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RADIOLOGICAL RESURVEY OF ANIMALS, SOILS AND GROUNDWATER AT BIKINI ATOLL, 1969

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Abstract

The results of radiometric and radiochemical analyses of samples, exclusive of land plants, collected at Bikini Atoll in 1969 are presented and discussed. Average values for radionuclides in food items in pCi/g wet are: reef fish, ^{60}Co-2.6, ^{90}Sr-.08, ^{137}Cs-.13; pelagic fish, ^{60}Co-.94; spiny lobster, ^{60}Co-.12; giant clams, ^{60}Co-24; curlews, ^{60}Co-.94, ^{137}Cs-380; turnstones, ^{60}Co-7.7, ^{137}Cs-56; terns, ^{60}Co-1.1, ^{137}Cs-.08. Average concentrations of ^{90}Sr in the muscle of coconut crabs from Bikini and Enyu Islands were 12 pCi/g wet and .05 pCi/g wet, respectively. There are no striking differences between the 1967 and 1969 average values for edible foods of marine origin, including the sea birds. Predominant radionuclides in undisturbed soils in 1969 are ^{55}Fe, ^{60}Co, ^{65}Zn, ^{90}Sr, ^{125}Sb, ^{137}Cs and ^{207}Bi. In the crater sediments ^{55}Fe, ^{60}Co, ^{90}Sr, and ^{207}Bi predominate. There are quantitative and qualitative differences in radionuclide content associated with the feeding habit of fish and there appears to be an increasing concentration of some radionuclides with increasing age of fish and clams. The radionuclide content of bird species presents a sharp contrast, both qualitatively and quantitatively, associated with feeding habit. It appears that some ^{60}Co and ^{207}Bi is being transported eastward by the bottom current in the lagoon. Silver-108m, previously unreported in fallout, was found in the hepatopancreas of the spiny lobster. The present levels of radionuclides and their distribution at Bikini are not likely to change significantly except for decrease in amounts, due to physical decay.

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RADIOLOGICAL RESURVEY OF ANIMALS, SOILS AND GROUNDWATER AT BIKINI ATOLL, 1969

INTRODUCTION

Bikini Atoll was a site for atmospheric tests of nuclear devices from 1946 to 1958. The population of 166 Bikinians was moved from the atoll in March, 1946, first to Rongerik Atoll, then to Kwajalein Atoll; in November, 1948, a final move was made to Kili Island. The land area at Kili is about one-tenth that at Bikini Atoll and there is no lagoon. Therefore, access to Kili is difficult, often impossible, and sea foods are scarce.

The results of a radiological resurvey of Bikini in 1964 by the University of Washington's Laboratory of Radiation Biology indicated that Bikini might be radiologically safe for permanent habitation. A request from the High Commissioner of the Trust Territories of the Pacific to the Atomic Energy Commission in 1966 to rehabilitate Bikini resulted in an extensive survey of the atoll in the spring of 1967. This survey emphasized external radiation measurements, including <u>in situ</u> gamma-ray spectrometry, although some food items were collected to supplement data from the 1964 survey. The 1967 survey party included personnel from the Atomic Energy Commission's Health and Safety Laboratory, the Division of Biology and Medicine, the U. S. Naval Radiological

Defense Laboratory, the Trust Territory, and the University of Washington.

The data were summarized by DBM and were presented to a panel of experts assembled by DBM for evaluation of potential radiological hazards. Most of the participants in the 1967 survey attended the presentation to provide details not included in the summary.

The panel concluded that Bikini could be safely reoccupied, but recommended some restrictions and suggested things to be done to rehabilitate the atoll. These included restriction of coconut crabs from the diet, because they contain high concentrations of ^{90}Sr, and covering the village area at Bikini Island with coral gravel from the beaches, to provide a shield against radiation from the soil. The panel also recommended that old structures and other such debris from the tests be removed from the islands and beaches and that the island be further monitored during the clean-up. Additional monitoring was necessary because dense vegetation on Bikini and Enyu Islets, especially, made it impractical to survey more than a few transects across the islands in 1967.

The panel's recommendations were made to the Chairman of the Atomic Energy Commission who informed the Secretary of the Interior, the administrator for the Trust Territory of the Pacific.

The clean-up phase of the rehabilitation of Bikini Atoll was begun in February, 1969, by Joint Task Force Eight. The AEC Nevada Operations Office is responsible for certification of the clean-up portion of the rehabilitation program, which was carried out under guidelines approved by the AEC Division of Operational Safety. At the request of NVOO, the U. S. Public Health Service took the responsibility for external radiation measurements, and for the collection and analysis of those land plants which are food items; the U of W Laboratory of Radiation Ecology was asked to sample and analyze other biological and environmental samples. This report presents the results of the Laboratory's analyses.

SELECTION OF SAMPLES AND SAMPLING SITES

The sampling program was based on the objective of obtaining data for evaluation of potential radiological hazards to man. The samples were limited, for the most part, to things which might be eaten by returning Bikinians, except for land plants. Some additional samples, for example soils, crater sediments and ground water, were taken to provide data for estimating the future distribution and amounts of radionuclides in the biota.

The fish collected are in two main categories: the reef fish and the pelagic, or "troll-caught" fish. The reef fishes are usually collected by throw nets by the Marshallese and are important items in their diet.

Of the more than 700 species of reef fishes at Bikini Atoll, we selected three species commonly eaten by the Marshallese and representative of three feeding habits: the mullet,* a plankton feeder; the convict surgeonfish, a grazing herbivore; and the goatfish, a bottom-feeding carnivore. The specific radionuclides found in fish and their concentrations are often associated with feeding habit, hence this was a necessary consideration in selecting samples representative of the kinds of fish which would be eaten when the Bikinians return. A fourth kind of reef fish, groupers, was also collected as respresentative of the higher order carnivores.

The troll-caught fishes are all high-order carnivores and fall into two broad subcategories: resident lagoon fish, ulua and dogtooth tuna; and migratory fish, yellowfin tuna. All were caught in or near Enyu Pass. Bikinians who were part of the clean-up crew cut filets from the yellowfin tuna and preserved them by salting. They said tuna is one of their favorite fish and, presumably, would fish for tuna if they return to Bikini.

The invertebrates sampled were the spiny lobsters (langouste), coconut crab and "giant" clams (Tridacna sp., and Hippopus hippopus). Some of the species of Tridacna never exceed a few centimeters in length, and only the smaller species were found

* For a list of common names and scientific names, see Appendix Table 16.

in the vicinity of Nam (Charlie) Islet. The larger species were found near Bikini Island.

In response to a special request to check the levels of radioactivity at Aerokoj Islet, received during the survey, the land hermit crab, a known concentrator of ^{90}Sr, was collected. Since coconut crabs are both an indicator organism and a food item, they would have been sampled instead of hermit crabs, but coconut crabs were not found on Aerokoj.

Thousands of terns nest at Bikini Atoll, mostly on the western islets. Both the birds and their eggs will be used as food. The terns almost always feed at sea, outside the lagoon or reefs. On the other hand, the curlews and turnstones feed along the shores and on the reef, and the curlew also eats the seeds of an endemic shrub, Scaevola serica, or the beach magnolia. Although the curlews and turnstones are transients and are present in small numbers, at most a few hundred, they contain the highest levels of radionuclides among the birds. Curlews, turnstones, noddy terns, and fairy terns were sampled.

Rats are not used as food but they are the only mammal living on the atoll, and a few were taken to determine their radionuclide content.

Groundwater was collected by driving half-inch pipe with well points into the soil. The well point sites on Bikini and

Eneman Islands were in areas found to be the most radioactive by the U. S. Public Health Service personnel. On Nam I. the well point was driven in a low area near the center of the island. Existing wells were sampled at Enyu. Attempts to obtain groundwater at Aerokoj were unsuccessful.

Soil samples were taken by one-inch depth increments to depths of ten inches or more near each well point. All depth increments for two sets of samples from Eneman were analyzed but only the surface one-inch of other sets of samples were analyzed. In addition to samples from soil pits at the well points, surface samples also were taken at Aomen and Oroken.

Sediments from the Bravo Crater were taken by dredge from depths of 40, 120, 140, and 160 feet.

ANALYTICAL METHODS

Gamma-Ray Spectrometry

All of the samples were analyzed by gamma-ray spectrometry. They were counted for at least 100 minutes with a 3 x 3-inch NaI(Tl) crystal used in conjunction with a 256-channel analyzer. Selected samples were counted for 1,000 minutes, either with a 3 x 3-inch detector or a detector system consisting of two opposing 5 x 5-inch crystals operating as a summing spectrometer.

Most of the biological samples were oven dried, ground and compressed in polyvinyl chloride (PVC) pipe to a volume resulting in a density of 1.0. Small samples, spiny lobster hepatopancreas for example, were ashed, dissolved in hydrochloric acid, and sealed in PVC pipe.

Oven-dried soil samples were compressed to a density of 1.35 in PVC pipe.

Spectrum resolution was done by Schonfeld's (1965) method of least squares. A set of previously prepared reference spectra for the different geometries and radionuclides was used. All values were corrected for decay to the date of collection. The error given for individual values is the 95% error.

Strontium-90 Analyses

Strontium-90 was determined by measuring the equilibrium concentration of its ^{90}Y daughter. Yttrium-90 was separated by solvent extraction and precipitation techniques (Petrow, 1965), with stable yttrium serving as both a carrier and a yield determinant. Recoveries ranged from 80 to 100%.

Iron-55 Analyses

Iron-55 was separated and purified by a combination of solvent extraction and electrodeposition techniques (Palmer and Beasley, 1967). Recoveries generally exceeded 90%. Counting was done by X-ray spectroscopy with a proportional counter in conjunction with a multichannel analyzer.

Bismuth-207 Analyses

The solvent extraction techniques of Sill and Willis (1965) were used for separating and purifying ^{207}Bi. Bismuth-212 was used as a yield determinant.

Plutonium-238, 239 Analyses

Plutonium-238,239 was separated by a combination of solvent extraction and anion exchange techniques (McCowan and Larsen, 1960; Kressin and Waterbury, 1962), with electrodeposition as the final step in the separation. Plutonium-236* was used to determine yield. A quantitative separation of plutonium from the coraline soils and sediments is exceptionally difficult and it is therefore essential that ^{236}Pu be used as a yield determinant and that counting be done by alpha spectrometry.

Tritium Analyses

Well water samples were measured for tritium content by a liquid scintillation technique with a minimum level of detection of 200 tritium units.

RESULTS AND DISCUSSION

The predominant radionuclides in the terrestrial organisms are ^{137}Cs and ^{90}Sr, whereas the marine organisms contain mainly

* Provided by the USAEC Health and Safety Laboratory, New York.

^{60}Co and ^{55}Fe. The concentrations of these radionuclides in edible portions of organisms range from undetectable amounts to the following maximum values:

^{137}Cs - 2260 pCi/g dry in the muscle tissues of a curlew from Nam I.

^{90}Sr - 204 pCi/g dry in the hepatopancreas of a coconut crab from Bikini I.

^{60}Co - 219 pCi/g dry in muscle and mantle tissue of a giant clam near Bikini I.

^{55}Fe - 40,900 pCi/g dry in the liver of an ulua.

The range in the amount of a radionuclide in the same tissue from the same species at the same islet is wide. When detectable amounts of radionuclides are present, the minimum and maximum values often differ by factors of four or five and sometimes by a factor of ten. The values for concentration of radionuclides in individual samples are given in Appendix Tables 1 through 15. Average values and ranges are given in text Tables 1 through 15.

Dry weights were used for the basic calculations because the true water content of some samples is difficult to determine. The average concentrations of radionuclides were converted to a wet-weight basis for convenience in calculating daily intake from the diet; the conversions were made by using average wet to dry weight ratios for each kind of sample.

The mean values for ^{90}Sr, ^{137}Cs, ^{60}Co, ^{65}Zn and ^{54}Mn in diet items at Bikini Atoll in 1967 were given in the Radiological Report on Bikini Atoll by Gustafson in 1968, and are listed in Table 1 with the average values determined from the 1968 samples. Three hundred fourteen-day ^{54}Mn and 245-day ^{65}Zn have been omitted from Table 1 because no detectable amounts of these radionuclides were found in the 1969 samples, and ^{55}Fe has been added, by using values for 1967 samples from an addendum to the 1968 report.

The 1967 values for fish include reef fish and troll-caught fish, whereas the 1969 data in Table 1 are for reef fish only. The average values for ^{60}Co in the muscle of troll-caught fish were,

Yellowfin tuna	0.15 pCi/g wet
Ulua	1.7 " "
Dogtooth tuna	0.04 " "

Thus, the 1969 values for fish in Table 1 are greater than if the values for troll-caught fish were included in the averages.

In Table 1 the data for giant clams are for 1969 samples taken from the vicinity of Bikini I. Clams were also collected around Nam I. but they were of a small species which is rarely eaten; also, the level of ^{60}Co in the Nam I. clams was lower than in the Bikini I. clams, presumably because the latter were older clams which had accumulated ^{60}Co for several years. No data for clams were available in 1967, but the maximum value for ^{60}Co in the edible portion of clams in 1964 was 73 pCi/g wet (Bonham, 1967).

The land crabs are listed separately for Bikini I. and Enyu I. because the panel convened by the DBM in 1968 recommended, on the basis of the data then available, that coconut crabs be omitted from the Bikini diet. Thirteen crabs collected at Enyu I. in 1969 were analyzed for ^{90}Sr and gamma emitters; the levels of all radionuclides are sufficiently low that a reconsideration of the restriction for Enyu I. is indicated.

The species of birds are listed separately for 1969 because an average value for all birds would be a poor estimate of the potential intake, since few curlews or turnstones are available.

In general, there are no striking differences between the 1967 and 1969 average values of radionuclides for edible portions of foods of marine origin, including the sea birds. The differences tend to show a decline in radionuclide content in 1969, but there are not sufficient data to provide a basis for a reasonable estimate of rates of decline because of the large variability in the data and the many poorly defined factors involved in the uptake and retention of radionuclides by organisms in the natural environment of Bikini. Some basic biological information such as rates of growth and life spans of the fishes is not known and the chemical form in which the radionuclides are present in the lagoon waters can only be surmised. We do not even know, for example, whether the radionuclides and their stable isotopes are present in the same chemical form. Furthermore, there are no uncontestable data on the trace element content of lagoon waters and probably will not be until the techniques of sampling and processing seawater samples is greatly improved. However, some hypotheses can be made and conclusions can be drawn from certain data.

All of the fallout radionuclides at Bikini are found in the surface of undisturbed soils. The predominant radionuclides in 1969 were ^{55}Fe, ^{60}Co, ^{65}Zn, ^{90}Sr, ^{125}Sb, ^{137}Cs, and ^{207}Bi. In the crater sediments only four predominate: ^{55}Fe, ^{60}Co, ^{90}Sr, and ^{207}Bi, although many more are present in smaller quantities. The soils and sediments are now the principal reservoirs of radionuclides at Bikini. The radionuclides are available to the land animals through the vegetation, or other animals, where there is selection of specific radionuclides, or through direct ingestion of soil. In the latter case, the animal selects certain radionuclides from a wider variety of nuclides than is in the vegetation.

Similarly, the marine animals may ingest radionuclides by eating another organism or by ingesting sediments. In addition, the marine organism may absorb radionuclides directly from the water, or radionuclides may be adsorbed on the surface of the animal. Although adsorption is an important means of contamination of organisms by fresh fallout, it is probably no longer important at Bikini, where the last significant fallout occurred in 1958. The astronomically large surface area presented by the masses of branching corals and their associated flora and fauna must have removed, from the water, all adsorbable radionuclides not already removed by the plankton soon after fallout.

The land organisms contain primarily the long-lived fission products ^{137}Cs and ^{90}Sr and, as expected, these radionuclides are found associated with those tissues or organs which contain potassium and calcium, respectively, since cesium and potassium behave similarly in metabolism, as do strontium and calcium.

There are quantitative and qualitative differences in radionuclide content of organisms associated with feeding habit. The goatfish, a bottom-feeding carnivore, contains more ^{60}Co and ^{207}Bi than the convict surgeonfish, a grazing herbivore, or the mullet, a plankton feeder (Tables 2 and 3). Higher order carnivores, the grouper and ulua, also contain more ^{60}Co and ^{207}Bi (Table 4) than the convict surgeonfish; however, the differences may be associated with age as well as with feeding habit.

The smaller, and presumably younger, reef fish of a species contain less ^{90}Sr than the larger fish of the same species

(Appendix Table 11). Presumably, the ^{90}Sr is being accumulated throughout the life of the fish and a steady state has not been reached. The values for ^{90}Sr in the ulua (Appendix Table 12) and the reef fish cannot be directly compared because the bone of the ulua was analyzed for ^{90}Sr and only whole eviscerated reef fish were analyzed. However, a comparison of Appendix Table 11 and 12 shows that there can be no great difference in ^{90}Sr content between larger, older fish of even the grazing herbivore and the higher order carnivore. On the basis of the differences between ^{60}Co content of goatfish and ulua, it might be assumed that there is an increasing concentration of the radionuclide in the ascending food chain. However, this is evidently not true for ^{90}Sr. The discrepancy probably exists because information is lacking on the radionuclide content of other organisms on which the ulua feed and which could well concentrate ^{60}Co, for example, squid.

Another example of increasing concentration of a radionuclide probably associated with age is the concentration of ^{60}Co in the kidney of the giant clams Tridacna sp. and Hippopus hippopus (Appendix Table 9). By far the highest levels of ^{60}Co, as much as 4,000 pCi/g dry, in any organism at Bikini Atoll is in the kidney of these clams. Obviously, there must be an accumulation of ^{60}Co in the kidney and the longer the clam lives in an environment where ^{60}Co is available, the more ^{60}Co it accumulates in the kidney, if ^{60}Co has a long biological half-life. This is not a concentration through the food web since the clams are filter feeders.

The radionuclide content of bird species presents a sharp contrast, both qualitatively and quantitatively, associated with feeding habit (Table 8 and Appendix Table 10). The fairy terns and noddy terns feed mostly at sea outside the lagoon and contain small amounts of fallout radionuclides, less than the amount of naturally occurring ^{40}K. They contain barely detectable amounts of ^{137}Cs. The curlew, on the other hand, feeds on the reef and on Scaevola sp. seeds, and consequently contains relatively large amounts of ^{137}Cs, as much as 2,300 pCi/g dry in muscle. The turnstones also feed along the beaches and on the reef, and contain both ^{60}Co and ^{137}Cs. The source of ^{137}Cs for the turnstones is not known, although it could be by direct ingestion of sand particles. The yellowfin tuna, which are feeding on essentially the same organisms as the terns, contain about the same levels of ^{60}Co as the fairy terns. The ^{60}Co levels in the noddy terns are somewhat higher but still are of the same order of magnitude. Thus the area in which an animal is feeding is a factor affecting its radionuclide content, as expected, in relation to the distance from the source of the radionuclide.

Fig. 1. Gamma-ray spectrum of sediment from Bravo Crater collected at a water depth of 160 feet, August, 1969.

The source of ^{60}Co for the tuna must be Bikini Atoll and not worldwide fallout because we analyzed tissues from 214 tuna, including 75 yellowfin tuna, taken from the Japanese tuna fishery during 1968 and 1969, and found no ^{60}Co (NVO-269-7, Annual Report). In contrast, the ^{55}Fe concentrations in the dark muscle of the tuna from the Japanese fishery ranged from 3.3 to 1600 pCi/g dry, most of the values fell in the range of 101 to 500 pCi/g dry. It appears, therefore, that a major amount of the ^{55}Fe in the Bikini tuna is from worldwide fallout.

One of the principal sources of radionuclides at Bikini is Bravo Crater in the reef adjacent to and southwest of Nam I. Figure 1 shows a gamma-ray spectrum of sediment taken from a depth of 160 feet. Clearly, ^{60}Co and ^{207}Bi predominate among the gamma emitters. In most soils, ^{137}Cs is the most abundant radionuclide. An intermediate condition exists at the southwestern end of Eneman I., where a low area is occasionally overwashed by seawater, and at the high tide line, where the ^{137}Cs is being leached from the soil.

The retention of ^{60}Co and ^{207}Bi by the sediments is reflected in the fact that the bottom-feeding goatfish in the vicinity of the craters contain ten times more ^{60}Co than the herbivorous convict surgeonfish and plankton feeding mullet. However, some ^{60}Co is being transported eastward by the bottom current in the lagoon either in solution or associated with fine (colloidal?) particles, because the difference in ^{60}Co content between convict surgeonfish and mullet in the vicinity of Bravo Crater and 16 miles eastward near Bikini I. is only by a factor less than two.

And, at the same time, the difference in ^{60}Co content between the goatfish from near the crater and those at Bikini I. is by a factor of about ten.

It appears that the physical redistribution of ^{207}Bi is similar to that of ^{60}Co, but since the levels of ^{207}Bi are lower than those of ^{60}Co by a factor of about 20, we are at the limits of detection, with the method used, for samples distant from the crater. The use of larger samples, chemical separation and more sensitive counting methods would make it possible to determine ^{60}Co:^{207}Bi ratios in sediments, lagoon water and organisms in different parts of the lagoon. These ratios would indicate whether transported radionuclides were primarily in solution or on particles. If the ratios remained constant, that would be a strong indication of transport on particles. The results of analyses of selected samples for ^{207}Bi by gamma-ray spectrometry and by chemical separation are compared in Table 13. Bismuth-207 will be a useful tracer in the future because it has a long half-life, 30 years compared to 5.2 years for ^{60}Co.

Plutonium-239, with a half-life in excess of 24,000 years, is another potentially useful tracer. The samples analyzed for plutonium were selected on the bases of collection location and content of gamma-emitting radionuclides, which indicate the greatest likelihood of the presence of plutonium. The values given in Table 14, therefore, probably are maximum values for each type of sample. The ratios of $^{239,240}Pu$ to ^{238}Pu approach 2:1 at Eniman I. and are about 15:1 in Bravo Crater. Bikini I. soils contained no detectable ^{238}Pu, although they contained the highest concentration of $^{239,240}Pu$ of the samples analyzed. The presence of $^{239,240}Pu$ and ^{207}Bi in goatfish viscera is consistent and probably results from direct ingestion of fine particles of sediment during feeding. The absence of ^{238}Pu in goatfish viscera as compared with the sediment merely reflects a low concentration of this radionuclide, below the limits of detection.

Although none of the 1969 samples were analyzed for the X-ray emitter ^{63}Ni, this radionuclide was found in concentrations of 80 d/m/g dry weight in Bravo Crater sediment collected in 1967 (Beasley and Held, 1969). Nickel-63 is of particular interest as a tracer since it has a half-life of 92 years. In addition, the clam kidney accumulates ^{63}Ni, as it does ^{60}Co, and is therefore an indicator organism for the presence of ^{63}Ni.

Another long-lived radionuclide, ^{108m}Ag, with a half-life of approximately 100 years, has been identified for the first time among the radionuclides at Bikini. This radionuclide was detected from the gamma-ray spectrum of the hepatopancreas of

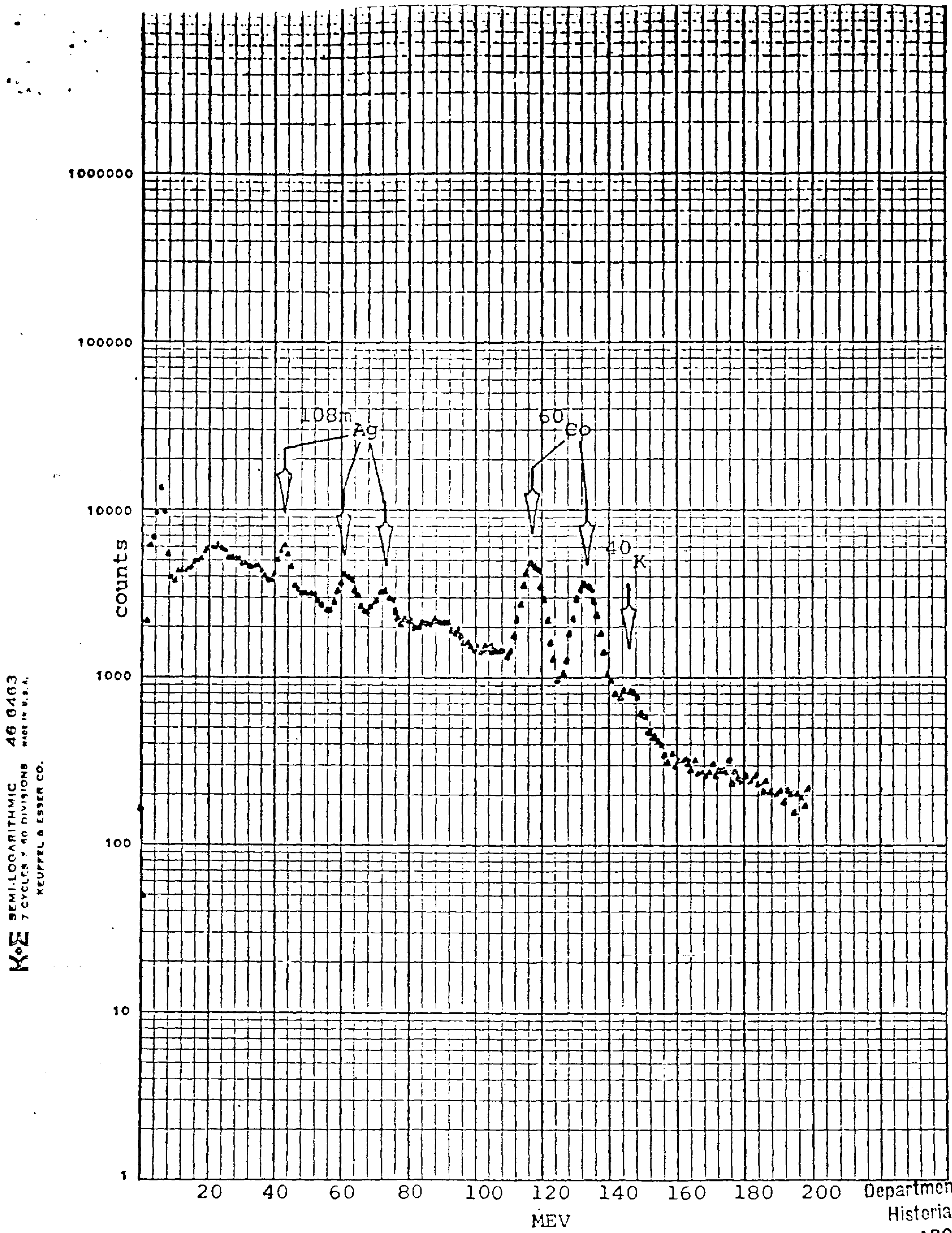

Fig. 2. Gamma-ray spectrum of spiny lobster hepatopancreas from Bikini Atoll, 1969.

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spiny lobsters collected in 1969 (Fig. 2). Although the identity of ^{108m}Ag has not been confirmed by chemical separation, there is little doubt of its presence because the spiny lobster hepato-pancreas is known to concentrate 260-day ^{110m}Ag (Seymour, 1963). Thus, ^{108m}Ag is another potentially useful long-lived tracer with its indicator organism.

Tritium in well water is present at low concentrations; the maximum value found was 14 pCi/ml, or 4300 tritium units, at Nam I., whereas at Bikini and Enyu Islands the concentration was 2 pCi/ml, or approximately 600 T.U. (Table 15). These values fall within the range of tritium concentrations in surface waters of the United States in 1966 reported by Moghissi and Porter (1968). Koranda (1965) has shown that there is approximately 10^4 times more tritium in bound water than in loose water in soils at Eniwetok Atoll, but that there is little exchange of the bound water with the loose water. Hence it is probable that there will be no major changes in the tritium concentration of well water at Bikini Atoll.

Bikini can be expected to remain a useful area for the study of the redistribution of radionuclides for at least several decades. This is especially true since rapid advances are being made in the technology of radionuclide detection.

The present levels of radionuclides and their distribution at Bikini are not likely to change significantly except for a decrease in amounts from physical decay. Exceptions are expected where physical disturbances occur during the replanting on land. If one of the rare typhoons should strike Bikini, there would be a major redistribution of the fine sediments, either a redistribution within the lagoon, a flushing from the lagoon, or both.