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RADIOACTIVE ORPHANS IN BARITE-RICH CHIMNEYS, AXIAL CALDERA, JUAN DE FUCA RIDGE

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ABSTRACT

Gamma-ray measurements on samples from two unusually radioactive barite-rich chimneys on Axial Seamount, Juan de Fuca Ridge, gave equivalent concentrations of ~0.1% Th and 0.2% U. The gamma-ray emitting nuclides in both the Th- and U-decay series were found to be unsupported by their parents ^{232}Th and ^{238}U and are therefore orphans. Measurements on three chimney fragments showed that ^{228}Th was not in radioactive equilibrium with ^{228}Ra, which allowed their apparent ages and initial ^{226}Ra/^{228}Ra ratios to be determined. Based on the ^{228}Th/^{228}Ra ages of 3 and 6 years for two adjacent samples, a maximum growth rate of 6 cm per year was established. Calculated initial ^{226}Ra/^{228}Ra ratios indicate that the fluids which precipitated the barite took 20 years to acquire their radiochemical signatures from mid-ocean ridge basalts.

Keywords: Axial Seamount, Juan de Fuca, chimneys, barite, sulfides, radioactivity, Th- and U-decay series, disequilibrium.

SOMMAIRE

Les mesures de rayons gamma faites sur des échantillons provenant de deux cheminées riches en barytine anormalement radioactive du mont sous-marin Axial (crête de Juan de Fuca) ont révélé des concentrations équivalentes d'environ 0.1% Th et 0.2% U. On a constaté qu'il n'y a pas de ^{232}Th ni de ^{238}U reliés à la présence des nucléides émetteurs de rayons gamma dans les familles radioactives du Th et de l'U; ces nucléides sont donc orphelins. Les mesures prises à partir des trois fragments des colonnes de minerai ont montré que le ^{228}Th n'est pas en équilibre radioactif avec le ^{228}Ra, ce qui a permis de déterminer l'âge apparent et les rapports initiaux ^{226}Ra/^{228}Ra des fragments. Selon les âges ^{228}Th/^{228}Ra de 3 et 6 ans, déterminés à partir de deux échantillons contigüs, le taux de croissance maximal est de 6 cm par année. Le calcul des rapports initiaux ^{226}Ra/^{228}Ra indique que les fluides qui ont précipité la barytine ont mis 20 ans à acquérir leurs signatures radiochimiques à partir des basaltes de la dorsale océanique.

Mots-clés: mont sous-marin Axial, crête de Juan de Fuca, colonne de minerai, barytine, sulfures, radioactivité, familles radioactives du Th et de l'U, déséquilibre.

INTRODUCTION

In the marine environment, the uranium and thorium decay products are frequently isolated from their parent nuclides. By monitoring these decay products it is possible to study the geochronology of corals, shells, sedimentation, and more recently, hydrothermal deposits (Lalou & Brichet 1982, Kadko *et al.* 1985). Lalou & Brichet (1987) discussed four potential methods for dating hydrothermal deposits to ~300 Ka in age.

For dating recent hydrothermal deposits with an age of less than about 20 years, the ^{228}Th/^{228}Ra method is potentially useful. The method was attempted by Lalou & Brichet (1982) for a hydrothermal deposit with an estimated age of 50 years. Lalou & Brichet (1987) have verified the method by dating a piece of active sulfide chimney, five years after it was sampled. Turekian *et al.* (1979) have used the method to determine the growth rate of clams. Later, Turekian & Cochran (1986) showed how the initial (^{228}Th/^{228}Ra) activity ratio of the clams could be used to estimate residence times of radium-bearing vent fluids.

During the summer of 1986, three areas of hydrothermal activity in the caldera of Axial Seamount on the Juan de Fuca Ridge were sampled using the Deep Submersible PISCES IV. At one site, samples were collected from two chimneys of massive barite. Subsequent examination of the barite samples with a total-count scintillometer showed that they were unusually radioactive. Using a portable gamma-ray spectrometer and comparing the chimney fragments with uranium and thorium standards, the fragments were found to have equivalent concentrations of ~0.1% Th and 0.2% U. These levels of radioactivity, due to decay products of ^{226}Ra and ^{228}Ra, are two orders of magnitude higher than those previously reported for similar chimney structures (Finkel *et al.* 1980, Lalou & Brichet 1982, Kadko *et al.* 1985). These initial radioactivity measurements prompted a more detailed gamma-ray spec-

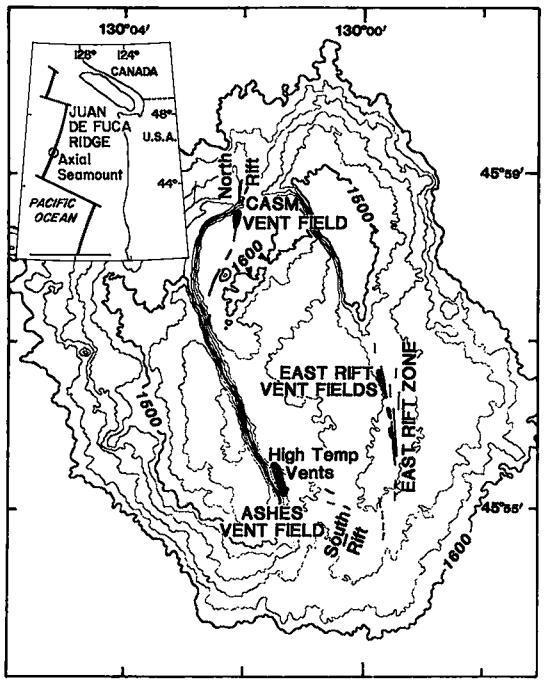

FIG. 1. Bathymetric chart (in meters) of Axial Seamount showing the CASM site and associated vent fields.

trometric investigation which revealed that ^{226}Ra and ^{228}Ra were unsupported by their parents ^{238}U and ^{232}Th and can therefore be considered orphans. This disequilibrium in the thorium series allowed the apparent age of the barite samples to be determined using the $^{228}Th/^{228}Ra$ method. In addition, the calculated initial $^{226}Ra/^{228}Ra$ ratios were used to estimate the length of time the hydrothermal fluids that produced the barite chemically reacted with basalt to acquire their radium signatures.

GEOLOGICAL SETTING

Axial Seamount is a prominent shield volcano astride the Juan de Fuca Ridge at about 46°N, 130°W. Its peak is 900 m above the abyssal plain surrounding the ridge. A distinctive caldera forms the summit of the volcano. The caldera has a U-shape, with steep walls about 100 m high that form the north, west and east sides, and a drain-out area in place of the south wall (Fig. 1). The floor of the caldera, at a depth of 1545 m, is composed of fresh glassy sheet flows, with less common lobate and pillowed flows near the margins of the caldera. Recent sheet flows are commonly deformed into pressure ridges, and form a highly irregular surface. Fissures are uncommon in the caldera, except near its northwest corner, the CASM (Canadian American Seamount Expedition) site, where a prominent fissure transects both the wall and the floor (CASM 1985).

During the summer of 1983, hydrothermal activity was discovered in three areas of the caldera. Low-temperature (*ca.* 20°C) fluids were emanating from the fissure at the CASM site (Hannington & Scott 1988). Hydrothermal precipitates within this fissure are masked by prolific biological material. Immediately to the east of the fissure is a single chimney, named Lamphere Spire (Hannington 1986). Approximately 3 km to the south, along the southwest wall, an area of high-temperature (*ca.* 330°C) hydrothermal venting (ASHES Vent Field) was studied during the 1986 dive series (Fig. 1). The East Rift fields, an area of low-temperature venting, appears from sidescan sonar records to be associated with a rift zone.

The chimneys investigated are of three types. Sulfide chimneys, the most common, are composed of near-massive sphalerite, with minor amounts of pyrite and chalcopyrite. The outer zones of these chimneys contain variable but generally minor amounts of barite. The second type, found at the CASM site, has massive barite walls up to 20 cm thick. The third type of chimney is composed almost entirely of anhydrite.

SAMPLE DESCRIPTION

Samples XL1731-1.8 (18 × 10 × 12 cm) and XL1731-6.7A1 (15 × 11 × 19 cm) were collected from the Lamphere chimney group at the CASM site (Hannington *et al.* 1986). They consist largely of barite (*ca.* 30%) and amorphous silica (*ca.* 50%) with minor pyrite (*ca.* 2%) and sphalerite (*ca.* 1%). Sample XL1731-1.8 is a fragment of the main Lamphere spire that fell on a metal collecting plate; XL1731-6.7A1 consists of pieces of a small (~50 cm) bulbous vent that grew from the base of Lamphere spire about 0.5 m from the main stack. None of the Lamphere vents was hydrothermally active at the time of sampling. Hannington & Scott (1988) recorded low temperatures (19°C) in one chimney of this group in 1983. Figure 2 shows the base of the main Lamphere spire and the broken-off sample XL1731-1.8.

Sample XL1731-1.8 was sectioned laterally into four pieces (Fig. 3). Because of the friable nature of the sample, it was soaked in an epoxy resin before sectioning. After sectioning, small conduits and cavities lined with mm-size barite crystals were visible (Fig. 3c). The irregular zones in Figure 3c appear to be roughly concentric to interior fluid conduits. Some typical material from the interior part of the sample (Fig. 4) was pulverized and analyzed (XL1731-1.8B). Sample XL1731-6.7A1, a collection of small fragments from the second vent structure, also was pulverized and analyzed. These fragments also displayed numerous small vugs lined with barite crystals.

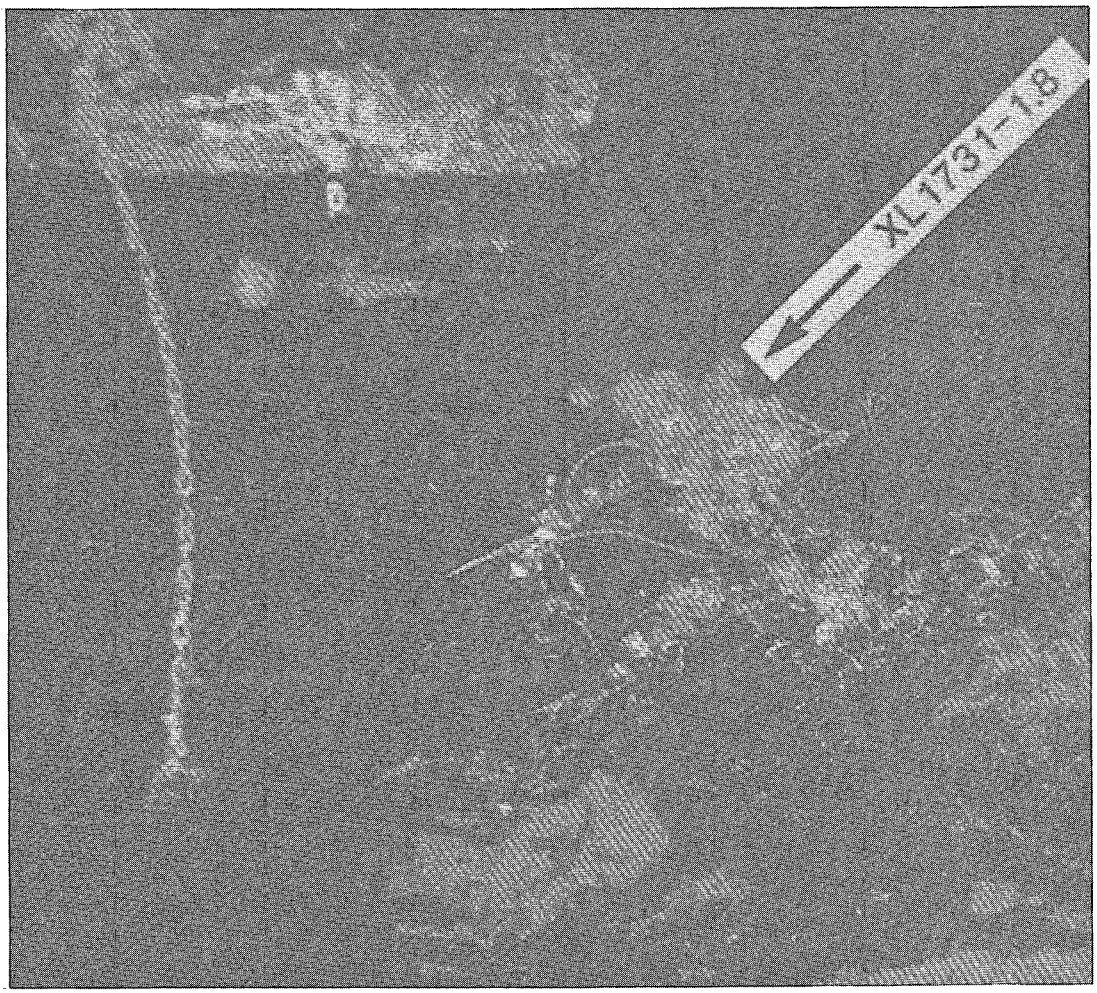

FIG. 2. Base of Lamphere Spire, the main chimney at the CASM site. The large fragment near the base is sample XL1731-1.8. The straight section of chain is ~130 cm long.

An autoradiograph (Fig. 5) of the lower section (Fig. 3d) indicates that greater radium activity is centered on the vuggy interior than at the leached crusted margins. The brightest areas correspond to cavities lined with crystalline barite. This brightness could be a geometric effect, or it could arise because the fluids containing the radium flow through the conduits and radium is concentrated on their surfaces.

ANALYTICAL METHODS

Gamma-ray measurements were made with an Aptec, high-purity germanium coaxial crystal spectrometer contained in a lead castle 10 cm thick. The crystal has an active volume of 56 cm, an Al window 0.6 mm thick, and a rated efficiency of about 12% at 1.33 MeV relative to a 76×76 mm (3×3 inch) sodium iodide crystal. A Canberra Series 85 multichannel analyzer system with 8192 channels was used for data aquisition.

Initial measurements were made on the whole-rock sample XL1731-1.8A (Fig. 3d) which was placed directly on the spectrometer window. Uranium, thorium, and uranium–thorium ore reference materials (BL–3, OKA–2, and DH–1a, respectively) were also measured for comparison. In both BL–3 and DH–1a, ^{226}Ra is known to be in equilibrium with ^{238}U (Ingles *et al.* 1977, Smith & Steger 1983, Grasty & Dyck 1984). The thorium ore, OKA–2 (Smith & Grasty 1984), would also be in equilibrium because the ^{232}Th series reaches equilibrium in a few tens of years.

Powdered samples of about 50 g of each reference material were vacuum-sealed in small metal con-

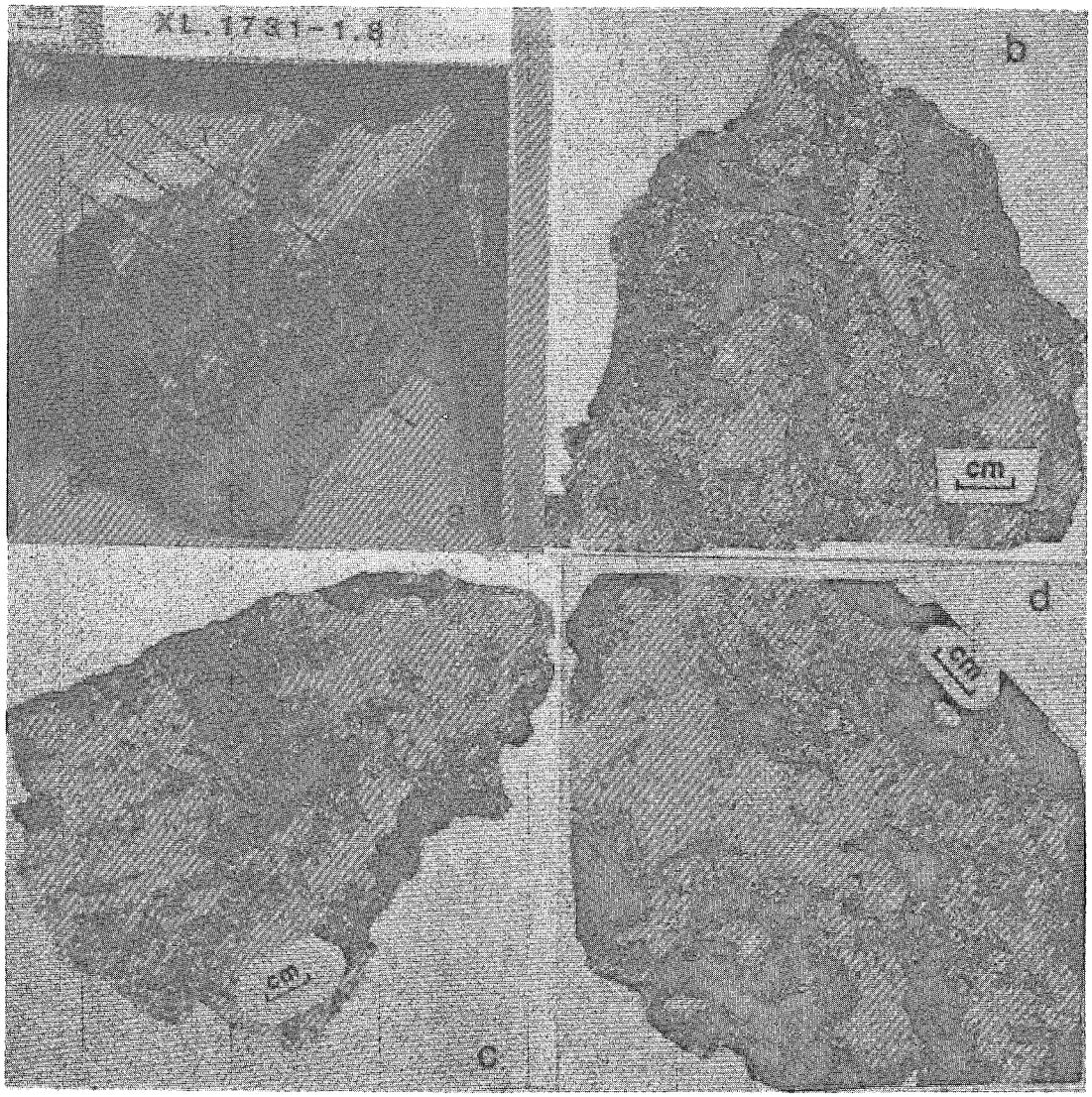

FIG. 3. Barite-rich chimney fragment (XL1731-1.8) from CASM site, Axial Seamount caldera. (a) View of vent fragment as collected – note large central fluid conduit exposed on left. Speciment was cut (– – – –) into two sections designated U and L, a top T, and a base, B. (b) Detailed view of section T displays a weathered exterior of replaced and infilled tube worms and other vent fauna. (c) Upper section (U) reveals the porous nature of the chimney; dark areas are conduits commonly lined with coarse grey to black barite crystals. White areas consist of plumose arrays of barite blades. (d) Lower section (L), designated XL1731-1.8A, was impregnated with epoxy, partly polished, and then subjected to autoradiography and radiochemical analyses. Dark areas are conduits or cavities (as for 1c). A cm scale is placed on the outer wall with the main interior conduit (seen in 1a) exposed on the lower left. Section L weighed 317.5 g.

tainers for at least 30 days. This prevents disequilibrium between ^{226}Ra and its decay products ^{214}Pb and ^{214}Bi due to emanation of the intermediate gaseous nuclide, ^{222}Rn. The gamma-ray spectra of a typical barite sample and the uranium and thorium reference ores are shown in Figure 6 for the energy range of about 750 to 1050 keV.

The ^{226}Ra to ^{228}Ra activity of sample XL1731-1.8A (lower section) was measured on the whole rock by comparing the ratios of gamma-ray peaks of similar energy for the barite and the uranium–thorium ore DH–1a, as described by Smith & Grasty (1984). The ^{228}Ra to ^{228}Th activity ratio was also measured using the thorium reference ore (OKA–2) as the

calibration material. Calculated ^{228}Ra to ^{228}Th activity ratios (Table 1) are based on four pairs of gamma rays of similar energy. The ^{226}Ra to ^{228}Ra activity ratio was calculated using three pairs of gamma rays (Table 2). In utilizing the gamma-ray peaks from ^{214}Pb and ^{214}Bi to monitor ^{226}Ra, emanation of the gas ^{222}Rn must be low. This was confirmed by monitoring for 10 days the gamma-ray emission of a sealed barite sample which showed no buildup of radon decay products.

Quantitative measurements were performed on the two powdered samples XL1731-1.8B and XL1731-6.7A1. The ^{226}Ra, ^{228}Ra and ^{228}Th activities of two 17-g samples were measured following the procedure described by Desgagné & Smith (1985). The activities per gram of the samples are presented in Table 3 in SI units of Becquerels (Bq), where one Becquerel is one atomic disintegration per second. To conform with current practice, activities are shown in the text in brackets, *e.g.*, (^{226}Ra). All errors on the activity measurements are at the 95% confidence level.

U and Th concentrations of the two powdered samples were also measured by neutron activation. These concentrations were converted to Bq/g, as shown in Table 3, using the relationships: 1μg/g of uranium = 0.0123 Bq/g of ^{238}U, and 1μg/g of thorium = 0.00404 Bq/g of ^{232}Th.

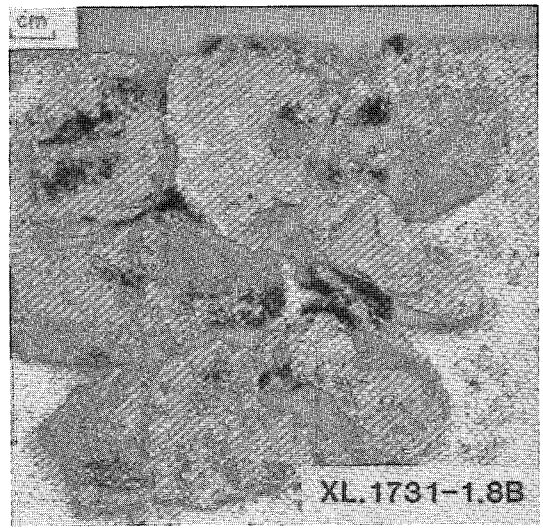

FIG. 4. Coarsely crystalline barite fragments from vuggy interior of vent sample (XL1731-1.8B). Sub-mm black barite prisms line cavities in white plumose barite matrix. Pieces are from the lower left area (near central conduit) of the basal portion B (Fig. 3a). These pieces were crushed for chemical and radiochemical analyses.

ANALYTICAL RESULTS

Almost all the gamma-ray peaks in the uranium spectrum are also in the barite. These gamma-ray peaks, which are due primarily to ^{214}Bi (half-life 19.9 minutes) and ^{214}Pb (half-life 26.8 minutes), are indicative of the presence of the long-lived nuclide ^{226}Ra (half-life 1600 years). A notable feature of the barite spectrum is the absence of the 1001 keV gamma-ray peak from ^{234}Pa in the uranium series. This peak is clearly present in the equilibrium uranium spectrum (Fig. 6). As ^{234}Pa will reach equilibrium with ^{238}U within a few months (the intermediate isotope ^{234}Th has a 24-day half-life), this indicates that the parent ^{238}U is absent or at a very low concentration. The low concentration of ^{238}U compared to ^{226}Ra was also suggested by the absence of: 1) 63 and 93 keV gamma rays from ^{234}Th; 2) uranium *K* X-rays (94.5 and 98.4 keV); and 3) 144 keV gamma rays from ^{235}U. The low concentration of ^{238}U was confirmed by neutron activation (Table 3).

Gamma rays from ^{210}Pb at 46 keV (outside the energy range of Fig. 6) were evident in the uranium spectrum, but not in the barite spectrum. As ^{210}Pb has a half-life of 22 years, it will reach equilibrium with any ^{226}Ra in about 100 years, provided there is no significant emanation of ^{222}Rn. This absence of a ^{210}Pb peak in the barite sample suggests it is considerably younger than 100 years. However, because the sample has a high percentage of barium, low-energy gamma rays are significantly attenuated. Therefore, the presence of ^{210}Pb could not be completely discounted.

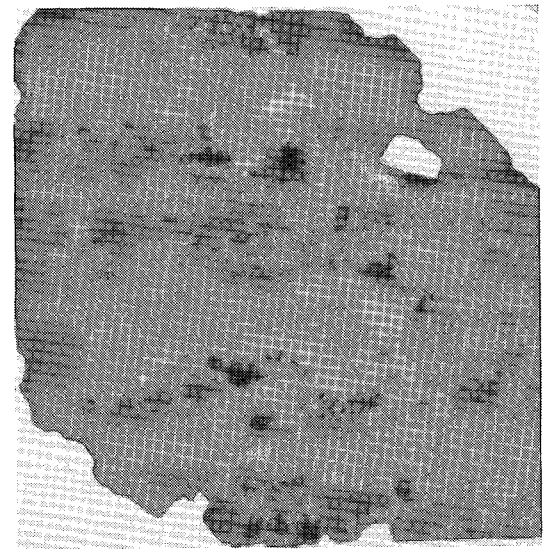

FIG. 5. Autoradiograph of sample XL1731-1.8. The image shown is of the lower section L of sample XL1731-1.8A (Fig. 3d). Brightest areas correspond to cavities lined with crystalline barite. Some cavities are filled with glue and appear dark in the image.

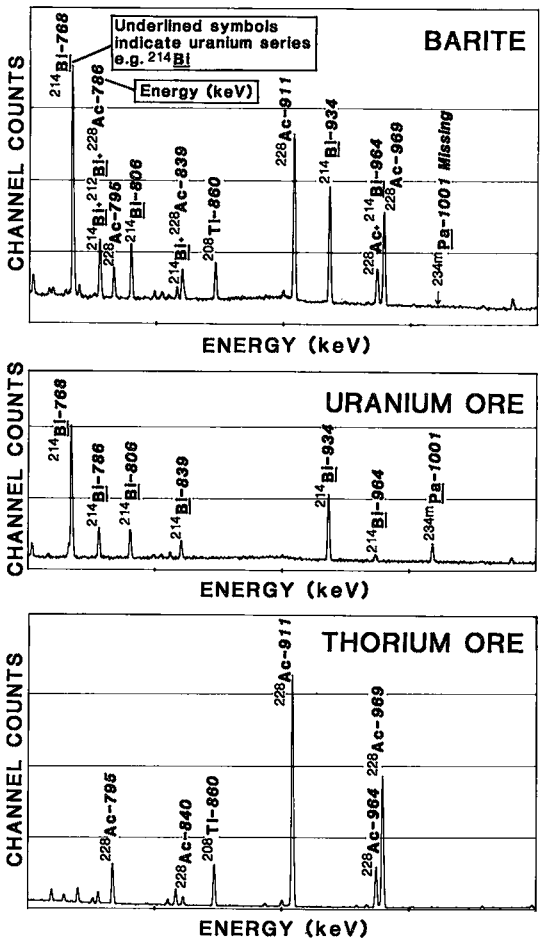

FIG. 6. Gamma-ray spectra of typical barite-rich material, and uranium and thorium reference ores.

Comparison of the thorium and barite spectra over the entire energy range indicates that all the gamma-ray peaks in the thorium spectrum are also observed in the barite (Fig. 6). Most of these gamma rays originate from ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl which were used to measure the (^{228}Ra)/(^{228}Th) ratio (Table 1). This (^{228}Ra)/(^{228}Th) ratio will reach an equilibrium value of 1.50 in about 20 years (Lalou & Brichet 1987). The measured ratio of about 1.1 (Table 1) confirms the young age of the barite.

The activities of ^{226}Ra, ^{228}Ra and ^{228}Th (Table 3) are two orders of magnitude higher than those previously reported for sulfide chimneys (Finkel *et al.* 1980, Lalou & Brichet 1982, Kadko *et al.* 1985). The ^{226}Ra values, however, are similar to those reported by Cecile *et al.* (1984) for continental barite produced from hot-spring precipitates. Radioactive barite has also been found in association with uranium deposits, the radioactivity being due to adsorption of radium on the crystal surfaces (Komarov 1974). The high activity of the barite chimneys and their young age makes them ideally suitable for chronological studies using the ^{228}Th/^{228}Ra method.

TABLE 1. (^{228}Th)/(^{228}Ra) RATIOS FOR WHOLE ROCK BARITE SAMPLE (XL-1731-1.8A)

228Th Daughter		228Ra Daughter		
Isotope	Energy (keV)	Isotope	Energy (keV)	(228Th)/(228Ra) Ratio
212Pb	239	228Ac	338	1.14 ±0.04
208Tl	583	228Ac	338	1.16 ± 0.04
212Bi	727	228Ac	795	1.05 ± 0.11
208Tl	860	228Ac	911	1.22 ± 0.10

Errors are at the 95% confidence level.

TABLE 2. (^{226}Ra)/(^{228}Ra) RATIOS FOR WHOLE ROCK BARITE SAMPLE (XL-1731-1.8A)

226 Ra Daughter		228 Ra Daughter		
Isotope	Energy (keV)	Isotope	Energy (kev)	(226 Ra)/(228 Ra) Ratio
214 Pb	295	228 Ac	338	6.9±0.5
214 Pb	352	228 Ac	338	7.2±0.3
214 Bi	1120	228 Ac	911	7.1±0.3

Errors are at the 95% confidence level.

TABLE 3. ACTIVITIES OF ISOTOPES IN THE URANIUM AND THORIUM SERIES (Bq/g) FOR TWO SMALL BARITE SAMPLES

Sample	(238U)1	(226Ra)	(232Th)1	(228Ra)	(228Th)
XL-1731-6.7A1	0.010	25.4±0.8	<0.004	5.61±0.07	2.84±0.03
XL-1731-1.8B	0.011	30.8±1.0	-----	6.56±0.13	4.74±0.07

Errors are at the 95% confidence level.
1 From neutron activation analyses.
----- Not analyzed.

TABLE 4. (^{228}Ra)/(^{228}Th) AND (^{226}Ra)/(^{228}Ra) RATIOS, ESTIMATED AGES AND FLUID RESIDENCE TIMES FOR BARITE SAMPLES

Sample	(228 Th)/ (228 Ra)	(228 Th)/ (228 Ra) Age (Years)	(226 Ra)/ (228 Ra)	Initial1 (226 Ra)/ (228 Ra)	Fluid Residence Time (Years)2
XL-1731-1.8A (Whole rock)	1.149±0.018	6.02±0.22	7.11±0.07	3.44±0.10	19.4±0.7
XL-1731-1.8B	0.723±0.018	2.73±0.09	4.69±0.15	3.38±0.11	18.9±0.9
XL-1731-6.7A1	0.506±0.008	1.70±0.04	4.53±0.14	3.69±0.12	21.3±0.9

Errors are at the 95% confidence level.
1 Calculated at the time of formation of the barite using the 228 Ra/228 Th age. Errors include uncertainties in the age and the measured (226Ra)/(228Ra) ratio.
2 Based on an estimated (226 Ra)/(228Ra) ratio of 1.33 for basalts from the Juan de Fuca Ridge.

^{228}Th/^{228}Ra AGE CALCULATIONS

To calculate the age of the barite samples the half-lives of ^{228}Ra and ^{228}Th were taken as 5.75 ±0.03 and 1.9132 ±0.0009 years, respectively (after Kocher 1981). The ^{228}Th/^{228}Ra method assumes that no

thorium is incorporated into the rock when it is formed. Consequently, any ^{228}Th must come from the decay of ^{228}Ra. The neutron-activation analyses of sample XL1731-6.7A1 show that the parent ^{232}Th has low activity (Table 3) and will make negligible contribution to the decay rate of its daughter ^{228}Ra. In addition, no gamma rays were detected from ^{234}Th in the uranium series.

Assuming no ^{228}Th is present initially, the variation of (^{228}Th)/(^{228}Ra) with time is given as:

$$(^{228}\text{Th})/(^{228}\text{Ra}) = \left(\frac{\lambda_T-\lambda_R}{\lambda_T}\right)\left[1-\exp(\lambda_R-\lambda_T)t\right] \quad (1)$$

where λ_T and λ_R are the decay constants for ^{228}Th and ^{228}Ra, respectively (Lalou & Brichet 1987). The ^{228}Th to ^{228}Ra activity ratio will therefore reach an equilibrium value of 1.50, given by $(\lambda_T-\lambda_R)/\lambda_T$, after ~20 years.

Table 4 shows the (^{228}Th)/(^{228}Ra) and (^{226}Ra)/(^{228}Ra) activity ratios for the two small samples and the whole rock. The activity ratios for the whole rock are the weighted averages of the values in Tables 1 and 2. The results show that the samples have not reached their (^{228}Th)/(^{228}Ra) equilibrium value of 1.50, thus allowing their ages to be calculated using equation (1).

The two small samples XL1731-6.7A1 and XL1731-1.8 came from two separate chimneys and give ages of 1.7 and 2.7 years. As these samples were chimney fragments, they probably do not represent the time the chimneys started forming. However, by comparing the ages of whole-rock sample XL1731-1.8A and the small sample XL1731-1.8B from the same chimney, some conclusions can be drawn.

The calculated age of 6.0 years for the whole rock XL1731-1.8A will be a weighted average of the ages of the parts of the sample that contribute to the observed count rates. Therefore the chimney must have started growing at least 6 years prior to the measurements. The age of the small sample XL1731-1.8B ajacent to the whole rock is 2.7 years. Other neighboring pieces may well be younger than this age of 2.7 years. We can therefore conclude that this part of the chimney took at least 3 years (6 – 2.7 years) to grow to the maximum dimension of the whole-rock section of 18 cm. This gives a maximum possible growth rate of 6 cm per year, which is not inconsistent with the value of 1.2 cm per year calculated for a similar chimney structure, also from the Juan de Fuca Ridge (Kadko *et al.* 1985).

Fluid Residence Times

Because ^{228}Ra decays exponentially with time, and ^{226}Ra can be assumed to have remained constant in the short life of the chimney, the initial (^{226}Ra)/(^{228}Ra) ratio can be calculated. The results (Table 4) give similar initial ratios for all three samples and should be compared to the value for Mid-Ocean Ridge Basalt (MORB).

Radioactivity measurements for MORBs from the Pacific and Atlantic oceans have revealed that fresh basalts are almost always out of radioactive equilibrium (Condomines *et al.* 1981, Newman *et al.* 1983). The ^{230}Th activity is significantly higher than the ^{238}U activity and is considered to be due to partial melting of mantle material (Allègre & Condomines 1982, McKenzie 1985). Equilibrium between ^{230}Th and ^{238}U will eventually occur after a time which is dependent on the half-life of ^{230}Th (7.5×10^4 years). Because of radioactive disequilibrium the (^{226}Ra)/(^{228}Ra) ratios of recent MORBs must be determined from their (^{230}Th)/(^{232}Th) ratio. If this (^{230}Th)/(^{232}Th) ratio is determined on rocks older than about ten thousand years, an age correction should be applied as carried out by Condomines *et al.* (1981). The problem of seawater contamination is an additional complication that frequently occurs. ^{230}Th production from ^{238}U in seawater has been found to give apparently high (^{230}Th)/(^{232}Th) ratios for oceanic basalts. Considerable care must therefore be taken in selecting and preparing samples for analysis. Fresh, unaltered basalt glasses are recognized as being the best (Newman *et al.* 1983).

Unfortunately, very few radioactivity measurements have been reported for fresh basalts from the Juan de Fuca region. Kadko *et al.* (1985) have reported ^{230}Th and ^{232}Th values for four basalts dredged from different sites along the ridge. Their data illustrate the problem of carrying out such measurements. Two of the samples show significant ^{230}Th contamination, evident from high (^{230}Th)/(^{238}U) ratios in excess of 2.2. There are also differences between their measurements of (^{232}Th) by alpha spectrometry and isotope-dilution mass spectrometry because of the low concentrations and associated activities of the samples. However, the (^{238}U)/(^{232}Th) ratios of all four samples calculated from their ^{238}U/^{232}Th mass-spectrometry values give an average of 1.13 ±0.06. This average is comparable to the value of 1.25 ±0.14 for seven basalt glasses from the southern Juan de Fuca Ridge, also calculated from U and Th concentrations (Hegner & Tatsumoto 1987). However, to determine the (^{230}Th)/(^{232}Th) ratios from the (^{238}U)/(^{232}Th) values some correction must be applied for disequilibrium in the uranium series. This correction factor is estimated to be 1.25 (Newman *et al.* 1983, Condomines *et al.* 1981), which gives a (^{230}Th)/(^{232}Th) ratio of 1.41 ±0.08 for the four dredged basalts, and 1.56 ±0.18 for the seven basalt glasses.

An alternative method of estimating initial (^{230}Th)/(^{232}Th) ratios makes use of ^{87}Sr/^{86}Sr ratios. Condomines *et al.* (1981) found a good correlation

between these two ratios for seven volcanic areas covering a wide range of values. Hegner & Tatsumoto (1987) gave $^{87}Sr/^{86}Sr$ ratios of around 0.7025 for the seven basalt glasses and three of the dredged basalts analyzed by Kadko *et al.* (1985). Sun & Nesbitt (1979) also gave a range of 0.7023 - 0.7027 for MORBs from the Juan de Fuca Ridge. Based on the $^{87}Sr/^{86}Sr$ correlation with $(^{230}Th)/(^{232}Th)$, the initial $(^{230}Th)/(^{232}Th)$ ratio would be in the range 1.32 - 1.34. This ratio is close to the values of 1.41 $\pm$0.08 and 1.56 $\pm$0.18 previously calculated from U/Th ratios. We have therefore taken 1.33 as being the initial $(^{230}Th)/(^{232}Th)$ ratio of fresh basalts from the Juan de Fuca Ridge. This value, which would also be the expected initial $(^{226}Ra)/(^{228}Ra)$ ratio, is significantly different from our measured initial ratios of ~3.5 for the barite chimney fragments (Table 4.)

Turekian & Cochran (1986) have found differences between the initial radium isotope ratios of clams and the MORB value. Kim & McMurtry (1988) have also determined an initial radium ratio of ~2.7, for a sulfide chimney on the Juan de Fuca Ridge, which is similar to our measured value. To explain these differences Turekian & Cochran (1986) proposed a simple model from which the time taken for chemical breakdown of the basalts could be calculated. In their model the following assumptions are made:

1) Prior to reaching the high-temperature area within the oceanic crust where the basalts are suffering chemical breakdown, the seawater contains negligible radium. This assumption can be justified from studies of the uranium decay products in groundwater. Radium, which enters groundwater by recoil, is quickly absorbed onto rock surfaces (Krishnaswami *et al.* 1982).

2) The dominant process by which radium enters the hot fluids is by chemical destruction of the basalts and not by recoil. Turekian & Cochran (1986) have discussed these two processes and concluded that recoil can be neglected because the Ra/Ba ratios of hydrothermal fluids are similar to MORBs.

3) Negligible ^{232}Th is incorporated into the hydrothermal fluids during chemical breakdown of the basalt. ^{228}Ra is therefore unsupported by its parent ^{232}Th.

4) The time taken for the fluids to travel from the region of chemical reaction to the seafloor is much shorter than the 5.75 year half-life of ^{228}Ra.

5) The quantity of radium removed from the basalts and entering the hydrothermal fluids is a linear function of the time the hot fluids are in contact with basalt.

This model accounts for the lack of correspondence betwen the $(^{226}Ra)/(^{228}Ra)$ ratios of the fluids and the basalts. As the chemical reaction time increases, the ^{226}Ra content of the fluids will increase. Over a time-span which is much less than the half-life of ^{226}Ra (1600 years), negligible ^{226}Ra will decay. However, because of the much shorter half-life of ^{228}Ra (5.75 years), with time, progressively more ^{228}Ra will decay in the fluids. Consequently, the $(^{226}Ra)/(^{228}Ra)$ ratio of the fluid will increase with time from its initial MORB value. This particular model is analogous to neutron activation in which a particular nuclide is created at a constant rate but also decays exponentially with time. Equations for the variation with time of a nuclide produced at a constant rate are given in standard nuclear physics textbooks (*e.g.*, Kaplan 1962). The equation for the variation in the $(^{226}Ra)/(^{228}Ra)$ ratio with time is given by Turekian & Cochran (1986):

$$(^{226}Ra)/(^{228}Ra) = (^{230}Th)/(^{232}Th)\frac{\lambda_8}{\lambda_6}\frac{\left[1-\exp(-\lambda_6 t)\right]}{\left[1-\exp(-\lambda_8 t)\right]} \quad (2)$$

where $(^{230}Th)/(^{232}Th)$ is the initial MORB value (1.33), and λ_8 and λ_6 are the decay constants of ^{228}Ra and ^{226}Ra, respectively.

Table 4 lists the fluid residence times for the whole rock and the two small samples calculated using equation(2). All three samples give similar residence times of about 22 years. The calculated residence time is not critically dependent on the $(^{226}Ra)/(^{228}Ra)$ MORB value because the ratio in the fluids changes fairly rapidly with time due to the short half-life of ^{228}Ra. For instance, increasing the initial $(^{226}Ra)/(^{228}Ra)$ basalt ratio from 1.33 to the value of 1.43 used by Turekian & Cochran (1986) only decreases the calculated residence time by one year.

Given that the three samples are from the same spire, it is likely that the radium-bearing solutions had a common source at depth, and had spent the same length of time acquiring their radium signatures. The fact that the samples have almost identical residence times in spite of their different ages and activities demonstrates the applicability of the method to barite-rich material. Turekian & Cochran (1986) concluded that for the Galapagos Mussel Bed area, the hydrothermal fluids had spent between 22 and 45 years acquiring their chemical signature. A residence time of ~15 years can be calculated from the initial $(^{226}Ra)/(^{228}Ra)$ ratio of a sulfide chimney from Juan de Fuca Ridge (Kim & McMurtry 1988). More work, particularly on barite, could confirm whether 20 years is a typical residence time for fluids producing such hydrothermal deposits.

CONCLUSIONS

Based on gamma-ray studies of samples from two barite-rich chimneys from Axial Seamount, Juan de

Fuca Ridge, we have come to the following conclusions.

1) The barite-rich chimneys are unusually radioactive. Two samples from different chimneys gave gamma-ray-equivalent concentrations of 0.1% Th and 0.2% U.

2) The radium isotopes in the U- and Th-decay series are unsupported by their parents ^{238}U and ^{232}Th, which together with their high activity and young age makes them ideal for chronological studies using the $^{228}Th/^{228}Ra$ method.

3) A whole-rock section through the chimney gave a minimum $^{228}Th/^{228}Ra$ age of 6 years. By analyzing an adjacent chimney fragment with an age of 2.7 years, we established a maximum growth rate of 6 cm per year.

4) Based on their $^{228}Th/^{228}Ra$ ages, we calculated initial $(^{226}Ra)/(^{228}Ra)$ ratios of ~ 3.5 for all three samples. This result is significantly different from an estimated value of 1.3 for basalts from the Juan de Fuca Ridge. This difference can be explained by a simple model in which both radium isotopes enter the hydrothermal fluids at a constant rate by chemical reaction of seawater with basalt. Because of the different half-lives of the two radium isotopes, the $(^{226}Ra)/(^{228}Ra)$ ratio of the fluids will increase with time from the initial MORB value.

5) Based on this model, we have calculated that the length of time the fluids were acquiring their radium signature is ~ 20 years for each of three samples. Similar residence times were found for samples from the Galapagos Rise spreading center (Turekian & Cochran 1986) and for a sulfide chimney from the Juan de Fuca Ridge using data presented by Kim & McMurtry (1988). More studies of barite from different areas would be useful to determine if 20 years is a typical residence time for the fluids which precipitate these deposits.

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