# XVII. RADIOCHEMICAL STUDIES ON THE PELAGIC SEDIMENTS COLLECTED IN THE EAST OF MARSHALL ISLANDS (GH78-1 AREA)

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## Introduction

The uranium and thorium series nuclides are useful means for the study of material transfer across the water-sediment boundary. The thorium isotopes, $^{232}$Th and $^{230}$Th, give an information on the sedimentation rate of deep sea sediments and they also can be used as tracers of insoluble metals in the ocean. A daughter of $^{230}$Th, $^{226}$Ra, belongs to the group of soluble alkaline-earths and thus $^{226}$Ra in sediments may give informations on the diffusivity of interstitial water and the early diagenesis of surface sediments.

We collected red clay samples and analyzed for these nuclides after separating each sample into various fractions according to a modified method of CHESTER and HUGHES (1967).

## Methods

The radio-isotopes in the sediments were divided into the following four fractions (Fig. XVII-1). At first a weak acid soluble fraction (A) was separated with acetic acid. This solvent may dissolve metals contained in water soluble salts, carbonates and the exchangeable sites of insoluble solids. Secondly an oxide fraction (B) was dissolved in the mixture of weak acid and hydroxyl amine. The third (C) was a strong acid soluble fraction, which was dissolved with hot 6 N HCl. Finally the remaining solid (D) was completely dissolved with HF.

Analytical methods for radium and thorium isotopes are shown in Fig. XVII-2, which will be given and discussed elsewhere in detail.

The samples used in this study were obtained during the cruise of Hakurei Maru, GH78-1 in the central North Pacific.

## Results and discussion

a) *Chemical states of thorium and radium*

Analytical data obtained are given in Table XVII-1 and percentage fractions are summarized in Table XVII-2.

The concentration of $^{230}$Th decreases clearly with depth, although that of $^{232}$Th is rather constant. This indicates that the sedimentation condition was nearly uniform at least during the past a few hundred thousand years.

We could not observe $^{226}$Ra in the weak acid soluble fraction. This may be due to the formation of an insoluble salt of radium with sulfate in interstitial

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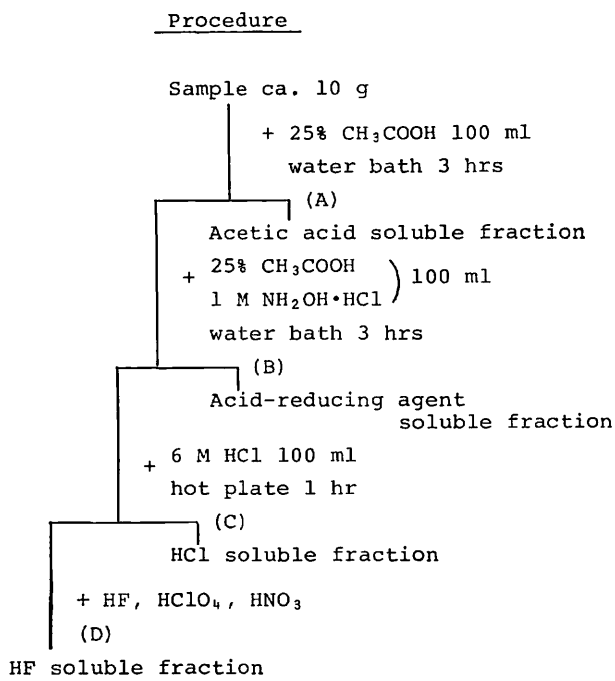


Fig. XVII-1 Stepwise separation method of radio-nuclides in the sediments according to their chemical states.

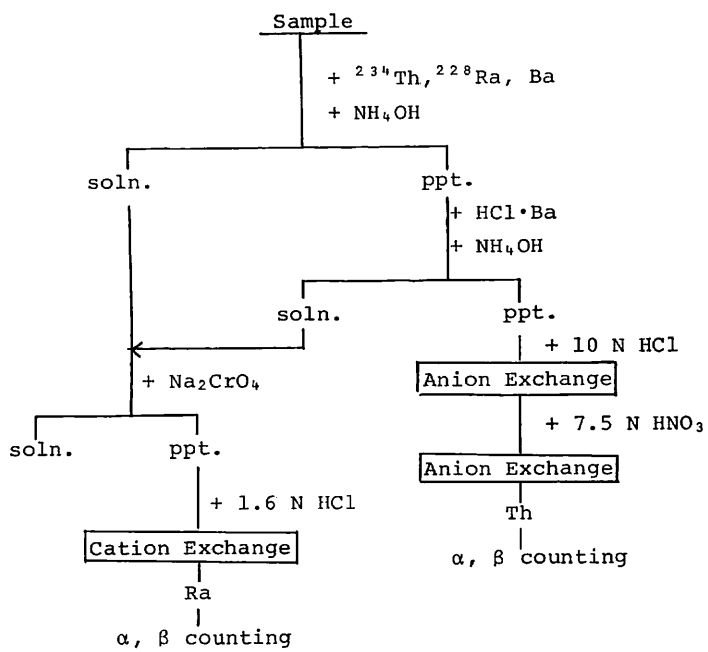


Fig. XVII-2 Schematic analytical procedure for the determinations of thorium and radium.

Table XVII-1 Concentrations of radio-nuclides separated into sequential fractions. (A) weak acid soluble fraction, (B) reducing agent soluble fraction, (C) strong acid soluble fraction and (D) refractory fraction, in the sediments

| Station | Depth (cm) | Fraction | $^{232}$Th (dpm/g) | $^{230}$Th (dpm/g) | $^{226}$Ra (dpm/g) | $\frac{^{230}\text{Th}}{^{232}\text{Th}}$ |
|---|---|---|---|---|---|---|
| 1045 | 0- 4 | A | 0.24±0.02 | 13.7 ±0.5 | 0 | 57.3 ±3.5 |
| 9°59.3′N | | B | 0.18±0.02 | 11.5 ±0.5 | 3.33±0.21 | 63.0 ±3.0 |
| 178°00.6′E | | C+D | 1.65±0.11 | 52.4 ±2.2 | 7.34±0.47 | 31.8 ±1.6 |
| 5380 m | 6-10 | A | 0.22±0.02 | 9.86±0.37 | 0 | 43.9 ±2.7 |
| | | B | 0.20±0.01 | 10.2 ±0.4 | 2.95±0.22 | 51.0 ±3.0 |
| | | C+D | 2.27±0.11 | 66.3 ±2.5 | 6.53±0.35 | 29.2 ±0.9 |
| | 12-16 | A | 0.17±0.01 | 8.09±0.31 | | 46.2 ±3.6 |
| | | B | 0.18±0.01 | 6.37±0.29 | 2.78±0.17 | 34.8 ±2.2 |
| | | C+D | 2.40±0.12 | 38.6 ±1.1 | 4.46±0.17 | 16.1 ±0.7 |
| | 18-22 | A | 0.19±0.01 | 6.18±0.22 | 0 | 32.1 ±1.9 |
| | | B | 0.15±0.01 | 3.93±0.17 | 2.38±0.32 | 26.3 ±1.9 |
| | | C+D | 1.80±0.12 | 27.0 ±1.2 | 6.51±0.34 | 15.0 ±0.9 |
| | 24-28 | A | 0.18±0.01 | 3.16±0.14 | 0 | 17.7 ±1.2 |
| | | B | 0.13±0.01 | 2.52±0.12 | 2.27±0.14 | 18.9 ±1.5 |
| | | C+D | 2.21±0.09 | 19.6 ±0.7 | 4.80±0.40 | 8.88±0.20 |
| 1050 | 0- 4 | A | 0.19±0.02 | 13.7 ±0.4 | 0 | 71.9 ±5.7 |
| 9°59.7′N | | B | 0.10±0.01 | 5.65±0.16 | 4.83±0.14 | 54.0 ±4.7 |
| 179°00.7′E | | C | 1.69±0.10 | 88.9 ±2.4 | 5.09±0.17 | 52.6 ±2.4 |
| 5603 m | | D | 0.62±0.06 | 2.11±0.13 | 1.37±0.14 | 3.38±0.33 |
| | 6-10 | A | 0.23±0.02 | 11.7 ±0.4 | 0 | 50.6 ±4.4 |
| | | B | 0.17±0.01 | 9.09±0.28 | 7.02±0.31 | 52.3 ±4.3 |
| | | C | (6.38±0.34)? | 69.4 ±3.6 | 5.08±0.20 | (10.9 ±0.4)? |
| | | D | 0.62±0.03 | 2.76±0.09 | 1.14±0.12 | 4.4 ±0.18 |
| | 12-16 | A | 0.17±0.02 | 7.51±0.24 | 0 | 45.1 ±5.4 |
| | | B | 0.19±0.01 | 7.81±0.23 | 3.97±0.20 | 40.6 ±2.7 |
| | | C | 1.56±0.12 | 62.4 ±3.0 | 6.19±0.32 | 39.9 ±2.5 |
| | | D | 0.57±0.04 | 1.87±0.10 | 1.14±0.14 | 3.30±0.25 |
| | 18-22 | A | 0.18±0.02 | 6.39±0.25 | 0 | 34.9 ±4.2 |
| | | B | 0.22±0.03 | 7.04±0.26 | 3.29±0.13 | 31.6 ±3.5 |
| | | C | (3.14±0.24)? | 35.5 ±2.0 | 3.93±0.15 | (11.3 ±0.7)? |
| | | D | 0.56±0.03 | 1.82±0.08 | 1.65±0.14 | 3.25±0.19 |
| | 24-26 | A | 0.13±0.01 | 2.70±0.08 | 0 | 20.7 ±1.7 |
| | | B | 0.26±0.02 | 5.45±0.19 | 6.58±0.25 | 20.9 ±1.9 |
| | | C | 1.53±0.08 | 29.9 ±1.0 | 5.84±0.28 | 19.5 ±0.9 |
| | | D | 0.69±0.03 | 1.28±0.06 | 0.57±0.04 | 1.86±0.14 |
| 1056 | 0- 4 | A | 0.25±0.02 | 15.9 ±0.6 | 0 | 63.4 ±3.2 |
| 10°58.8′N | | B | 0.23±0.01 | 11.7 ±0.4 | 5.53±0.23 | 51.0 ±2.8 |
| 179°58.6′N | | C+D | 2.01±0.08 | 67.9 ±2.3 | 2.23±0.20 | 33.8 ±0.7 |
| 5930m | 6-10 | A | (2.17±0.11)? | 11.7 ±0.5 | 0 | (5.37±0.17)? |
| | | B | 0.14±0.01 | 7.07±0.26 | 6.54±0.43 | 51.4 ±4.2 |
| | | C+D | 1.70±0.08 | 61.5 ±2.1 | 3.96±0.33 | 36.2 ±1.0 |
| | 12-16 | A | 0.25±0.02 | 10.4 ±0.4 | 0 | 41.6 ±2.1 |
| | | B | 0.12±0.01 | 5.02±0.17 | 3.00±0.32 | 43.0 ±3.0 |
| | | C+D | 1.44±0.07 | 39.4 ±1.4 | 1.44±0.07 | 27.4 ±0.8 |
| | 18-22 | A | 0.16±0.02 | 6.02±0.26 | 0 | 38.0 ±5.0 |
| | | B | 0.18±0.02 | 6.11±0.23 | 9.39±0.66 | 34.1 ±3.3 |
| | | C+D | 0.75±0.04 | 11.3 ±0.4 | 7.29±0.57 | 15.1 ±0.7 |
| | 24-28 | A | 0.10±0.01 | 2.27±0.10 | 0 | 22.5 ±3.2 |
| | | B | 0.16±0.02 | 5.60±0.23 | 14.5 ±0.6 | 35.5 ±4.8 |
| | | C+D | 0.96±0.07 | 11.4 ±0.5 | 0.88±0.10 | 11.9 ±0.7 |

Table XVII-2 Percentage fraction of data given in Table XVII-1

| Nuclide | Station | A | B | C | (C+D) | D |
|---|---|---|---|---|---|---|
| $^{232}$Th | 1045 | 8.4 | 7.0 | | (84.6) | |
| | 1050 | 6.4 | 7.1 | 62.1 | | 24.4 |
| | 1056 | 11.7 | 11.4 | | (77.0) | |
| $^{230}$Th | 1045 | 14.7 | 11.8 | | (73.4) | |
| | 1050 | 11.0 | 10.5 | 75.9 | | 2.8 |
| | 1056 | 17.6 | 17.1 | | (65.3) | |
| $^{226}$Ra | 1045 | 0 | 31.7 | | (68.3) | |
| | 1050 | 0 | 43.7 | 45.5 | | 10.8 |
| | 1056 | 0 | 70.4 | | (29.6) | |

water, because we also could not find barium in the fraction (TSUNOGAI *et al.*, 1979).

Table XVII-2 shows that $^{232}$Th is more contained in the refractory fraction than $^{230}$Th. This is considered that radiogenic $^{230}$Th makes authigenic minerals in seawater or recoilled $^{230}$Th is loosely bounded. It is interesting to note that $^{226}$Ra, one of alkaline-earth metals, is not so soluble as compared with $^{230}$Th or $^{232}$Th which rather easily make an insoluble solid phases in seawater, but we must remind that only a small fraction of $^{226}$Ra remains in the sediments.

b) *Sedimentation rate of red clay*

The large excess of $^{230}$Th relative to its parent $^{234}$U was observed for the surface sediments. This excess $^{230}$Th should decrease with a half-life of $7.5 \times 10^4$ years. Therefore we can evaluate the sedimentation rate of the sediments using the vertical profile of $^{230}$Th with the aid of the following equation and the least square method.

$$A = A_0 \exp(-\lambda t)$$
$$= A_0 \exp(-\lambda z/S)$$

where A is the radioactivity of excess $^{230}$Th, $\lambda$ is the decay constant, z is the depth and S is the sedimentation rate. In this equation the sedimentation rate and the porosity of sediments are assumed to be constant, and thus we get $z = St$.

$$\ln A = \ln A_0 - \lambda z/S$$

A plot of logarithms of $^{230}$Th content versus depth gives a straight line and the sedimentation rate from its gradient.

The sedimentation rate is obtained to be 1 to 2 mm/$10^3$ yr as shown in Table XVII-3. This value seems to be probable in this region.

c) *Sedimentation rate of $^{230}$Th and the flux of $^{226}$Ra from the sediments to the bottom water*

In Table XVII-3 is included the sedimentation rate of $^{230}$Th (dpm/cm$^2$/yr) at the surface, which is calculated from the sedimentation of the surface sediments, its in situ density and the concentration of $^{230}$Th. The values range from $5 \times 10^{-3}$ to $10 \times 10^{-3}$ dpm/cm$^2$/yr, and agree fairly well with the production rate of $^{230}$Th in the water column of 4,000 m depth ($8 \times 10^{-3}$ to $9 \times 10^{-3}$ dpm/cm$^2$/yr, KRISHNASWAMI, 1976). This coincides with the fact that thorium in

Table XVII-3 Sedimentation rates of sediments and $^{230}$Th, and flux of $^{226}$Ra from the sediments to water calculated from $^{230}$Th and $^{226}$Ra in the sediments

| Station | Sdimentation rate mm/10$^3$yr | Sedimentation rate of $^{230}$Th dpm/cm$^2$/yr (atoms/cm$^2$/yr) | Flux of $^{226}$Ra to water dpm/cm$^2$/yr (atoms/cm$^2$/yr) |
|---|---|---|---|
| 1045 | 1.7 | $5.9\times10^{-3}$ | $2.6\times10^{-1}$ |
| | | ($3.4\times10^{8}$) | ($3.2\times10^{8}$) |
| 1050 | 2.0 | $9.7\times10^{-3}$ | $4.1\times10^{-1}$ |
| | | ($5.6\times10^{8}$) | ($5.0\times10^{8}$) |
| 1056 | 1.2 | $5.8\times10^{-3}$ | $2.5\times10^{-1}$ |
| | | ($3.3\times10^{8}$) | ($3.1\times10^{8}$) |

seawater is rapidly removed from seawater and there is a negligible source for $^{230}$Th in seawater other than those produced from radioactive decay of $^{234}$U in seawater.

In the surface sediments we also have observed the deficiency of $^{226}$Ra activity relative to the $^{230}$Th activity. This deficiency should be caused by the diffusive flux of $^{226}$Ra in interstitial water and the flux is easily calculated as shown in Table XVII-3.

About 90% of $^{226}$Ra produced in the sediments returns to the bottom water. This flux is about one order of magnitude larger than that observed in the western North Pacific ((3.3 to 5.1) $\times$ $10^{-2}$ dpm/cm$^2$/yr, YAMADA, 1979). where the sedimentation rate is also larger, and that observed in the coastal area, New York Bight ($5\times10^{-2}$ dpm/cm$^2$/yr, LI *et al.*, 1979).

## References


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