SILVER-108m IN BIOTA AND SEDIMENTS AT BIKINI AND ENIWETOK ATOLLS

By
T. M. BEASLEY
and
E. E. HELD

(*Reprinted from Nature, Vol. 230, No. 5294, pp. 450–451, April 16, 1971*)

(*Reprinted from Nature, Vol. 230, No. 5294, pp. 450–451, April 16, 1971*)

Silver-108m in Biota and Sediments at Bikini and Eniwetok Atolls

THE occurrence[1] of the long lived silver radionuclide ^{108m}Ag ($T_{1/2} > 100$ yr) in biota from the Pacific Ocean has been held to suggest that the silver radionuclide ratio $^{110m}Ag/^{108m}Ag$ may be useful as a tracer of environmental processes. It also appears[1] that large amounts of ^{110m}Ag and ^{108m}Ag were not produced during the 1958 test series or earlier, and that the production activity ratio $^{110m}Ag/^{108m}Ag$ was 162, a ratio derived solely from the thermal neutron activation of stable silver. We consider that their observations require reconsideration.

We have found ^{108m}Ag in biota and sediments collected at previous nuclear testing sites in the Pacific. We first detected the radionuclide in a composite sample of the hepatopancreases of spiny lobsters collected at Bikini Atoll in 1969, and have since measured its concentration in several samples from Bikini and Eniwetok Atolls. Gamma-ray spectra were made using both NaI(Tl) and a solid state Ge(Li) detector system. The resolution of the Ge(Li) detection system permitted precise identification of the radionuclide photopeaks and the NaI(Tl) crystal systems were used to quantitatively measure the ^{108m}Ag. More specific information was obtained by chemically isolating the ^{108m}Ag using a solvent extraction technique[2] by means of which the silver radionuclides are effectively separated from a number of other radio elements, especially ^{125}Sb, ^{207}Bi, ^{60}Co, ^{54}Mn, ^{102}Ru and $^{230,232}Th$.

Comparison (Table 1) of the concentrations of ^{108m}Ag in the hepatopancreas of spiny lobsters taken from Bikini with biota reported in ref. 1 shows that the Bikini specimens contain 2 to 3 times more ^{108m}Ag. By contrast, the concentration of ^{108m}Ag in the composite sample from Eniwetok is less than that in the Guadalupe lobster specimen. The last test series at Bikini and Eniwetok Atolls occurred in 1958, so that the time between the cessation of tests and the collection of the specimens ranges from 6 to 12 yr. It is reasonable to conclude that higher concentrations of both ^{108m}Ag and ^{110m}Ag were present at earlier times. Indeed, Seymour[3] has reported a ^{110m}Ag concentration of approximately 100 d.p.m./g of wet tissue for the hepatopancreas of a spiny lobster collected at Guam in November 1959, and although ^{108m}Ag was not determined in the sample, it is significant that this activity was observed in biota collected 1,200 miles downstream of the test site as early as 1959.

Table 1 Concentrations of ^{108m}Ag in Samples from Bikini and Eniwetok Atolls

Location	Samples	Date of collection	Concentration of ^{108m}Ag (d.p.m./g dry wt)
Eniwetok Atoll	Spiny lobster (20)* (hepatopancreas)	August 1964	0.20 ± 0.04†
Bikini Atoll	Spiny lobster (3) (hepatopancreas)	June 1969	1.1 ± 0.3
Bikini Atoll	Spiny lobster (9) (hepatopancreas)	June 1970	0.75 ± 0.15
Bikini Atoll	Crater sediment	July 1969	0.09 ± 0.05

* No. in parentheses signifies the number of individuals comprising the composite sample analysed. Species: *Panulirus* sp.

† Errors represent the 67% confidence level of the count rate measurements.

The sediment (Table 1) was taken from the site of a large thermonuclear detonation in 1954 (Bravo Crater), the crater of which is exposed to both lagoon and sea water and which is therefore a source of ^{108m}Ag for the biota of the area (^{110m}Ag having largely decayed between 1954 and 1969). Thermonuclear tests at other sites in the atoll have also produced silver radionuclides. The last test at Bikini was in 1958, yet we have measured a ^{110m}Ag concentration of 5.2 ± 0.06 d.p.m./g of soil on Eninman Islet at the atoll and thus the amount of ^{108m}Ag present in the Pacific Ocean and elsewhere as a result of pre-1959 testing may be significant.

The second observation about the initial production activity ratio of ^{110m}Ag/^{108m}Ag in ref. 1 may be conservative in its estimate of ^{108m}Ag production. As we have said, the initial activity ratio of ^{110m}Ag/^{108m}Ag was assumed to be due only to thermal neutron activation of stable silver. There are other production mechanisms for these two radionuclides, the most notable being (n,2n) reactions on stable silver (^{108m}Ag) and (n,p) reactions on stable cadmium (^{108m,110m}Ag). The cadmium reaction is probably not important because the quantity of stable cadmium in nuclear devices would probably be kept to a minimum on account of the large thermal neutron cross-sections. The excitation functions for (n,p) reactions at atomic weights between 100–110 and neutron energies between 10–20 MeV (ref. 4) are small (1–100 millibarns) by contrast with the excitation functions for (n,2n) reactions at atomic weights > 100 and similar neutron energies, which are significantly higher (> 1 barn).

of ^{108m}Ag in Samples from Bikini and Eniwetok Atolls

	Date of collection	Concentration of ^{108m}Ag (d.p.m./g dry wt)
20)* reas)	August 1964	0.20 ± 0.04 †
3) reas)	June 1969	1.1 ± 0.3
9) reas)	June 1970	0.75 ± 0.15
it	July 1969	0.09 ± 0.05

fies the number of individuals comprising sed. Species: *Panulirus* sp.
7% confidence level of the count rate

was taken from the site of a large in 1954 (Bravo Crater), the crater of lagoon and sea water and which is Ag for the biota of the area (^{110m}Ag between 1954 and 1969). Thermo- in the atoll have also produced silver st at Bikini was in 1958, yet we have ntration of 5.2 ± 0.06 d.p.m./g of soil atoll and thus the amount of ^{108m}Ag n and elsewhere as a result of pre-1959

about the initial production activity n ref. 1 may be conservative in its ction. As we have said, the initial ^{08m}Ag was assumed to be due only tion of stable silver. There are other or these two radionuclides, the most ons on stable silver (^{108m}Ag) and (n,p) m (^{108m,110m}Ag). The cadmium react- ant because the quantity of stable cad- ould probably be kept to a minimum ermal neutron cross-sections. The (n,p) reactions at atomic weights utron energies between 10–20 MeV illibarns) by contrast with the excita- reactions at atomic weights > 100 gies, which are significantly higher

We have irradiated stable silver in a fast neutron beam at the University of Washington cyclotron [^{9}Be(d,n)^{10}B] and have produced easily measurable quantities of ^{106m}Ag [^{107}Ag(n,2n)^{106m}Ag] and ^{105}Ag [^{107}Ag(n,3n)^{105}Ag]; ^{108m}Ag has not been found so far in the irradiated sample. The energy similarities of the gamma rays emitted by ^{106m}Ag and the very low specific activity of ^{108m}Ag will require several months decay time before the presence or absence of ^{108m}Ag can be confirmed. The (n,γ) cross-sections for ^{107}Ag and ^{109}Ag (based on a radiative capture initial activity ratio of 162) seem, however, to be almost equal for ^{110m}Ag/^{108m}Ag and so the nuclear states of the two isotopes seem to be similar. It would be surprising if ^{108m}Ag production by (n,2n) reactions on ^{109}Ag were inconsequential, for (n,2n) reactions on ^{107}Ag have been shown experimentally to produce relatively large amounts of ^{106m}Ag. Production pathways are important, because the type and design of a nuclear device can generate different ratios of fast and thermal neutron fluxes.

It is difficult to resolve our findings with the close agreement previously shown[1] between silver radionuclide activity ratios and the time of large scale nuclear testing. If it is possible for the silver radionuclides produced in 1961 and 1962 to remain in rather shallow isolated water masses for several years[1], then the same possibility must be extended to silver radionuclides produced before 1959. But the absence of definite knowledge of the rates at which silver is removed from the surface layers of the ocean makes it impossible to decide whether or not ^{108m}Ag from pre-1961–62 testing confuses subsequent dating.

The determination of ^{110m}Ag/^{108m}Ag ratios in air filters from the time period of interest would clarify the production activity ratios, and the analysis of undisturbed lichen samples collected annually since 1958 (and earlier) would elucidate the relative amounts of ^{108m}Ag caused by pre-1959 tests and those in 1961–62. The potential usefulness of the ^{110m}Ag/^{108m}Ag ratio in dating is apparent but we feel that some caution should be exercised in their use for the description of natural processes until the outstanding problems are resolved.

We thank C. W. Vick for technical assistance and Dr W. Weitkamp for help with the cyclotron irradiation. The work was supported by the US Atomic Energy Commission.

T. M. Beasley *
E. E. Held

Laboratory of Radiation Ecology,
College of Fisheries,
University of Washington

* Present address: Division of Biology and Medicine, US Atomic Energy Commission.

Received November 2, 1970.

[1] Folsom, T. R., Grismore, R., and Young, D. R., *Nature*, **227**, 941 (1970).
[2] Handley, T. H., and Dean, J. A., *Anal. Chem.*, **32**, 1878 (1960).
[3] Seymour, A. H., in *Radioecology* (edit. by Schultz, V., and Klement, jun., A. W.), 151 (Reinhold and American Institute of Biological Science, 1963).
[4] Barbier, M., in *Induced Radioactivity*, 177 (North-Holland, Amsterdam, 1969).

Printed in Great Britain by Fisher, Knight & Co., Ltd., St Albans.