COO-3462-15

# FISSION- AND ALPHA-TRACK STUDY OF BIOGEOCHEMISTRY OF PLUTONIUM AND URANIUM IN CARBONATES OF BIKINI AND ENEWETAK ATOLLS

Final Report

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## DISCLAIMER

INTRODUCTION

In the last three years, coral samples from the Bikini and Enewetak lagoons were analyzed for their alpha emitter concentration using solid state alpha track detectors.

For several years prior to the last three year contract, we had applied solid state track detectors (SSTD) to determine the distribution and concentration of uranium in carbonates by detecting the fission events caused by the thermal neutron induced fission of uranium 235.

During the final year of this contract we decided that it would be useful to determine the U concentration and distribution in coral samples from the Bikini and Enewetak lagoons. It was decided that these corals, which were alive during the period of the nuclear tests in these sites and some samples of which were shown previously to be contaminated with alpha emitters (Noshkin et al, 1975; Levy et al, 1978; COO-3462-14), would allow a better understanding of the effect of a radioactively contaminated environment on the assimilation of U by corals. Through the combined use of solid state alpha track detectors (SSAD) and solid state fission track detectors (SSTD) it is possible to relate the concentration and distribution of plutonium and americium (by SSAD) and uranium (by SSTD) to the coral textures in the same sample.

The discrimination between Pu and Am on the one hand and uranium on the other is not possible in areas of the sample where high plutonium (> 1000 pCi/g Pu) concentrations are found. In a fission

track experiment the number of tracks produced by Pu fission are negligible, provided the Pu concentration in the area of the sample analyzed does not exceed 1000 pCi/g. Accordingly, both contaminated and uncontaminated corals were exposed to SSAD, and, only portions of these corals showing no "hot spots," i.e. no high Pu or Am concentrations, were analyzed for uranium by fission. Nonetheless these samples represent corals which grew in an environment contaminated by nuclear testings, and in one group of them high concentrations of alpha emitters were detected by SSAD.

Eight coral samples were chosen from the Bikini and Enewetak lagoon for U analysis.

## COOPERATION WITH OTHER INVESTIGATORS

Samples were obtained through the courtesy of Dr. V.E. Noshkin from Lawrence Livermore Laboratories and Dr. S. Smith and Mr. B.P. Lamberson, Mid-Pacific Marine Laboratories.

## PRESENTATION OF SCIENTIFIC CONTRIBUTIONS

During this year and a half three papers were published, the first in Health Physics (Levy, et al, 1978) and the second in Geo Journal (Levy, et al, 1977) and the third paper in the Third International Coral Reef Symposium (Levy, et al, 1977).

## ANALYTICAL METHODS

The method of analysis of alpha emitters by solid state alpha emitters is described in one paper (Levy, et al, 1978) and in several different reports (Levy, et al, 1975, 1976, 1977). The method of

uranium measurement by fission track analysis has become an established method (Fleisher, et al, 1975). Using both methods allows the measurement of uranium concentrations and distribution in corals, provided the Pu + Am content does not exceed 1000 pCi/g. Calculations show that:

1) 18 ppb (1000 pCi/g) of Pu will, in a fission experiment, give the same number of fission tracks as 3 ppm of natural uranium, namely 160 tracks/reticle (1.2 $mm^2$) when using a thermal neutron flux of 1 x $10^6$ neutrons/$cm^2$.

2) 18 ppb of Pu will, in an alpha experiment, give 20 alpha tracks/reticle, while 3 ppm U give only 0.12 alpha tracks/reticle. Thus, if in an alpha experiment, 20 alpha tracks/reticle are detected in a portion of a certain sample, they can be attributed either to 18 ppb Pu or 2500 ppm natural uranium but it is not possible to determine by using alpha detectors whether the 20 alpha tracks/reticle originate from Pu or U; on the other hand a fission experiment can help differentiate between the Pu and U.

If the 20 alpha tracks/reticle were due to 18 ppb Pu, then in the fission experiment this concentration of Pu would result in about 160 fission tracks/reticle. If, on the other hand, the 20 alpha tracks/reticle detected by SSAD were due to 2500 ppm U, then the number of fission tracks expected (flux 1 x $10^{16}$ neutrons/$cm^2$) should be about 135,000 fission tracks/reticle, which would form a very dense fission track star, in fact not measurable using the microscope at 625 x magnification.

Thus, it is possible by the use of alpha and fission track

analysis to attribute an alpha track concentration either to Pu or to U. Using a similar approach it can be shown that at levels of natural uranium concentrations (about 3 ppm) the number of fission tracks due to U is significantly larger than the number due to any low level Pu concentration. Only concentrations of about 1000 pCi/g Pu will give a comparable number of fission tracks. Accordingly in analyzing for U it can be concluded that a sample containing Pu does not interfere seriously in the fission experiment unless its concentration exceeds 1000 pCi/g (about 18 ppb Pu). In most portions of corals from Bikini and Enewetak lagoon the expected Pu concentration is less than 10 pCi/g. With a typical thermal neutron irradiation this concentration yields only about two fission tracks/reticle or the ratio of number of fission tracks due to U to the number due to Pu (10 pCi/g) is around 100. Thus fission tracks due to 10 pCi/g Pu can be neglected in U analysis by fission.

By analyzing one of the contaminated coral samples, the coral head Porites lutea ($E_7$), the general relations shown by the calculations were confirmed. Exposing a thin section $E_7$ of the coral first to a SSAD for 30 days a "hot spot" in the form of 111 alpha tracks/reticle (1.2 $mm^2$) was detected. Then by applying the SSTD technique to the same thin section a "star" of fission tracks was detected. Approximately 60 fission tracks/reticle were counted which formed this star. Thus it was shown that the hot spot detected by SSAD was due to Pu rather than U, because the concentration of U necessary for the emission of 111 alpha T/R (during a

30-day exposure) is 20,000 ppm and in a fission experiment such a uranium concentration would have yielded about one million fission tracks.

SAMPLES

The coral samples analyzed from the Bikini and Enewetak lagoons are described in Table 1. Figures 1 and 2 are maps of Bikini and Enewetak Atolls showing sample locations.

Two groups of coral heads were chosen for uranium analysis by the fission track method. The basis for coral sample segregation was the degree of contamination with Pu and Am.

Group A (contaminated samples)

Previous work with the coral samples listed in Table 2 showed that they included portions highly contaminated with Pu and Am. The results presented in the 1978 Annual Report (Levy, et al, 1977, COO-3462-14) are summarized in this table.

Group B (non-contaminated samples)

The second group of samples analyzed included samples in which no areas of high concentrations of alpha emitters were detected by SSAD in previous work; these samples are $E_3$, $E_4$, $E_5$, $E_6$ and $E_8$ described in Table 1.

RESULTS

Results of uranium analysis of the corals are shown in Table 3.

Group A (Corals contaminated with Pu and Am)

1) Analysis of outer portion of the coral Favites virens

TABLE 1

Coral Heads Analyzed, Bikini and Enewetak Lagoons

| Number | Coral Species | Location[2] Letter | Supplier[3] |
|---|---|---|---|
| $B_1$ | Favites virens | | VEN |
| $E_2$ | Goniastria retiformis | R | VEN |
| $E_3$ | Porites lutea | E* | PML |
| $E_4$ | Porites lutea | A* | PML |
| $E_5$ | Favia pallida | A* | PML |
| $E_6$ | Favia pallida | A* | PML |
| $E_7$ | Porites lutea | B* | PML |
| $E_8$ | Favia pallida | B* | PML |

[1]B - Bikini lagoon; E-Enewetak lagoon

[2]Locations of samples are shown in Figures 1 and 2.

[3]VEN - Dr. Victor E. Noshkin; PML - Dr. P.M. Lamberson

*close to the Mike and KOA craters.

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TABLE 2

Coral Samples from Bikini and Enewetak Lagoons,
Containing High Concentrations of Pu and Am - (Group A)

| Sample No. | Species | Location | Maximum Plutonium Concentration pCi |
|---|---|---|---|
| $B_1$ | Favites virens | Bikini Lagoon | > 2000 |
| $E_2$ | Goniastria retiformis | Enewetak " | 480 |
| $E_7$ | Porites lutea | " " | 6000 |

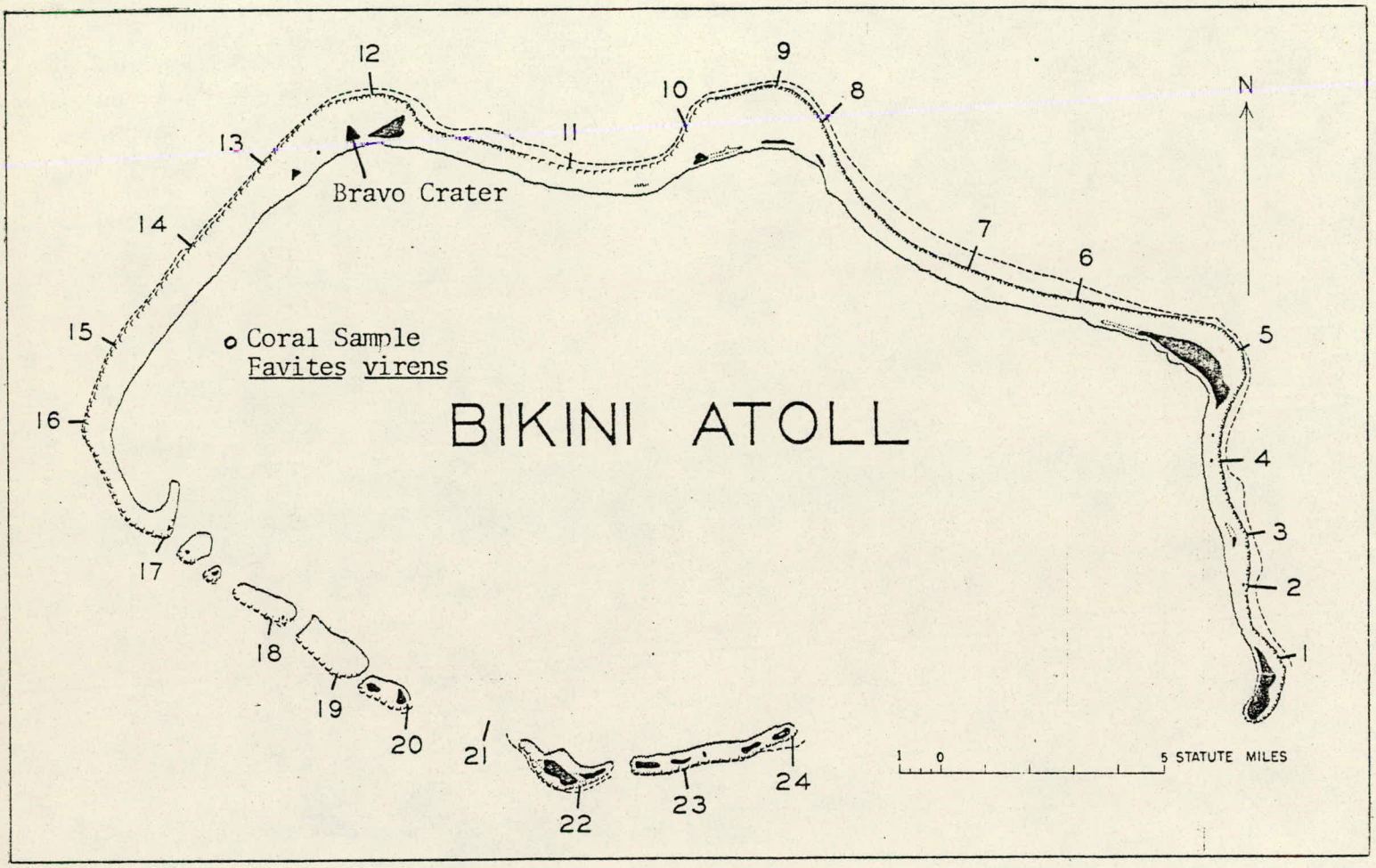


Figure 1 Map of Bikini Atoll and sample location

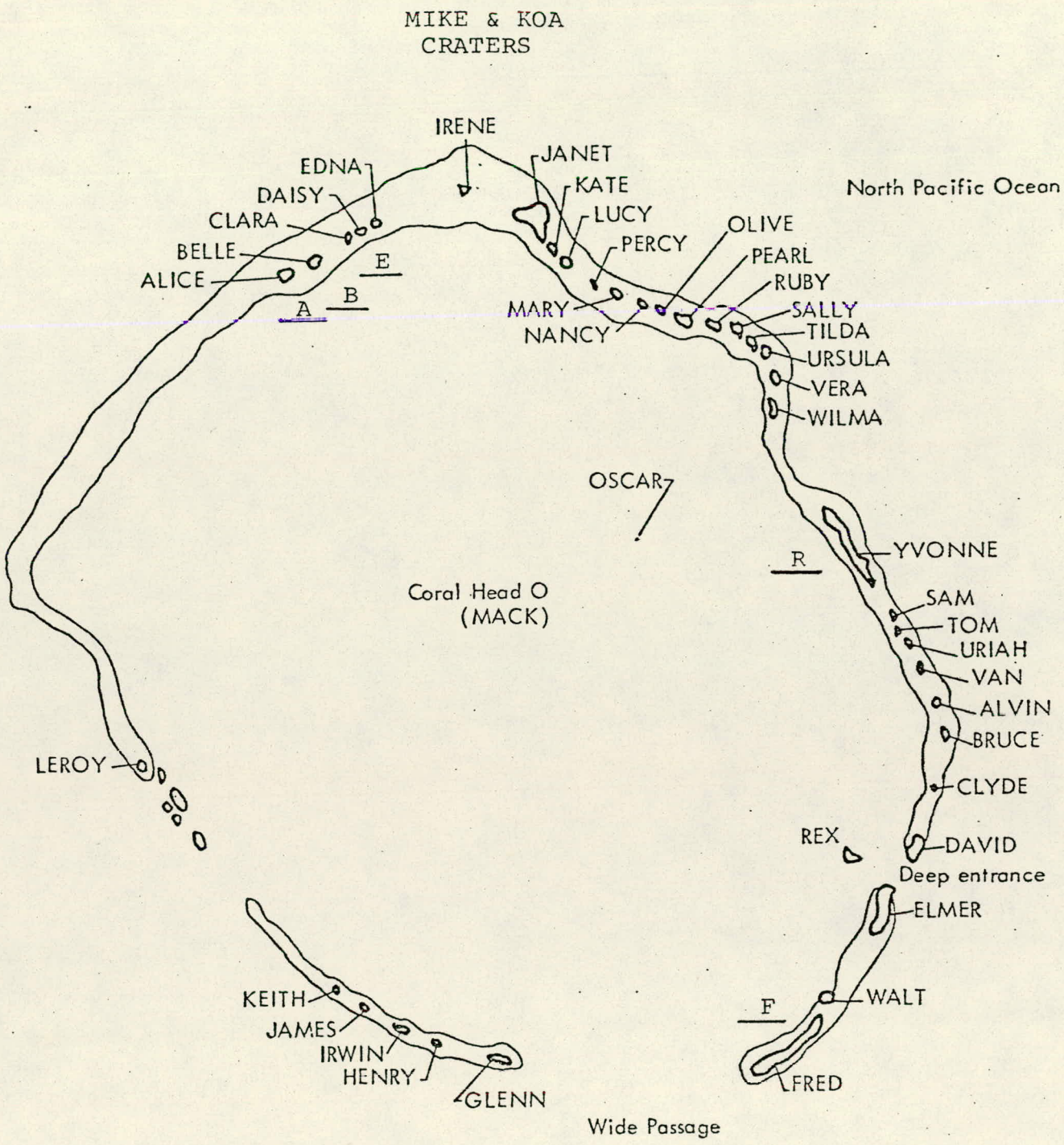


Figure 2 Map of Enewetak Atoll and sample locations

($B_1$) [thin section A (COO-3462-14) Bikini Lagoon (see Figure 3)]. The top 1/4 of thin section A (Fig. 3) of the coral Favites virens represents the period of growth of the coral which followed the nuclear blasts. (See report, COO-3462-14, pp. 18, 19).

a) Alpha track analysis

Thin section A (upper portion) was exposed to a SSAD and confirmed that this portion of the coral Favites virens does not contain high concentrations of alpha emitters.

b) Fission track analysis.

The fission track analysis of the portion of the coral Favites virens (known to contain no high concentrations of Pu or Am) showed that the uranium concentration ranges from 1.2 to 2.2 ppm with an average of about 1.8 ppm. These concentrations were found in the coral texture senso stricto.

2) Analysis of the coral Goniastrea retiformis $E_2$ - Enewetak Lagoon.

a) Alpha Track Analysis

As shown in the summary report (COO-3462-14) this sample contained several hot spots with up to 480 pCi/g. Alpha track analysis of the portion of the thin section used in this report showed one star of alpha tracks equivalent to a concentration of 2700 pCi/g. Pu concentrations as shown by SSAD do not exceed 10 pCi/g in all other portions of the samples.

Portion analyzed by fission and alpha detectors (this study).

Areas showing high alpha emitters concentrations (Pu and Am) (COO-3462-14).

Figure 3. Portion of thin section a, coral Favites virens, analyzed by fission and alpha track detectors' methods.

b) Fission Track Analysis

In fission track analysis, the area containing the star of alpha particles was excluded from the counting. In areas containing < 10 pCi/g, Pu does not affect the analysis of U by fission as discussed earlier in this report. Uranium concentrations were found to range from 1.3 to 1.7 with an average of about 1.6 ppm.

3) Analysis of the coral Porites lutea $E_7$ - Enewetak Lagoon.

a) Alpha Track Analysis

A study of the thin section showed a hot spot in the form of a star of alpha tracks equivalent to a concentration of 6000 pCi/g Pu. In all other areas of the thin section Pu concentrations did not exceed 18 pCi/g.

b) Fission Track Analysis

Analysis of the detector excluding the hot spot area showed concentrations ranging from 1.6 to 2.0 ppm with an average of about 1.9 ppm u.

Group B (Corals free of contamination with Pu and Am)

Analysis of these corals ($E_3$, $E_4$, $E_5$, $E_6$, $E_8$) by SSAD showed that they contain no hot spots; furthermore the maximum Pu concentration in them does not exceed 4 pCi/g. Accordingly the number of fission tracks found by SSTD can be attributed to U only.

The results of the fission track analysis of these samples are shown in Table 3.

TABLE 3

Plutonium and Uranium Concentrations in Coral Heads from Bikini and Enewetak Lagoons

| Sample No. | Coral Species | Max. Pu Concentration pCi/g | Uranium Concentration (ppm) Min. | Max. | Mean |
|---|---|---|---|---|---|
| Group A (contains area with hot spot > 400 pCi/g Pu) | | | | | |
| $B_1$ | Favites virens | 2 | 1.5 | 2.3 | 1.8 |
| $E_2$ | Goniastria retiformis | 2700 | 1.8 | 2.3 | 1.9 |
| $E_7$ | Porites lutea | 6600 | 1.7 | 2.1 | 1.9 |
| Group B (no areas with more than 10 pCi/g Pu) | | | | | |
| $E_3$ | Porites lutea | 3 | - | - | 1.9 |
| $E_5$ | Favia pallida | 2 | 1.5 | 2.3 | 1.8 |
| $E_6$ | Favia pallida | 2 | 2.9 | 3.1 | 3.0 |
| $E_4$ | Porites lutea | 2 | 2.6 | 2.9 | 2.7 |
| $E_8$ | Favia pallida | 2 | 2.9 | 3.1 | 3.0 |

## DISCUSSION AND CONCLUSIONS

Both groups of samples, A and B, were taken from a radioactively contaminated environment; only Group A samples were shown to contain high Pu and Am concentrations while in samples of Group B no "hot spots" were detected. Thus, samples of Group A represent corals which accumulated high concentrations of alpha emitters and thus in them it is expected to find the most accentuated effects of radioactive contamination on the assimilation and or accumulation of U in corals.

In Group B, on the other hand, no "hot spots" were detected and one would expect less of a change in both U concentrations and distribution compared to what was found in corals from normal marine environments e.g. the Red Sea, where no nuclear tests were performed (Gvirtzman, et al, 1973).

The results of the analysis of uranium concentrations in the 8 coral heads sampled from the Bikini and Enewetak lagoons lead to the following conclusions: 1) No parallel increase in uranium concentration was found in the corals contaminated by Pu and Am; 2) In the non contaminated corals, the fission track analysis shows wider ranges of uranium concentrations (1.8 - 3.1). Thus, in the corals not contaminated by Pu and Am, uranium concentrations similar to the uranium concentration in the contaminated corals were found; 3) Uranium content in all corals analyzed was rather homogeneously distributed, no "hot spots", "stars" or areas differing in concentration by more than a few percent were detected by the fission track analyses.

It should be noted that the portions of the contaminated samples analyzed for U were chosen to be clear of high Pu and Am concentration and thus do not represent the distribution and concentration of uranium in the same portion of the sample affected by the nuclear blast.

A U analysis by SSTD of portions of coral samples containing high Pu and Am concentrations is not possible due to the fact that Pu and Am as well as U fission under the effect of thermal neutrons. Thus a discrimination between Pu and Am (if these occur in concentrations exceeding 1000 pCi/g as in the coral samples contaminated by the nuclear blasts $B_1$, $E_2$ and $E_7$) and U is not possible by the use of the SSTD techniques.

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