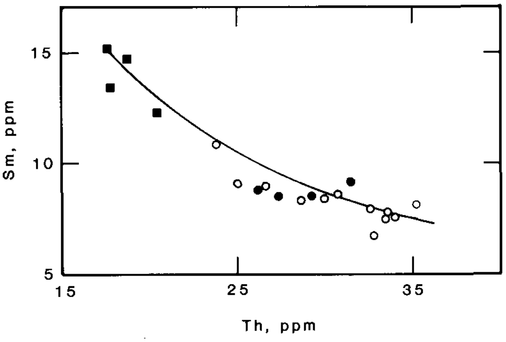

FIG. 3. Sm concentration *vs.* Th concentration in pumice from D, E, and G. Curve is calculated Rayleigh fractionation line using the least differentiated sample as parent and apparent distribution coefficients from Wolff and Storey (1984). Other notation as Fig. 1. Typical errors are ±6% for Th, and ±4% for Sm.

(ii) As mentioned above, the entire Granadilla Series appears to have been rapidly emplaced. Booth and Walker (unpublished) have suggested typical repose periods of *ca.* 1000 years for the Granadilla Series. This estimate is on a similar time scale to that required for significant fractionation to occur within high-level alkaline systems (Wolff and Storey, 1984; Wolff *et al.*, in prep.). At any rate, field evidence suggests a shorter repose time separating D and E than between E and G.

(iii) Magmatic compositions are geochemically coherent. This is true both for incompatible (Figs 1 and 2) and compatible elements (Fig. 3). The abundances of compatible elements are particularly sensitive to differences in crystallizing assemblages and to recharge or mixing with more mafic magma. The Tenerife phonolites contain titanite as a phenocryst phase, which causes progressive depletion in middle rare earths (*MREE*) with fractionation. *MREE* are least depleted in unit E, and become progressively removed with fractionation through G (Fig. 3). The duplication of chemical trends for the whole data set with those defined within an individual zoned deposit (G) is compelling evidence for a close genetic relation between the three deposits.

E is regarded as parental to D and G. D represents the uppermost portion of a zoned magma chamber that was more deeply excavated during the E eruption, with little fractionation occurring in the chamber between the two events. Fractionation of residual phonolite remaining in the

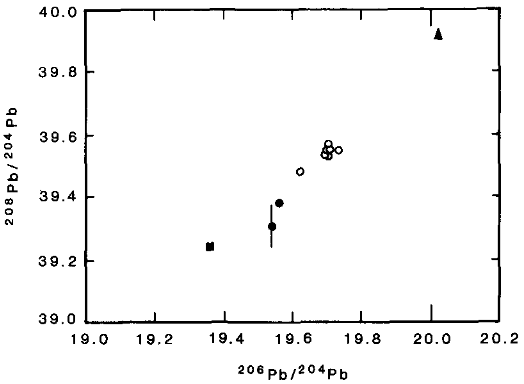

FIG. 4. ^{208}Pb/^{204}Pb *vs.* ^{206}Pb/^{204}Pb in pumice from D, E, and G. Triangle: 5.35 Ma trachyte from Sun (1980); other notation as Fig. 1. The larger error for one sample from deposit D applies to that sample only; for all other points, $2r$ errors are less than twice the diameter of the plotted point.

system following the E eruption resulted in rezonation of the chamber, which then erupted again to yield zoned deposit G. The least fractionated compositions sampled from G are only slightly more evolved than the most fractionated E sample.

Pb isotope behaviour

Pb analytical details are given in Palacz and Wolff (1988). In contrast to Sr and Nd, Pb isotope ratios are highly variable in the whole Tenerife suite (Sun, 1980; Z. A. Palacz, unpublished data); values for D, E, and G lie within the range for Tenerife basanites. There is no correlation between Sr and Pb isotope ratios, indicating that measured ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, and ^{208}Pb/^{204}Pb are magmatic values unaffected by the fluid contamination process which dominates observed Sr-isotope ratios. ^{206}Pb/^{204}Pb and ^{208}Pb/^{204}Pb vary appreciably between D, E, and G, and to a lesser extent within G (Fig. 4); ^{207}Pb/^{204}Pb exhibits little analytically significant variation. E contains the least radiogenic Pb, and G the most radiogenic. The least fractionated sample from G also has the least radiogenic Pb in that deposit (Fig. 5). Pb isotope variation demonstrates that the high-level, zoned Tenerife magma chamber was acting as a open system during production of the Granadilla Series. This could either result from variations in the Pb isotopic composition of 'fresh' magmas replenishing the system at depth, or from assimilation of isotopically distinct material within the volcanic pile.

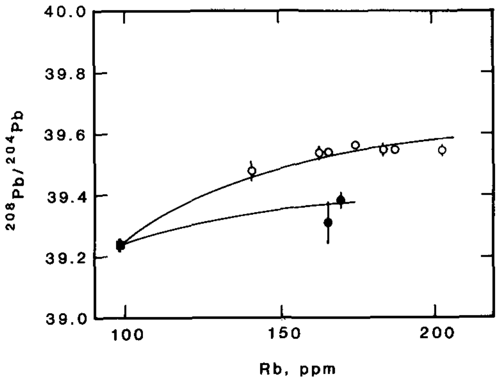

FIG. 5. ^{208}Pb/^{204}Pb *vs.* Rb concentration in pumice from D, E, and G. Curves: calculated AFC curves using E (sample EF1) as initial magma, and a hypothetical contaminant having the same ^{208}Pb/^{204}Pb as Sun's (1980) 5.35 Ma trachyte. Other parameters: Pb in assimilant = 40 ppm; Pb in initial magma = 20 ppm; Rb in assimilant = 60 ppm; $D_{Pb} = 0.2$; $D_{Rb} = 0$.

Trace element evidence for AFC processes

Although trace element variations can be modelled by closed-system fractionation to a first approximation, closer examination suggests that this is unsatisfactory. A plot of two trace elements obeying the Rayleigh fractionation law and having the same bulk solid/liquid distribution coefficient should yield a straight line that passes through the origin. Incompatible–incompatible element plots for D, E, and G yield straight lines within analytical error, but their intercepts are markedly non-zero (Figs 1 and 2). This applies to nearly all element pair combinations among U, Th, Rb, Zr, and Hf, which exhibit the most incompatible behaviour in the suite.

Powell (1984) inverted the AFC equations of DePaolo (1981) in order to explore how assimilant compositions could be constrained from geochemical data on sample suites where AFC is suspected, and found that linear trends with non-zero intercepts between two incompatible elements are a strong indicator of AFC (perhaps the only indicator where there is no isotopic contrast between magma and assimilant). Although other interpretations of the present data are possible (e.g. slight curvature of trends that cannot be resolved), it seems that, within the limitations of the data set, AFC processes are a more plausible explanation of the trace element variations than closed system fractionation. For negative intercepts, the assimilant composition lies above the best-fit line through the data (Powell, 1984). Therefore, the assimilant in this case has higher U/Zr and Th/Rb than the magma (Figs 1 and 2). For the five incompatible elements listed above, relative concentrations in the assimilant compared to the magma are U $\geqslant$ Th $\geqslant$ Hf $>$ Zr $>$ Rb.

Discussion

Given the geochemical coherence of the three deposits, the increasingly radiogenic character of Pb with fractionation, and the trace element relations, assimilation of country rock into phonolitic magma is a more satisfactory mechanism of inducing the isotopic variation than magmatic recharge of the system, for which there is in any case no evidence prior to post-G times. Replenishment with isotopically variable parental magma is unlikely to result in coherent relationships. For example, trace element and isotopic variations in units from the younger Fasnia Series, in which several recharge events are recorded, are markedly discontinuous (Wolff, 1983 and unpublished data).

The nature of the contaminant can be constrained to some extent by geological, geochemical and thermal considerations. Assimilation into phonolitic magma, which forms the capping layer to the zoned Tenerife system (Wolff and Storey, 1984; Wolff, 1985*a*), requires that the contaminant be present within the volcanic pile itself. The top of the magma system may be as shallow as 4 km (Wolff, 1987), which is approximately at sea level for present day Tenerife. Within the island structure, available igneous rocks will be the full range of mafic through felsic types, variably altered by seawater and meteoric water, with intercalated marine sediments. Atlantic Ocean sediments reported by Sun (1980) have less radiogenic Pb isotope compositions than our samples, and cannot be regarded as plausible contaminants (the same conclusion was reached by Sun, op. cit., for Canary Islands volcanics in general). Furthermore, magmatic Sr, Nd, and Pb isotope ratios in the pumices fall within the range for Tenerife mafic magmas even where that range is very narrow (e.g. for Sr). The assimilant is therefore very unlikely to be a sediment or an igneous rock whose isotopic composition has been significantly modified by alteration.

The contaminant is almost equally unlikely to be a mafic composition. The ability of felsic magma to assimilate basaltic compositions is limited, especially at the rather high rates indicated by attempts at modelling the isotope–trace-element trends (Fig. 5). Also, assimilation of significant quantities of basic material should leave a detectable imprint on the major element and compatible trace element chemistry of the magma, which we have already shown not to be the case. Therefore, the most probable contaminant is felsic igneous rock. Clearly this material must have significantly more radiogenic Pb compositions than the analysed magmas.

A possible candidate for material similar to our required contaminant is provided by Sun (1980), who reports a trachyte from the early shield-building phase (5.35 Ma) with $^{206}Pb/^{204}Pb = 20.022$ and $^{208}Pb/^{204}Pb = 39.918$, considerably more radiogenic than Granadilla Series phonolites. Although our data require a contaminant with slightly lower $^{208}Pb/^{206}Pb$ (Fig. 4), Sun's sample clearly demonstrates that material with Pb sufficiently radiogenic to provide the required leverage as a contaminant for our magmas existed within the Tenerife volcano during the lifetime of the Granadilla Series magma chamber.

Attempts at quantitative modelling. Without precise information on contaminant compositions and bulk solid/liquid distribution coefficients for elements and isotopic systems of interest, virtually any correlated geochemical trend can be modelled using the AFC equations of DePaolo (1981) by judicious manipulation of input parameters. This is especially problematic for felsic magmas, where distribution coefficients may be highly variable within a single system (Wörner *et al.*, 1983). Some useful indications can however be gained by computing AFC curves for Granadilla Series Pb-isotope–trace-element covariations, assuming that (i) the assimilant is a felsic igneous rock, with trace element abundances similar (e.g. within a factor of 3) to those of analysed samples, and Pb isotope composition similar to that of Sun's (1980) early trachyte, and (ii) that distribution coefficients for Pb and the incompatible elements discussed above are similar and low. Good fits to the E–G trend are favoured by high values of r around 0.5, and low concentrations of Rb in the assimilant relative to magma, the latter consistent with the indications from trace-element–trace-element plots. An example curve is shown in Fig. 5. The requirement for high r values remains over a wide variation in other input parameters, although it appears from Fig. 5 that AFC may not have been significant in the generation of the most strongly fractionated compositions in G (Rb $>$ 180 ppm). For any one combination of initial element concentrations and bulk distribution coefficients, the E–D isotopic shift can be accommodated using an r value approximately half that required for the E–G trend.

The high r values may not be totally unreasonable. Given a low initial temperature contrast, any magma should be capable of absorbing a solid of similar composition and mineral assemblage through AFC up to a value of r close to 1 ($r = 1$

corresponds to ideal zone refining), if the magma and assimilant are in intimate contact. In a sub-caldera magma chamber, this intimate contact will be favoured due to the disrupted nature of the roof, a consequence of repeated caldera collapse. This is particularly true of Tenerife, where the caldera volume is approximately equal to the *total* (magma equivalent) volume of the Granadilla and Fasnia Series ($\sim 70\,\mathrm{km}^3$), but is much greater than any individual deposit, suggesting that collapse occurred incrementally in response to each major eruption. High rates of assimilation in the Tenerife chamber may be due to large pieces of the volcanic edifice foundering into the magma chamber during major collapse episodes.

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