Radionuclide Disequilibria Studies for Investigating the Integrity of Potential Nuclear Waste Disposal Sites: Subseabed Studies

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September 1981

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INTRODUCTION

The isolation of high-level nuclear wastes requires a high degree of integrity for the proposed isolation facility. Geologic history largely indicates the long-term stability of the formations being considered as potential high-level waste sites. For example, if it can be shown that specific elements have been leached in the past or during recent times, then such a site would not be a likely candidate for nuclear waste storage. However, if little or no movement of specific elements can be demonstrated for periods up to a million years or more, one may conclude that a site could be suitable for long-term isolation.

Ideally one wishes to know the migration characteristics of specific waste-related, long-lived radionuclides at a potential nuclear waste burial site. Since many of the radionuclides are transuranics and their direct migration is indeterminate, no direct measurement method is possible. No satisfactory methods except ^{14}C dating, which has its practical limitations, now exist for determining directly the slow migration of specific elements within a geologic site over a very long time or in the intervals between episodic migration events. Hence we have turned to studying naturally occurring radionuclides in geologic material as analogs to the long-lived radionuclides in high-level wastes. Although in this report the radionuclides disequilibria concept was applied to subseabed migration, it may be equally valid for other potential waste storage sites such as salt domes, basalt and granite.

The dating methods used in this preliminary study are based on ratio measurements of the natural uranium and thorium decay chain members. All the pertinent natural radionuclides in these chains are shown in Table 1. In addition, certain long-lived spontaneous fission products could serve as useful analogs to certain waste radionuclides, and these are included in Table 1 for completeness. If the natural radionuclides are used as analogs of radionuclides in nuclear wastes, then their wide range of chemical properties (valence states, solubilities, etc.) and half-lives will provide information and insight for these two important questions regarding the integrity of the potential disposal sites:

- the importance of past solution transport of radionuclides in the geological environment of proposal disposal sites,
- the time scale of any past steady-state migration based on disequilibria of the radioisotope pairs.

CONCEPT

The approach is based on the concept of parent-to-daughter relationships shown in Table 1. If they are in equilibrium, then their activity ratio is unity. A departure from unity denotes disequilibrium on the time scale, depending on the half-lives of daughter and parent involved. The important ratios for consideration are $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{234}\text{U}$, $^{226}\text{Ra}/^{230}\text{Th}$, $^{231}\text{Pa}/^{230}\text{Th}$, $^{231}\text{Pa}/^{235}\text{U}$, $^{230}\text{Th}/^{232}\text{Th}$, $^{228}\text{Ra}/^{232}\text{Th}$ and ^{238}U /spontaneous fission products (^{129}I , ^{99}Tc , ^{135}Cs), respectively. These ratios cover the time span of 10^3 to 10^8 years.

TABLE 1. Radionuclide Relationships in Migration and Age-Dating Studies

^{238}U CHAIN

^{235}U CHAIN

^{232}Th CHAIN

^{238}U SPONTANEOUS FISSION PRODUCTS

The ratios are useful because the various radionuclides have different half-lives and/or solubilities in aqueous media. As an example, the ratio of $^{234}\text{U}/^{238}\text{U}$ is of special interest because ^{238}U is primordial whereas ^{234}U is a decay product. The radionuclide ^{234}U is known to dislodge by a recoil process and may become more mobile relative to ^{238}U (Koczy 1961; Burton 1975). Thus a $^{234}\text{U}/^{238}\text{U}$ ratio of unity in a material implies that no migration of ^{234}U has occurred relative to ^{238}U within the time scale comparable to the 250,000 half-life of ^{234}U . Similar examples can be drawn from the elements in Table 1. Since these radionuclides possess a range of half-lives from a few years to millions of years, migration over such time periods can be calculated from radioisotope ratios.

To illustrate the concept, let's assume that a geologic material was vigorously leached some time in the past and that the concentrations of ^{235}U and ^{238}U were reduced eightfold and tenfold relative to their daughters ^{231}Pa and ^{230}Th ; ^{234}U , because of its increased mobility, was reduced 100-fold relative to ^{230}Th . Likewise, ^{129}I was reduced by 200-fold, and ^{226}Ra , because of its high solubility was completely removed. The return of these radionuclides to equilibrium is shown schematically in Figure 1. Immediately following leaching, the radionuclides ^{230}Th , ^{231}Pa , and ^{135}Cs are in large excess relative to their parents ^{238}U and ^{235}U ; the radionuclides ^{234}U , ^{129}I , and ^{226}Ra are severely depleted relative to their parents.

The ingrowth of ^{226}Ra reaches equilibrium with ^{230}Th in a few thousand years. The decay of the excess radionuclides spans a period of 10^4 to 10^7 years, and the ingrowth of the depleted radionuclide a period from 10^4 to 10^8 years. Thus the relative ratios of ^{235}U , ^{238}U and their decay products can be used to determine time scales of disequilibrium processes occurring from 10^3 to 10^8 years ago.

In this study we have demonstrated the feasibility of using members of the natural radionuclide decay series to determine sedimentation rates and radionuclide migration within deep sea cores in an area that has been selected as a generic study site for possible high-level waste disposal. While the data in this report address this specific study, it should be noted that the

FIGURE 1. Schematic Showing How Various Radionuclides Approach Equilibrium from a Given Disequilibrium Condition

techniques are applicable to other types of candidate disposal sites (salt dome, basalt and granite) currently being considered for permanent storage of high-level nuclear waste.

APPROACH

The naturally occurring radionuclides are usually present at very low levels; thus to use them for dating and migration calculations sensitive analytical techniques must be available for their measurement. At Pacific Northwest Laboratory (PNL) very sophisticated and sensitive counting techniques and other analytical methods have been developed to measure members of the ^{235}U , ^{238}U , and ^{232}Th chains in geologic materials by nondestructive methods. However, if the concentrations of uranium and thorium and their daughters are low, then the radionuclides must be radiochemically separated and concentrated to achieve the required measurement sensitivity. For measuring the spontaneous fission products ^{99}Tc , ^{129}I , and ^{135}Cs in sediment, the current detection sensitivities are 10^8 , 10^7 , and 10^6 atoms, respectively. These sensitivities are in the range of those required for geochronological and migration rate studies. Thus, with existing techniques and improved analytical methods, geochemical dating and radioisotopes ratio measurements can provide a firm basis for helping to establish the integrity of potential high-level waste sites.

If sufficiently precise radionuclide measurements can be made, then migration models can be employed to answer important questions regarding the integrity of candidate disposal sites. Examples of the information that can be obtained are as follows:

1. Establish any past movement of natural radionuclides or spontaneous fission products over time scales ranging from 1000 years to tens of millions of years.
2. Characterize adsorption/desorption rates of the various natural radionuclides between natural waters and the associated strata and thus establish actual in situ distribution coefficients.

3. Establish the rate of chemical migration of natural radionuclides that are analogs to radionuclides in potential disposal sites.

This information is both useful and actually necessary to the selection of candidate sites for the isolation of high-level radioactive wastes.

To demonstrate the analytical approach and techniques, we obtained samples of subseabed sediment cores taken in the North Pacific gyre from a location chosen as one of the generic study areas. These were acquired through D. R. Anderson, Sandia National Laboratories program manager for the subseabed disposal program, and included samples from a 25-m core, and a 2.5-m core both taken near 30°N , 158°W . These cores had been extensively studied by the subseabed program scientific team. We measured both radionuclides and stable elements in the core samples to gather information on the current sedimentation rate, radionuclide migration rates, and general profiles of selected stable elements. From these data we were able to establish diffusion rates within the sea sediment.

SAMPLES

About 30 samples were obtained from a 25-m long core, LL-4 GPC-3 (30° , 19.9°N , 157° , 48.4°W). As the upper 0.5 m of the long core was not available, we also obtained 10 samples from a 2.5-m short core, MARA 02 GC-4, taken from a nearby area (30° , 6.3°N , 157° , 57.1°W). To investigate the near-surface sediment processes, the radionuclides ^{238}U , ^{235}U , ^{234}U , ^{230}Th , ^{232}Th , ^{231}Pa , and ^{226}Ra were measured on selected samples from this series by direct counting and radiochemical separation techniques. A suite of about 30 stable elements was determined on all samples. These samples are "red" clays though their appearances change from tan-medium to dark-medium to a light-brown color with increasing depth of the core. Figure 2 depicts the physical appearances and location of the core samples that we analyzed and also indicates the types of radionuclide or chemical analyses performed.

FIGURE 2. Samples Chosen for Radionuclide and Chemical Studies Shown in Both the Short GC-4 and Long Cores GPC-3

PROCEDURE

The measurement of many of the radionuclides in geologic materials requires carrier-free radiochemical separations, which are labor-intensive. Approximately 5 g of material was fused in a Na_2CO_3 matrix. The fused cake was decomposed with water and neutralized with acid. Uranium and thorium were coprecipitated with $\text{Fe}(\text{OH})_3$ scavenging using NH_4OH . Subsequently, uranium and thorium were further radiochemically purified and separated using modifications of the ion-exchange and electroplating procedures (Grindler 1962; Hyde 1960). These radiochemical separations were repeated at least twice to ensure radiochemical purity. After the separations, the radionuclides ^{238}U , ^{235}U , and ^{234}U were measured by both alpha spectrometry and mass spectrometry. The precision of the measurements was ± 1 to 2% by mass spectrometry and ± 3 to 8% by alpha spectrometry. The mass spectrometry results, which were more precise, are presented in this report. The radionuclides ^{230}Th , ^{232}Th , and ^{231}Pa were determined by alpha spectrometry. The precision of the data by this method is ± 1 to 3%. The chemical yields of uranium and thorium were determined by using ^{233}U and ^{228}Th tracers. Radium-226 was measured by direct gamma counting of its low-energy photons using an intrinsic germanium detector. The errors due to counting statistics were small, ranging from ± 1 to 3%. Chemical analyses of some 30 stable elements were performed by instrumental neutron activation analysis (Laul 1979). The overall error in the analysis of radionuclide and chemical data displayed in Figures 3 to 7 is included in the solid and open circle symbols.

DISCUSSION OF RESULTS

THORIUM-230/THORIUM-232 PROFILE

Figure 3 shows the depth profile of the activity ratio of $^{230}\text{Th}/^{232}\text{Th}$ in the short core GC-4. The ratio is high, approximately 22 at the surface, falls exponentially, and levels off to 0.7 at about 2 m below the sediment surface. This observed profile is a product of the ocean chemistry of thorium and uranium. As the ^{234}U is dissolved in seawater and decays, the daughter ^{230}Th

FIGURE 3. $^{230}\text{Th}/^{232}\text{Th}$ Depth Profile in Short Core GC-4. The sedimentation rate for both the short and long core GPC-3 is estimated to be 2.5 mm/1000 yr or 2.5 m/m·yr. The error bars are included in the solid and open symbols in Figures 3 to 7.

(7.7×10^4 yr) hydrates and precipitates in the sediment (Burton 1975). Hence, ^{230}Th collects slowly at the seawater-sediment interface, causing the high-activity ratio at the sediment surface. As the sediment continues to accumulate, the precipitated ^{230}Th decays and finally reaches some equilibrium value determined by the uranium content of the sediment. Assuming the ^{232}Th content of the accumulating sediment is constant, the ratio $^{230}\text{Th}/^{232}\text{Th}$ permits a direct calculation of the sedimentation rate. Based on the data in Figure 3 and correcting for ingrowth of ^{230}Th within the original sediment, the sedimentation rate of the short core is estimated as 2.5 mm/1000 yr. The data from the long core also yield the same value.

Protactinium-231, like thorium, is readily hydrated in seawater and is produced from a soluble uranium isotope, ^{235}U . For an independent determination the $^{231}\text{Pa}/^{232}\text{Th}$ ratios were determined, which gave an identical sedimentation rates in the short and long cores. The profile of $^{231}\text{Pa}/^{230}\text{Th}$ suggests a constant sedimentation rate, and these data showed that the sedimentation rate was constant through 2.5 m, which is about 10^6 years. However, based on ichthyolith stratigraphic study the sedimentation rate has been rather slow (0.2-0.5 m/m.y) and not constant below a 6-m depth interval, and the long core represents an accumulation history of about 60 million years (Doyle 1980).

RADIUM-226/THORIUM-230 PROFILE

Since radium is very soluble in seawater, ^{226}Ra formed from ^{230}Th decay in ocean sediment readily dissolves in interstitial water. As a result, ^{226}Ra readily diffuses into bottom waters (Koczy 1961; Burton 1975) and is constantly being supplied from the sediment. Radium-226, a daughter of ^{230}Th , has a half-life of 1600 years. With this short half-life, ^{226}Ra dating would be sensitive over a period of about 5000 years. The near-surface sedimentation rate as determined above is 2.5 mm/1000 yr, so any diffusion of ^{226}Ra into overlying seawater should be reflected in the top portion of the sediment column. Figure 4 shows the depth profile of $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio in the short core. Near the core surface ^{226}Ra is deficient by a factor of 2.2 relative to ^{230}Th but is essentially in equilibrium at greater depth. Thus ^{226}Ra appears to be diffusing out of the sediment surface. More data points are needed in the top 20 cm of sediment to characterize the profile for very precise diffusion calculations; the diffusion rate calculations which have been made are described in the section on diffusion. The ^{226}Ra profile implies a fairly rapid diffusive process, which can be explained with little reworking of the sediment surface by marine organisms.

We do not know how sediment will behave around a waste canister. A strong thermal gradient may develop which, in turn, may cause physical convection of wet sediment. The question of whether this may occur cannot be inferred from our data.

FIGURE 4. $^{226}\text{Ra}/^{230}\text{Th}$ Profile in Short Core GC-4. Smooth buildup suggests diffusive process instead of reworking of the sediment by physical processes.

URANIUM-234 PROFILE

If migration of uranium isotopes were extremely slow one might logically expect the $^{234}\text{U}/^{238}\text{U}$ ratio to be unity in the sediments well below the surface, but with diffusion the ratio may be less than 1. When ^{234}U is born from radioactivity decay it is in a more readily leachable form than ^{238}U . Seawater is enriched in ^{234}U by 15% (Burton 1975) as a result of high $^{234}\text{U}:^{238}\text{U}$ ratios in river water and preferential upward migration of ^{234}U from ocean sediments. Depth profiles of ^{234}U in both the short core (open circles) and the long core (solid circles) are shown in Figure 5. (Note that these profiles are in atom percent which is essentially equivalent to a plot of $^{234}\text{U}/^{238}\text{U}$ in these samples. At equilibrium, the ^{234}U content is 55×10^{-4} atom percent.) The measured profiles clearly demonstrate the disequilibrium between ^{234}U (2.4×10^5 yr) and its parent ^{238}U (4.5×10^9 yr).

FIGURE 5. Depth Profiles of ^{234}U (10^{-4} atom %) in Short Core GC-4 (open circles and Long Core GPC-3 (solid circles). A good fit by the dashed line in the short core is obtained by assuming $D = 1.0 \text{ cm}^2/\text{yr}$ and 22% free material migrating upward, whereas 10% free material is assumed for the long core. The core fitting is more sensitive to the amount of free material than to the D value.

At the bottom of the 25-m core in Figure 5, secular equilibrium has probably been attained. There is a progressively larger depletion of ^{234}U going up the core, indicating upward diffusion of ^{234}U . However, at about 2.5 m the profile turns suddenly and the ^{234}U content increases toward the surface. Deposition material with a $^{234}\text{U}/^{238}\text{U}$ ratio of greater than unity accumulates

at the water-sediment interface and is responsible for the sharp increase in the atom percent of ^{234}U . Thus, the observed profile of ^{234}U (Figure 5) is actually a combination of sedimentation, which tends to enhance the ^{234}U : ^{238}U ratio, and upward diffusion, which reduces their ratio. Near the surface, the first process is dominating while below the inflection the second process is dominating. How these data may be interpreted with these two processes is discussed in the next section.

CALCULATION OF MIGRATION RATES

The linear diffusion model of Ku (1965) was used in the calculation of apparent diffusion or migration rates for ^{226}Ra and ^{234}U . The model simply states that diffusion processes in sediment depend on an apparent diffusion coefficient, a sedimentation rate and the percentage of free isotope that can migrate. Under steady-state conditions, these parameters are assumed constant with depth.

For ^{226}Ra the calculation is simple because the isotope is so soluble that most of it migrates. Using the data points for the short core, we estimated the value of the diffusion coefficient (D) for ^{226}Ra to be $0.3 \text{ cm}^2/\text{yr}$.

To calculate the migration rate of a radionuclide we used the following relationship:

$$\text{Migration rate} = \frac{\text{relaxation length}}{\text{half-life}}$$

$$\text{or} \quad = \frac{\sqrt{D/\lambda}}{1/\lambda} = \sqrt{\lambda D}$$

Where D = apparent diffusion coefficient

λ = decay constant for isotope of interest

Using $D = 0.3 \text{ cm}^2/\text{yr}$ and $\lambda = 4.33 \times 10^{-4}/\text{yr}$ for ^{226}Ra , we calculate the migration rate of radium to be about 12 cm/1000 yr.

For ^{234}U all the variables in the diffusion model must be considered. For the calculation we used the sedimentation rate determined from the $^{230}\text{Th}/^{232}\text{Th}$ ratio of 2.5 mm/1000 yr and then computed a best fit to the ^{234}U data on the short and long core. In the computation the amount of free isotope and the apparent diffusion coefficient (D) were varied to obtain the best fit. The fit around the inflection point depends strongly on the amount of free isotope. The lower portion of the curve determines the calculated value of D. This variation can be seen in Figure 5, where two D values of $1.0 \text{ cm}^2/\text{yr}$ and $1.8 \text{ cm}^2/\text{yr}$ are plotted for the long core. There are not enough data on the short core to estimate a precise diffusion coefficient value. However, by assuming $D = 1.0 \text{ cm}^2/\text{yr}$ and 22% of the ^{234}U as free material migrating upward, a good fit was obtained for the short core, whereas 10% free material migrating upward was assumed for the long core. Apparently the curve-fitting is more sensitive to the amount of free material migrating upward than to D value (see Figure 5). The values of D and migrating materials are similar to those reported by Ku (1965) on other deep ocean sediments. With D of 1.0 to $1.8 \text{ cm}^2/\text{yr}$ and $\lambda = 2.89 \times 10^{-6} \text{ yr}$, the migration rate of ^{234}U in the sediment was calculated to be 1.7 to 2.3 cm/1000 yr.

To put these results in perspective, let's suppose that a canister of high-level waste was buried in the deep ocean sediment at a depth of 25 m. When the package is breached, radionuclides begin diffusion into the sediment under ambient conditions. A radionuclide with a chemistry similar to that of uranium would migrate 17 to 23 m in one million years. A radionuclide with a chemistry similar to that of radium would migrate approximately 120 m in one million years. Thus, by comparison, ^{226}Ra chemistry permits 5 to 7 times faster migration than ^{234}U chemistry. These results have major significance in assessing the relative migration rate of various radionuclides in nuclear wastes. Movement of other nuclear waste isotopes could be inferred from similar measurements of other natural radionuclides such as ^{99}Tc , ^{135}Cs and ^{129}I which are produced continuously by spontaneous fission of ^{238}U .

Thus the measurement of natural radionuclides ^{238}U , ^{235}U and ^{232}Th and their daughter products is a powerful tool for estimating the migration rate of various elements and for understanding and assessing the integrity of a potential disposal site.

CHEMICAL DATA

Chemical correlations and depth profiles of various elements were also studied. The depth profiles of La, an analog of the rare earth fission products, and Th, an analog of the transuranic elements, are shown in Figures 6 and 7. The patterns of rare earth elements and ratios of La/Yb in the long core are used as markers of the nature of material present. There appear to be basically three major chemical units present in the core. Unit 1 extends from 0 to 7 m, Unit 2 is from 7 to 20 m, and Unit 3 extends from 20 to 25 m. Among these units, depth profiles of various elements such as La, Th, Pb, As, Sr, Zn, Ni, Cu, Mn, Sc, Hf, Ba, and Rb vary widely in concentration and show sharp peakings at different depth intervals. For example, Ba shows sharp peaking at 4 m, Sc at 9 m, Hf and Th at 11 m, Ni at 13 m, Pb at 15 m, Mn at 16 m, As at 17 m, La at 18 m and Sr at 19 m. The sharp peaking of individual elements at different depth intervals indicates that these sedimentary layers are highly heterogeneous in composition. More importantly, the pronounced stratification implies that the layers do not seem to have been disturbed since their emplacement and that the major fraction of the various elements have not diffused more than a few meters in a million years.

SUMMARY

This study of subseabed sediments indicates that natural radionuclides can be employed to define past long-term migration rates and thereby evaluate the integrity of potential disposal sites in ocean sediments. The study revealed the following conclusions:

1. The sedimentation rate of both the long and short cores collected in the North Pacific is 2.5 mm/1000 yr or 2.5 m/m.yr in the upper 3 meters.
2. The sedimentation rate has been rather constant over the last one million years.
3. Slow diffusive processes dominate within the sediment. Reworking of the sediment by physical processes or organisms is not observed.

FIGURE 6. Depth Profiles of Lanthanum in the Short and Long Cores. Lanthanum shows peaking at 1.8 m in the short core and 18 m in the long core.

FIGURE 7. Depth Profiles of Thorium in the Short and Long Cores. Thorium shows depletion at 2.5 m and peaking at 11 m in the long core.

4. The migration rate of ^{234}U is 1.7 to 2.3 cm/1000 yr, whereas the migration rate for ^{226}Ra is about 12 cm/1000 yr, i.e., ^{226}Ra migrates 5 to 7 times faster than ^{234}U .
5. The cores are heterogeneous in chemical and mineral composition, and the sediment layers do not appear to have been disturbed significantly since their emplacement.

6. Since diffusive processes appear to provide the only significant method for radionuclide migration at this mid-Pacific site, it seems that locations of this type should provide considerable long-term protection for high-level waste burial. Further understanding of the behavior of waste radionuclides in the sediment could be gained by further application of these analytical techniques and by developing methods that would permit the use of long-lived radionuclides such as ^{99}Tc (2.1×10^5 yr), ^{135}Cs (2×10^6 yr), and ^{129}I (16×10^6 yr) in diffusion rate studies.

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