

URANIUM-THORIUM ACTIVITIES AND DISEQUILIBRIUM IN VOLCANIC ROCKS FROM THE ANDES (33-46°S): PETROGENETIC CONSTRAINTS AND ENVIRONMENTAL CONSEQUENCES

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ABSTRACT

This is a two part study of recent lavas from the Southern Volcanic Zone (SVZ) of the Andes (33-46°S). In the first part, radioactive equilibrium between ^{238}U and ^{230}Th was tested by analyzing 9 samples from historic eruptions for $^{230}\text{Th}/^{232}\text{Th}$ activity ratio and for U and Th abundances and activities. In the second part, ^{238}U and ^{232}Th activities were determined in 23 volcanic rocks ranging in composition from basalt to rhyolite.

Based on the 9 samples tested for radioactive equilibrium between ^{238}U and ^{230}Th , lavas from the 33-37°S segment of the SVZ, where the continental crust is thick (55-35 km), show ^{238}U - ^{230}Th equilibrium. Lavas from south of 37°S, where the continental crust is thin (30-35 km), show either ^{238}U - ^{230}Th equilibrium or disequilibrium and ^{238}U enrichment. $^{238}\text{U}/^{230}\text{Th}$ (a direct measure of radioactive disequilibrium determined by the ratio of $^{238}\text{U}/^{232}\text{Th}$ and $^{230}\text{Th}/^{232}\text{Th}$) correlates with the common indicators of subduction zone processes ^{10}Be , Rb/Cs and La/Yb, suggesting that the source of the ^{238}U enrichment is subducted oceanic crust and sediments. Based on the correlation of disequilibrium with indicators of subducted slab input, the rate of magma ascent from the subduction zone is 0.4 to 1.33 meters/year. This rate is consistent with calculated rates for magma ascent as diapirs or through fractures, but is too rapid for a significant portion of the ascent path to consist of either porous media flow or crustal storage. The magmatic source of Andean lavas from the volcanic front appears to be experiencing U enrichment through time by metasomatism.

^{238}U specific activities vary from 3 to 42 Bq/kg and ^{232}Th specific activities vary from 3 to 69 Bq/kg in the 23 samples studied. The highest ^{238}U and ^{232}Th concentrations were found in rhyolites, and in general U is more compatible than Th during magmatic differentiation from basalt to rhyolite. These values are within the concentration range of soils in areas of normal natural activity. There may be a major health risk to populations living near volcanoes that erupt silicic compositions resulting from inhalation of ash or soil particles and suffering exposure to radiation from decay of U and Th.

Key words: U-Th systematics, Radioactive equilibrium, Quaternary volcanic rocks, Southern Andes, Chile.

RESUMEN

El presente es un estudio, en dos partes, de lavas recientes de la Zona Volcánica Sur (ZVS) de los Andes (33-46°S). Con el fin de determinar la razón de actividad $^{230}\text{Th}/^{232}\text{Th}$ y la abundancia y actividad del U y del Th, en la primera parte se estudió, en nueve muestras de erupciones históricas, el equilibrio radiactivo entre el ^{238}U y el ^{230}Th . En la segunda parte se determinaron, en 23 rocas volcánicas cuya composición varía entre basáltica y riolítica, las actividades del ^{238}U y del ^{232}Th .

El análisis radiactivo de las nueve primeras muestras permitió observar que las lavas provenientes del segmento 33-37°S de las ZVS, donde la corteza continental es más gruesa (55-35 km), exhiben un equilibrio entre el ^{238}U y el ^{230}Th ; en cambio, las lavas provenientes de la zona al sur de los 37°S, donde la corteza es más delgada (30-35 km), muestran ya sea un equilibrio entre ambos isótopos o un disequilibrio acompañado por un enriquecimiento en ^{238}U . La razón $^{238}\text{U}/^{230}\text{Th}$, que es una medida directa de disequilibrio radiactivo determinada por la relación $^{238}\text{U}/^{232}\text{Th}$ y $^{230}\text{Th}/^{232}\text{Th}$, se correlaciona

con indicadores comunes de procesos de subducción, tales como: ^{10}Be , Rb/Cs y La/Yb , sugiriendo que los causantes del enriquecimiento en ^{238}U son la corteza y los sedimentos oceánicos subducidos. Sobre la base de esta correlación, la velocidad de ascenso magmático, desde la fuente, es de 0,4-1,33 m/año. Esta velocidad es comparable con las calculadas para el ascenso de magmas en forma de diapiros y a través de fracturas, pero es demasiado elevada en relación con las estimadas para ascensos a través de material poroso y para ascensos que incluyen un almacenamiento prolongado en la corteza continental. La fuente magmática de las lavas del frente volcánico de los Andes del Sur parece haber experimentado, con el tiempo, un enriquecimiento en ^{238}U por metasomatismo.

Las actividades específicas del ^{238}U varían entre 3 y 42 Bq/kg y las del ^{232}Th fluctúan entre 3 y 69 Bq/kg. Estos valores están dentro del intervalo de actividad presentado por suelos de áreas de actividad natural normal. Las mayores concentraciones de ^{238}U y ^{232}Th se encuentran en las riolitas. En general, el U es más compatible que el Th durante la diferenciación magmática de basalto a riolita. Los volcanes que emiten material rico en SiO_2 presentan un mayor riesgo para la salud de la población que vive en los alrededores que los de naturaleza basáltica, no sólo por la inhalación de ceniza o partículas de polvo, sino que, también, por la exposición a la radiación causada por el decaimiento radiactivo del U y del Th.

Palabras claves: Sistemática U-Th, Equilibrios radiactivos, Rocas volcánicas cuaternarias, Andes del Sur, Chile.

INTRODUCTION

^{238}U decays by emitting a total of 8 alpha and 6 beta particles to reach the stable isotope ^{206}Pb , with a total half-life of 4.47×10^9 years. This decay series of 15 nuclides includes ^{230}Th , which has a half life of 7.52×10^4 years. At radioactive equilibrium, the activity of each isotope in the decay chain has the same activity. If some process fractionates the elements, then radioactive disequilibrium will be measured between the elements that were fractionated. The half life determines the rate at which radioactive equilibrium is reestablished, and hence radioactive disequilibrium is a chronometer of fractionation processes.

This paper presents the results of two approaches to studying U and Th isotopes in volcanic rocks. In the first approach, ^{238}U and ^{230}Th are used both as isotopic tracers of distinct sources and as indicators of melting processes in the subduction zone. It considers measured radioactive disequilibrium between ^{230}Th and its parent ^{238}U . The activities of ^{230}Th (daughter) and ^{238}U (parent) are normalized to the activity of ^{232}Th , the parent of the separate Th decay series. Samples in $^{230}\text{Th}/^{232}\text{Th}$ - $^{238}\text{U}/^{232}\text{Th}$ radioactive equilibrium have equal activity ratios and define a line with slope equal to one known as the equiline (Fig. 2; Allegre and Condomines, 1976). At a given $^{230}\text{Th}/^{232}\text{Th}$ ratio, fractionation of U from Th will displace the composition from the equiline, either to the right (in case of U enrichment) or to the left (in case of Th enrichment). With time after fractionation, $^{230}\text{Th}/^{232}\text{Th}$ changes along a vertical trajectory according to the following equation:

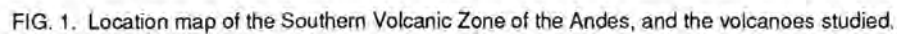
$$\frac{^{230}\text{Th}/^{232}\text{Th}}{^{238}\text{U}/^{232}\text{Th}} = \left(\frac{^{230}\text{Th}/^{232}\text{Th}}{^{238}\text{U}/^{232}\text{Th}} \right)_0 e^{-\lambda t} + \lambda$$

where

$\lambda = ^{230}\text{Th}$ decay constant = $9.22 \times 10^{-6} \text{ years}^{-1}$ (Allegre, 1968; Newman *et al.*, 1986). Therefore, whether disequilibrium is caused by U or Th enrichment, equilibrium is achieved on a timescale given by the half life of ^{230}Th (7.52×10^4 years). Depending on the initial fractionation, radioactive equilibrium is reestablished in the geologically short time of 7.5×10^4 to 2.3×10^5 years.

Quaternary volcanism in the Southern Andes is one of the products of the subduction of the oceanic Nazca Plate beneath the continental South American Plate. Latitude 37°S is an important segment boundary within the Southern Andes; from 33° to 37°S , continental crust ranges in thickness from 55 to 35 kilometers, while south of 37°S continental crust ranges in thickness from 30 to 35 kilometers (Hildreth and Moorbath, 1988). The authors find, in agreement with Sigmarsson *et al.* (1991), that components derived from the subducting slab enrich mantle partial melts in ^{238}U over its daughter product ^{230}Th . This enrichment is only observed in lavas from the thin crust segment south of 37°S . The preservation of ^{238}U - ^{230}Th disequilibrium in lavas south of 37°S implies that the ascent rate of magma through the mantle and crust must be greater than 0.4 to 1.33 meters/year.

In the second portion of the study, ^{238}U and ^{232}Th were determined in lavas from 23 centers of the SVZ (Fig. 2). When U and Th rich ash particles are inhaled during an eruption, they can lodge in lung tissue.



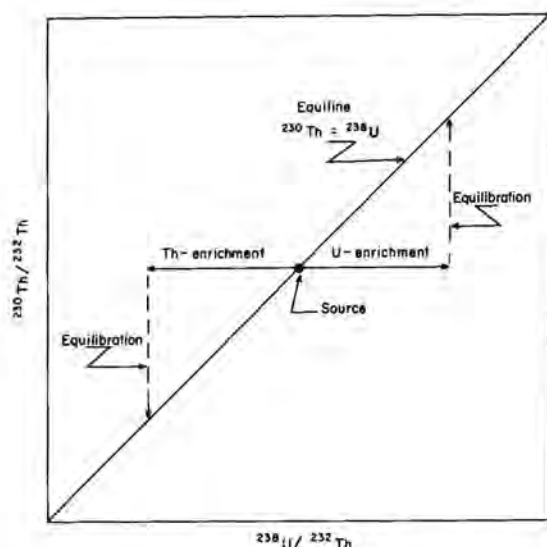


FIG. 2. Schematic representation of ^{238}U and ^{230}Th (normalized to ^{232}Th) fractionation on an equiline diagram. Values are activity ratios. The equiline is defined by radioactive equilibrium between ^{238}U and ^{230}Th . From a source initially in equilibrium, disequilibrium with Th enrichment moves the composition to the left, while in disequilibrium with U enrichment the composition moves horizontally to the right. Samples evolve towards the equiline along vertical trajectories, and depending on the original fractionation return to radioactive equilibrium after 1 to 3 half lives of ^{230}Th .

Episodic degassing of volcanoes can also release the daughter product radon, which can also be inhaled and lodge in lung tissue. In either case, as the elements decay within human tissue the radiation emitted can have adverse health effects. Hence, it is important

to quantify any increased health risk due to living near a volcano and inhaling excess amounts of radioactive elements.

RESULTS

The analytical methods used in this study are described in the Appendix. ^{238}U and ^{232}Th specific activities (expressed in Bq/kg), SiO_2 abundance (expressed in weight per cent), $^{238}\text{U}/^{232}\text{Th}$ and $^{230}\text{Th}/^{232}\text{Th}$ activity ratios, and the chemical classification of each lava sample are shown in Table 1. This table

also includes the name and latitude of the volcanic centers, ^{10}Be , Rb/Cs and La/Yb ratios, and eruption dates of samples analyzed for the first part of this study.

^{238}U and ^{232}Th specific activities increase as the SiO_2 content of the samples increases (Fig. 3). At

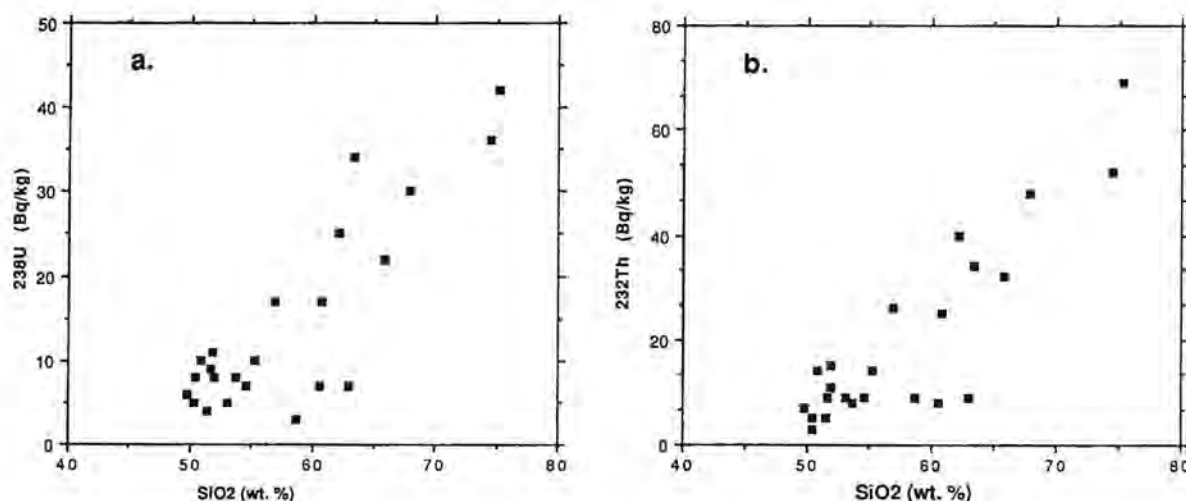


FIG. 3. Correlation of ^{238}U (Fig. 3a) and ^{232}Th (Fig. 3b) with SiO_2 . The correlation of ^{238}U and SiO_2 is $r = 0.85$; $p < 0.001$, and that of ^{232}Th and SiO_2 is $r = 0.88$; $p < 0.001$.

TABLE 1. SiO₂ (wt%), ²³⁸U (Bq/kg) AND ²³²Th (Bq/kg) CONCENTRATIONS, AND ²³⁸U/²³²Th AND ²³⁰Th/²³²Th RATIOS IN QUATERNARY VOLCANIC ROCKS FROM THE SOUTHERN ANDES

Sample N°	Volcano	Latitude	SiO ₂	²³⁸ U	²³² Th	²³⁸ U/ ²³² Th	²³⁰ Th/ ²³² Th	¹⁰ Be	Rb/Cs	La/Yb	Eruption Date
160283-01	Carrán	40°24'S	—	—	—	0.95	0.91	—	—	3.40	1957
111275-09	Huichatio	39°25'S	51.93	—	—	0.84	0.88	—	18.9	7.91	1971
260186-01	Villarrica	39°25'S	52.20	—	—	1.10	0.97	2.5	9.6	3.45	1984
051177-07	Antuco	37°25'S	52.86	—	—	0.81	0.80	1.0	16.0	5.19	1853
PT-9	Peteroa	35°12'S	55.10	—	—	0.80	0.80	—	20.0	7.76	1937
030282-02	Calbuco	41°25'S	55.76	—	—	1.00	0.86	—	13.9	3.16	1961
281282-03	San José	33°47'S	62.30	—	—	0.85	0.88	1.4	28.9	14.9	19th cent.
070282-02	Puyehue	40°30'S	69.00	—	—	0.91	0.82	—	13.7	5.29	1921
070283-02	Descabezado Gr.	35°30'S	71.98	—	—	0.83	0.85	—	19.1	9.15	—
070284-05	Cay	45°04'S	49.71	6 ± 1	7	0.86	—	—	—	—	—
070284-04	Macá	45°06'S	50.36	5 ± 1	5	1.00	—	—	—	—	—
151282-09	Osorno	41°10'S	50.42	8 ± 1	3	2.67	—	—	—	—	—
010284-03	Hudson	45°55'S	50.82	10 ± 1	14	0.71	—	—	—	—	—
120284-07	Hualaihué	41°53'S	51.42	4 ± 1	5	0.80	—	—	—	—	—
120284-01	Corcovado	43°11'S	51.68	<9	9	<1.00	—	—	—	—	—
120284-05	Michinmahuida	42°48'S	51.88	11 ± 1	15	0.73	—	—	—	—	—
010284-01	Hudson	45°55'S	51.90	8 ± 1	11	0.73	—	—	—	—	—
120284-10	Hornopirén	41°53'S	53.05	5 ± 1	9	0.56	—	—	—	—	—
151282-10	Osorno	41°10'S	53.69	8 ± 1	8	1.00	—	—	—	—	—
100284-09	Yanteles	43°28'S	54.57	7 ± 1	9	0.78	—	—	—	—	—
070284-03	Macá	45°06'S	55.32	10 ± 1	14	0.71	—	—	—	—	—
100284-05	Melimoyu	44°04'S	56.84	17 ± 2	26	0.65	—	—	—	—	—
120284-07	Huequi	42°23'S	58.66	3 ± 1	9	0.33	—	—	—	—	—
100284-03	Mentolat	44°42'S	60.55	7 ± 1	8	0.88	—	—	—	—	—
120284-11	Yate	41°48'S	60.81	17 ± 2	25	0.68	—	—	—	—	—
090283-01	Descabezado Gr	35°30'S	62.11	25 ± 3	40	0.63	—	—	—	—	—
100284-04	Mentolat	44°42'S	63.02	7 ± 1	9	0.78	—	—	—	—	—
281282-06	San José	33°47'S	63.35	34 ± 2	34	1.00	—	—	—	—	—
050283-01	Descabezado Gr.	35°30'S	65.85	22 ± 2	32	0.69	—	—	—	—	—
060283-04	Descabezado Gr.	35°30'S	67.90	30 ± 3	48	0.63	—	—	—	—	—
120284-04	Chaitén	42°50'S	74.44	36 ± 4	52	0.69	—	—	—	—	—
280183-05	Lag. del Maule	36°00'S	75.25	42 ± 4	69	0.61	—	—	—	—	—

Note: Analytical methods and uncertainties are discussed in the Appendix.

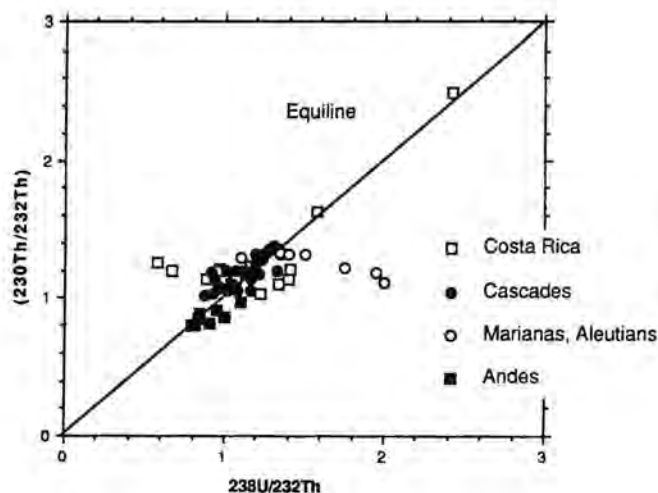


FIG. 4. ^{238}U - ^{230}Th data for arcs (Costa Rica from Allegre and Condomines, 1976; Cascades from Newman *et al.*, 1986; Marianas and Aleutians from Newman *et al.*, 1984; Andes from this study).

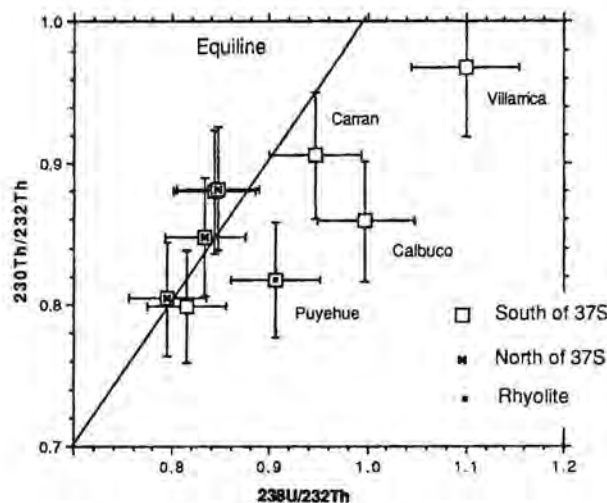


FIG. 5. ^{238}U - ^{230}Th data for Southern Volcanic Zone Andean lavas. One sigma uncertainty for this study is 2%; the uncertainty bars shown are all 5%. Only lavas from the thin crust segment south of 37°S display disequilibrium and ^{238}U enrichment. Other lavas are in radioactive equilibrium.

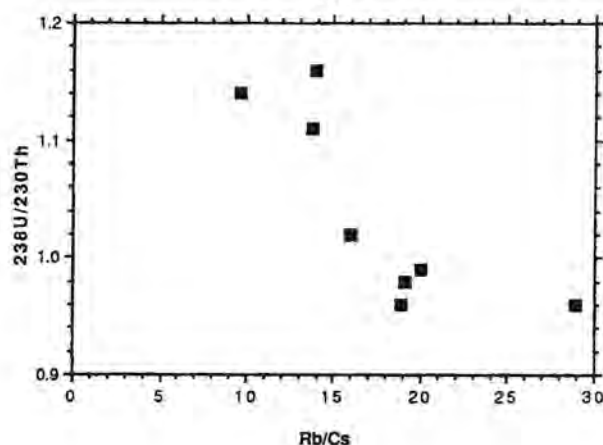
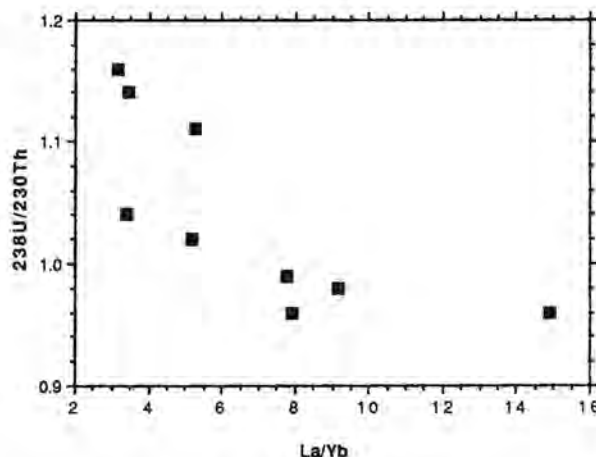
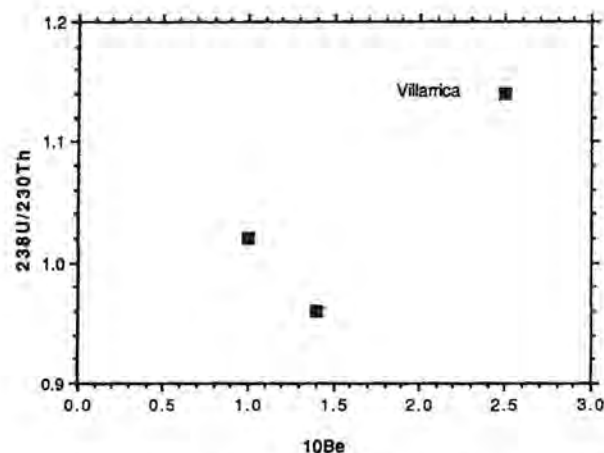


FIG. 6. Correlation of Rb/Cs, La/Yb, and ^{10}Be with $^{238}\text{U}/^{230}\text{Th}$ ratio (a measure of radioactive disequilibrium). High ^{10}Be , low Rb/Cs and La/Yb indicate greater slab derived inputs to magmas (López-Escobar *et al.*, 1977; Morris *et al.*, 1985; Hickey *et al.*, 1986; Tormey *et al.*, in press), and in all cases corresponds to high $^{238}\text{U}/^{230}\text{Th}$. This coupled with the observation that only arcs show ^{238}U enrichment (Allegre and Condomines, 1982), indicates that the source of the ^{238}U enrichment is derived from subducted oceanic crust and sediments.

Descabezado Grande, $^{238}\text{U}/^{232}\text{Th}$ increases from 0.62 in an andesite to 0.62 - 0.69 in dacites, and to 0.83 in a rhyolite. At Maca the opposite is observed: 1.0 in a basalt decreasing to 0.69 in basaltic andesite. Considering all the samples, U/Th in basalts ranges from 0.7 to 1.0 (a single sample from Osorno is 2.7), and U/Th in rhyolites ranges from 0.61 to 0.89. The data are currently too limited to determine the relative partition coefficients of U and Th during evolution from basalt to rhyolite, but generally Th is more incompatible than U in the crustal environment.

Compared to other arc lavas, Andean lavas have low $^{230}\text{Th}/^{232}\text{Th}$ ratios (0.82-0.96; Fig. 4; Table 1). Figure 5 illustrates that lavas from south of 37°S tend to either have higher $^{230}\text{Th}/^{232}\text{Th}$ than lavas from north of 37°S or will evolve to higher $^{230}\text{Th}/^{232}\text{Th}$ as

they establish radioactive equilibrium. Lavas from the thick crust segment (north of 37°S) are in radioactive equilibrium, but lavas from the thin crust segment, Villarrica (39°25'S), Puyehue (40°30'S) and Calbuco (41°15'S), are in disequilibrium with ^{238}U enrichment. Similar results were obtained by Sigmarsson *et al.* (1991), who in addition found that a basalt from Osorno (41°S) was in radioactive disequilibrium with ^{238}U enrichment. Figure 6 shows that disequilibrium as measured by the $^{238}\text{U}/^{230}\text{Th}$ ratio (determined from the ratio of $^{238}\text{U}/^{230}\text{Th}$ to $^{230}\text{Th}/^{232}\text{Th}$), correlates with ^{10}Be , Rb/Cs and La/Yb, which are common parameters of subduction zone inputs (López-Escobar *et al.*, 1977; Morris *et al.*, 1985; Hickey *et al.*, 1986; Tormey *et al.*, in press; see Sigmarsson *et al.*, 1991 for correlation with $^{10}\text{Be}/^9\text{Be}$).

DISCUSSION

PETROGENETIC CONSTRAINTS

Currently, only lavas from volcanic arcs (either island or continental) show ^{238}U activity enrichments over the activity of the daughter ^{230}Th (Allegre and Condomines, 1982; Condomines *et al.*, 1988). All cases of U-Th disequilibrium in non-arc oceanic lavas are enrichments in ^{230}Th , that is, to the left of the equiline. The difference between island arcs and oceanic lavas therefore probably arises in the subduction zone. Newman *et al.* (1986) observed that the few cases of U-Th disequilibrium in historic lavas from Cascades continental arc are towards Th enrichment, postulating that either partial melting ($D_{\text{Th}} < D_{\text{U}}$; Allegre and Condomines, 1982; Condomines *et al.*, 1988) or contamination of subcrustal magmas with high Th/U crustal melts could be responsible for this enrichment. In other words, while subduction zone processes would cause U enrichments, crustal contamination processes would cause Th enrichments. The U and Th activities measured in this study are consistent with U being more compatible than Th during magmatic differentiation in the crust.

In contrast to the Cascades continental arc, the cases of ^{238}U - ^{230}Th disequilibrium in Andean lavas are ^{238}U enrichment (Figs. 4, 5), and this disequilibrium is observed in lavas of the thin crust segment south of 37°S. Lavas from the thick crust segment of the SVZ, where crustal contamination has been shown to occur (Hildreth and Moorbath, 1988; Tormey *et al.*, in press) show radioactive equilibrium. The ^{238}U

enrichment in primary magmas from the thick crust segment may have been erased by contamination with high Th/U crustal melts (Tormey, 1989). In the thin crust segment of the SVZ, where crustal contamination is minor (Hickey *et al.*, 1986; Tormey *et al.*, 1991), the ^{238}U enrichment remains.

In addition to $^{238}\text{U}/^{230}\text{Th} > 1$, lavas south of 37°S have higher ^{10}Be (Morris *et al.*, 1985), lower Rb/Cs (Tormey *et al.*, in press), higher degree of mantle melting and lower La/Yb (López-Escobar *et al.*, 1977; Tormey *et al.*, in press) than lavas north of 37°S. These parameters indicate a greater proportion of material derived from the subducted slab in lavas from south of 37°S than north of this latitude. Disequilibrium, as measured by the $^{238}\text{U}/^{230}\text{Th}$ ratio, correlates with all these subducted slab features (Fig. 6; see Fig. 3 of Sigmarsson *et al.*, 1991 for $^{10}\text{Be}/^9\text{Be}$). This observation suggests that the recent ^{238}U enrichment is caused by slab-derived components. Under the oxidizing conditions expected from dehydration of the subducted oceanic crust, U would be in the +6 oxidation state and would readily fractionate from Th. Fractionation of U from Th under these conditions enriches the derived aqueous phase in U, and the overlying metasomatized mantle in turn is enriched in ^{238}U .

TIME SCALE AND MECHANISM OF MAGMA ASCENT

Accepting that the subducted oceanic crust is the source of U-Th disequilibrium and enrichment of ^{238}U over ^{230}Th , then it is possible to constrain the

time elapsed between magma generation and eruption. The time required to reestablish radioactive equilibrium is between one and three half lives of ^{230}Th , that is, between 7.52×10^4 and 2.26×10^5 years. This range is more conservative than the value of 2.0×10^4 years used by Sigmarsson *et al.* (1991).

If the depth to the zone of magma generation is approximately 100 kilometers (Barazangi and Isacks, 1976; Bevis and Isacks, 1984), then the magmatic ascent rate is 0.4 to 1.33 meters/year. This chemically derived ascent rate compares favorably with ascent rates calculated from physical principles. Ascent rates for fracture transport range from kilometers/day to meters/day (Eaton and Murata, 1970; Szekely and Reitan, 1971; Shaw, 1980). Ascent rates for diapirs range from 3 to 300 meters/year (Spera, 1980; Marsh, 1982). Ascent rates calculated for porous media flow driven by buoyancy of the partial melt range from centimeters/year to millimeters/year (Spera, 1980; McKenzie, 1984). Based on this comparison between the ascent rate constrained by chemistry and ascent rates constrained by physics, only a small portion of the ascent of magmas in subduction zones can be by porous media flow. Most of the ascent must be as diapirs or through fractures. Furthermore, south of 37°S storage of basaltic lavas during ascent through the mantle and crust for more than 20,000 years is inconsistent with the chemical data.

Th/U IN THE SOURCE OF ANDEAN MAGMAS

There are three methods to infer Th/U in the source of Andean magmas. The first is to directly measure Th/U in basaltic lavas (K_m); this method gives the shortest time scale measurement. The second method is the ratio of ^{232}Th (parent of the ^{232}Th decay chain) to ^{230}Th (daughter product of ^{238}U decay); this method, yielding K_{Th} , measures a longer time scale than K_m . The longest time scale Th/U, K_{Pb} , is measured by the ratio of ^{208}Pb (final nuclide of the ^{232}Th decay chain) to ^{206}Pb (final nuclide of the ^{238}U decay chain). For the nine lavas used in the first portion of this study, K_{Pb} is 3.95 (Data from Hickey *et al.*, 1986), K_{Th} is 3.53, and K_m is 3.41. For comparison, bulk earth Th/U is about 3.9, and the Th-depleted source of mid-ocean ridge basalts is about 2.3 (Galer and O'Nions, 1985). For the Andean lavas, $K_{Th} < K_{Pb}$ implies that there has been U enrichment on a shorter time scale than measured by the Pb isotopic system. $K_m < K_{Th}$ implies further U enrichment

of the mantle source on a shorter time scale than measured by the Th isotopic system. One explanation for the decreasing Th/U with time observed in the Andean magmas is progressive metasomatism of the mantle by low Th/U fluids derived from the subducted oceanic crust. Arc magmatism has occurred semi-continuously since the Jurassic in the southern Andes, and metasomatism by low Th/U fluids derived from subducted oceanic crust may be decreasing Th/U in the overlying mantle.

ENVIRONMENTAL EFFECTS

Of the estimated annual effective dose equivalent from natural sources of radiation in areas of normal background, about 50% is due to the ^{238}U -series and about 17% to the ^{232}Th -series. Radon-isotopes are the major component of both series (United Nations Scientific Committee, 1982). The range of ^{238}U and ^{232}Th specific activities obtained in this work (3-42 and 3-69 Bq/kg respectively), is similar to the range reported for soils from areas of normal natural activity (10-50 and 7-50 Bq/kg respectively; United Nations Scientific Committee, 1982). The ^{238}U abundances obtained in this work are also similar to the range of total U abundances reported by Zentilli and Dostal (1977) for Quaternary volcanic rocks ranging in composition from basalts to rhyolites from Central Andean ($16-28^\circ\text{S}$) centers. There is a positive correlation between the variables ^{238}U , ^{232}Th , and SiO_2 in the analyzed rocks (Fig. 3). Because dacitic and rhyolitic eruptions are more frequent in the Central Volcanic Zone of the Andes ($16-28^\circ\text{S}$) and in the northern part of the SVZ ($33-37^\circ\text{S}$) than south of 37°S , populations in these volcanic areas are exposed to a higher natural radiation dose than populations living in areas where basaltic rocks predominate. However, in terms of health risk, inhalation of fine ash particles during a volcanic eruption is comparable to inhalation of fine soil particles during a dust storm.

Because radon is thermodynamically stable as a gas under the conditions of a volcanic eruption, fractionation of radon into the vapor plume during an eruption will lead to a greater abundance of radon than predicted by the U and Th contents of the magma. Hence, although the ^{238}U and ^{232}Th specific activities are within the normal range described for soils, gases associated with volcanic eruptions may

be highly enriched in radon (Kuroda *et al.*, 1984; Lambert *et al.*, 1985-1986). Continuous or semi-continuous degassing of radon from volcanoes may also elevate the radon content of the atmosphere near them, and hence present an exposure pathway to humans through inhalation. At very high exposure levels, this dose may cause an acute reaction. Chronic

health effects may be suffered as a result of periodic or continuous degassing of magmas within volcanoes through fractures, fumarole fields, and other hydrothermal activity. Quantifying this radon flux and the resulting human exposure is beyond the scope of this paper.

CONCLUSIONS

1. Lavas from the 33-37°S segment of the SVZ of the Andes, where the continental crust is relatively thick (~55 km at 33°S to ~35 km at 37°S) and crustal contamination is evident (Hildreth and Moorbath, 1988; Tormey *et al.*, 1991), show ^{238}U - ^{230}Th equilibrium; lavas from the 37-46°S segment, where the continental crust is relatively thin (30-35 km) and crustal contamination is minor (Hickey *et al.*, 1986; Tormey *et al.*, 1991), show either ^{238}U - ^{230}Th equilibrium or disequilibrium and ^{238}U enrichment.
2. The correlation of ^{238}U - ^{230}Th with subducted slab indicators such as ^{10}Be , Rb/Cs, and La/Yb, as well as the observation that disequilibrium with ^{238}U enrichment only occurs in subduction zones, implies that the U-Th fractionation as ^{238}U enrichment are due to subduction zone processes rather than crustal ones. If so, the magmatic ascent rate is between 0.4 to 1.33 m per year. This rate compares favorably with rates calculated for magma ascent in fractures or as diapirs, but is too rapid for a significant portion of the ascent to be by porous media flow.
3. The source of Andean magmatism appears to have been metasomatized by low Th/U fluids over time, leading to a lowering of Th/U as measured by Pb isotopes (K_{Pb}), Th isotopes (K_{Th}), and directly in the lavas (K_{m}).
4. The ^{238}U and ^{232}Th specific activities found in Quaternary volcanic rocks from the SVZ of the Andes (3-42 and 3-69 Bq/kg, respectively) are similar to the concentration ranges of these nuclides reported for soils from areas of normal natural activity (10-50 and 7-50 Bq/kg, respectively). Because U and Th correlate with SiO_2 , people living in areas of more frequent rhyolitic eruptions (CVZ and northern SVZ) are exposed to a higher radiation burden. The most critical component of the radiation dose due to proximity of volcanoes is caused by radon degassing from magmas, both during eruptions and on a semi-continuous basis from fractures and hydrothermal activity.

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REFERENCES

- Allegre, C.L. 1968. ^{230}Th dating of volcanic rocks: a comment. *Earth and Planetary Science Letters*, Vol. 5, p. 209-210.
- Allegre, C.L.; Condomines, M. 1976. Fine chronology of volcanic processes using ^{238}U - ^{230}Th systematics. *Earth and Planetary Science Letters*, Vol. 28, p. 395-406.
- Allegre, C.L.; Condomines, M. 1982. Basalt genesis and mantle structure studied through Th-isotopic geochemistry. *Nature*, Vol. 299, p. 21-24.
- Barazangi, M.; Isacks, B. 1976. Spatial distribution of earthquakes and subduction of the Nazca plate beneath South America. *Geology*, Vol. 4, p. 686-692.
- Bevis, M.; Isacks, B. 1984. Hypocentral trend surface analysis: Probing the geometry of subduction zones. *Journal of Geophysical Research*, Vol. 89, p. 6153-6170.
- Condomines, M.; Hemond, C.; Allegre, C.L. 1988. U-Th-Ra radioactive disequilibria and magmatic processes. *Earth and Planetary Science Letters*, Vol. 90, p. 243-262.
- Eaton, J.P.; Murata, K. 1970. How volcanoes grow. *Science*, Vol. 132, p. 925-938.
- Galer, S.; O'Nions, R. 1985. Magmatogenesis and the mapping of chemical and isotopic variations in the mantle. *Chemical Geology*, Vol. 56, p. 45-621.
- Hickey, R. L.; Frey, F. A.; Gerlach, D. C.; López-Escobar, L. 1986. Multiple sources for basaltic arc rocks from the Southern Volcanic Zone of the Andes (34° - 41°S): Trace element and isotopic evidence for contributions from subducted oceanic crust, mantle and continental crust. *Journal of Geophysical Research*, Vol. 91, p. 5963-5983.
- Hildreth, W. E., Moorbath, S., 1988. Crustal contribution to arc magmatism in the Andes of Central Chile. *Contributions to Mineralogy and Petrology*, Vol. 98, p. 455-489.
- Knopke, J.; Kühn, W. 1985. Determination of uranium in soils samples by different analytical extraction methods. In *International Contact Seminar in Radioecology, Proceedings, Preprint*. Swedish University of Agriculture Sciences. Uppsala.
- Kuroda, P.K.; Liou, J.C.H.; Banavali, A.D.; Akridge, J.D.; Burchfield, L.A. 1984. Polonium- 210 fallout from the 1980 eruption of Mount St Helens and the mystery cloud of 1982. *Geochemical Journal*, Vol. 18, p. 55-60.
- Lambert, G.; Le Cloarec, M.F.; Ardouin, B.; Le Rouley, J.C. 1985-1986. Volcanic emission of radionuclides and magma dynamics. *Earth Planetary Science Letters*, Vol. 76, p. 185-192.
- López-Escobar, L.; Frey, F.; Vergara, M. 1977. Andesites and high alumina basalts from the central-south Chile High Andes: Geochemical evidences bearing on their petrogenesis. *Contributions to Mineralogy and Petrology*, Vol. 63, p. 199-228.
- Marsh, B.D. 1982. On the mechanics of igneous diapirism, stopping, and zone melting. *American Journal of Sciences*, Vol. 282, p. 808-855.
- McKenzie, K. 1984. The generation and compaction of partially molten rock. *Journal of Petrology*, Vol. 25, p. 713-765.
- Morris, J.S.; Tera, F.; Harmon, R.S.; López-Escobar, L.; Klein, J.; Middleton, R. 1985. Be^{10} in lavas from the Andean Southern Volcanic Zone (35° - 40°S): Evidence for sediment subduction. *Universidad de Chile, Departamento de Geología y Geofísica, Comunicaciones*, No. 35, p. 157-160.
- Newman, S.; Macdougall, J.D.; Finkel, R.C. 1984. ^{230}Th - ^{238}U disequilibrium in island arcs: evidence from the Aleutians and the Marianas. *Nature*, Vol. 308, p. 268-270.
- Newman, S.; Macdougall, J.D.; Finkel, R.C. 1986. Petrogenesis and ^{230}Th - ^{238}U disequilibrium at Mt. Shasta, California, and in the Cascades. *Contributions to Mineralogy and Petrology*, Vol. 93, p. 195-206.
- Shaw, H. 1980. The fracture mechanism of magma transport from the mantle to the surface. In *Physics of Magmatic Processes* (Hargraves, R.B.; editor). Princeton University Press, p. 201-262.
- Sigmarrsson, O.; Condomines, M.; Morris, J.D.; Harmon, R.S. 1991. Uranium and ^{10}Be enrichments by fluids in Andean arc magmas. *Nature*, Vol. 346, p. 163-165.
- Spera, F. 1980. Aspects of magma transport. In *Physics of Magmatic Processes* (R.B. Hargraves; editor). Princeton University Press, p. 263-323.
- Szekely, J.; Reitan, P.H. 1971. Dike filling by magma intrusion and by explosive entrainment of fragments. *Journal Geophysical Research*, Vol. 76, p. 2602-2608.
- Tormey, D.R. 1989. Geology and geochemistry of the active Azufre-Planchón-Peteroa volcanic center ($35^{\circ}15'\text{S}$, Southern Andes): Implications for cordilleran arc magmatism. Ph.D. Thesis. Massachusetts Institute of Technology. 331 p.
- Tormey, D.R.; Hickey-Vargas, R.L.; Frey, F.A.; López-Escobar, L. (In press). Recent lavas from the Andean volcanic front (33° - 42°S): Interpretations of along-strike compositional variations. *Geological Society of America, Special Paper*, No. 265.
- United Nations Scientific Committee on the Effects of Atomic Radiation. 1982. Ionizing radiation: sources and biological effects. *Report to the General Assembly*, 773 p.
- Zentilli, M.; Dostal, J. 1977. Uranium in volcanic rocks from the Central Andes. *Journal of Volcanology and Geothermal Research*, Vol. 2, p. 251-258.

APPENDIX

MATERIALS AND METHODS

The techniques used for the first part of the study to determine disequilibrium between ^{238}U and ^{230}Th are described in detail by Tormey (1989). Three to 7 grams of sample were dissolved at 100°C using 100 ml 3:1 HNO_3 -HF; while drying down, addition of ~ 10 ml HClO_4 inhibits formation of insoluble fluorides. The sample was then conditioned with ~ 150 ml 6.2N HCl , dried down and conditioned again in 1:1 (8N) HNO_3 . Four column chromatography steps are required to isolate Th (Tormey, 1989). Thorium was then electroplated, and the activity ratio $^{230}\text{Th}/^{232}\text{Th}$ was determined from their α emissions utilizing an Ortec alpha spectrometer and multi-channel analyzer. Precision based on duplicate analyses of five samples is better than the one sigma counting statistics for the ratio of 1% (Tormey, 1989). U and Th abundances (hence $^{238}\text{U}/^{232}\text{Th}$) were measured by isotope dilution on a $12''$ mass spectrometer. Within run precision is better than 0.1%, but the chance of within run fractionation leads us to a 1% precision.

For the second part of the study, 23 lava samples from volcanic centers located between latitudes $33^\circ 00'\text{S}$ and $46^\circ 00'\text{S}$ (Fig. 1, Table 1) were analyzed. ^{238}U was extracted by using the hydrogen fluoride method as described by Knopke and Kühn (1985) and Köhler *et al.* (1988). According

to this method, 0.185 Bq of ^{232}U were added as a spike to 5 g of lava powder and the mixture was digested with 40% HF during 1-2 days. The resulting solution was evaporated with HNO_3 and the residue dissolved in 2M HNO_3 . Sodium nitride was added to reduce the Fe^{+3} in solution. The latter was then salted out with NaNO_3 . This mixture was shaken with triethyl phosphine oxide in cyclohexane and two phases were obtained. The aqueous one was discharged, and the phase containing the uranium, was treated with saturated NH_4HCO_3 . Again two phases were formed. The aqueous one, which in this case contains the uranium, was added to H_2SO_4 and its pH was adjusted between 2 and 3 by using an internal indicator. Uranium was electroplated from this solution. The α emissions were measured with an Ortec Dual 576 α -spectrometer connected to a multichannel analyzer. At least two measurements of ^{238}U activity were made per sample, and their respective averages and absolute errors were calculated.

The abundances of SiO_2 were determined by atomic absorption at the Departamento de Geología of the Universidad de Chile. According to their SiO_2 content, expressed in weight per cent, the samples analyzed were classified into basalts ($\text{SiO}_2 < 52\%$), basaltic andesites ($\text{SiO}_2 = 52-56\%$), andesites ($\text{SiO}_2 = 56-63\%$), dacites ($\text{SiO}_2 = 63-70\%$), and rhyolites ($\text{SiO}_2 > 70\%$).