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There were 67 tests associated with the U.S. nuclear testing program on Bikini and Enewetak Atolls in the northern Marshall Islands. Various quantities of sulfur (S), arsenic (As), yttrium (Y), tantalum (Ta), gold (Au), rhodium (Rh), indium (Ir), tungsten (W), and thallium (Tl) were added to test devices to determine the neutron energy spectrum and flux or for other diagnostic purposes. Other natural occurring and manmade radioisotopes including polonium-210 (^{210}Po), selected uranium and thorium isotopes, americium-241 (^{241}Am) and curium-242 (^{242}Cm) were also added to the devices to increase the explosive yield of multistage explosions or for other for experimental purposes. The original information on the inventory of materials involved in the test program was prepared by the Office of Defense Programs (DP) of the United States Department of Energy (DOE), and summarized in a letter dated 17 January of 1995, from the Assistant Secretary, Vic Reis, addressed to Dr. Paul Seligman, the Deputy Assistant Secretary for the Office of Health Studies. The question has since been raised as to the possible human health or ecological impacts of potential residual quantities of these materials remaining at the test sites. It should also be noted that with the exception of ^{241}Am and ^{242}Cm all these elements and isotopes occur in nature. The basic argument put forward in this discussion is that fallout contributions of tracer materials would need to be comparable with levels found in the natural environment before any serious consideration could be given to the possibility that residual quantities these materials at the atolls may pose a significant risk to human health.

A nuclear explosion produces a fireball of incandescent gas and vapor in excess of 10 to 100 million degrees. As the fireball dissipates it assumes a toroidal shape as strong convective forces uplift cool air and other surface debris into the cloud. The height and stabilization of the fireball depends on the explosive yield of the device, the altitude of the detonation, and meteorological conditions at the time of the blast. Condensation products formed from vapors within the fireball contain a complex mixture of fission products, activation products, unspent nuclear fuel and fuel products as well as other vaporized materials from the non-fuel bomb components and the surrounding environment. The energy released from large near-surface detonations in the Marshall Islands was sufficient to lift and immediately vaporize several thousand m^3 of soil and associated debris. Soil particles entering the fireball served as nuclei on which radioactive and other condensation products attached. These larger sized particles settled to earth more quickly and lead to significant levels of local (or close-in) fallout contamination. Some fractionation may occur between different products but for the purposes of our initial assessment of the current day impacts of tracer materials used by the U.S. in the nuclear test program in the Marshall Islands, we assume that unreacted tracer materials were partitioned in the much the same manner as volatile fission products (Robison et al., 2001). Accordingly, we approximate the deposition rates of residual tracer materials at the test sites using measured distribution patterns and inventories of cesium-137 (^{137}Cs) and strontium-90 (^{90}Sr). No additional fieldwork was carried out for this current assessment although some relevant new information did become available on uranium isotopes (Hamilton, unpublished data). We also distributed the full inventory of tracer

material by neglecting the small fraction (<10 to 20%) that would have been consumed in the device by nuclear reactions.

The total fission yield from the U.S. nuclear test program on Bikini and Enewetak Atolls in the Marshall Islands was around 38.4 and 19.9 Mt, respectively, or a total of 58.3 Mt for both atolls (Robison et al., 2001). Based on fission product production estimates from UNSCEAR (2000), 58.3 Mt of nuclear fission will produce around 345 PBq of ¹³⁷Cs and 227 PBq of ⁹⁰Sr. By comparison, the ¹³⁷Cs inventory at the atolls in 1954 was estimated to be around 1 PBq or about 0.29% of the total amount dispersed into the atmosphere. For calculation purposes this fraction was increased by 50% (to 0.42%) to account for uncertainty in the distribution of refractory materials. Based on these partitioning estimates, we then calculated the total fractional amount of each tracer element or isotope deposited over the test sites (Table 1). It was concluded that because

Table 1. Quantity of tracer material available for local distribution at the atolls (listed highest to lowest) and associated estimates of residual tracer concentrations in surface soil compared with abundances in the earth's crust

Element	Estimated amount distributed at the test sites (kg)¹	Estimated soil concentration (µg g⁻¹ dry soil)²	Crustal abundance (µg g⁻¹ dry soil)³
²³⁸ U	281	0.004	2.7
²³² Th	4.6	0.00006	0.6
Sulfur	3.1	0.00004	260
Thallium	0.65	0.000008	0.45
Tungsten	0.013	0.0000002	1.5
Indium	0.011	0.0000001	0.1
²³⁰ Th	0.0063	0.00000008	–
²³³ U	0.0046	0.00000006	–
Gold	0.0021	0.00000003	0.004
Yttrium	0.001	0.00000001	33
Rhodium	0.00084	0.00000001	0.001
Tantalum	0.00037	0.000000005	2
Arsenic	0.00032	0.000000004	1.8

¹Estimated amount of the total materials inventory distributed at the atolls based on the partitioning of ¹³⁷Cs into the local environment.

²Estimated surface soil concentration of material associated with a sample containing a present-day ¹³⁷Cs activity concentration of ~2 Bq g⁻¹ dry soil.

³Data source: Parker (1967)

of the extremely small quantity of tracer material available for distribution on the atolls, any potential residual anthropogenic contribution will pose little or no risk to human health. Moreover, there was a net loss in total inventory of the vast majority of these elements because high-energy nuclear detonations such as Mike and Bravo tests vaporized several thousand m³ of coral soil and sediment in forming large bomb craters. Naturally occurring tracer elements and isotopes in the associated coral soil and sediment

were uplifted into the atmosphere and dispersed globally with other forms of radioactive fallout debris.

A comparison of the estimated concentration of residual tracer materials at the atoll with that found in the earth's crust also shows that the anthropogenic contribution is largely negligible with the possible exception of ^{238}U (Table 1). The declared amount of ^{238}U was, by far, the largest quantity of any declared tracer material used in the test program (Robison et al., 2001). A review of historical environmental measurements indicates that there is no statistical difference in total uranium concentrations in soil and sediment within the Marshall Islands that can be directly attributable to the test program (Robison et al., 2001). However, the total uranium content of soils and sediments do not provide a definitive measure of small anthropogenic inputs of uranium because of the high natural variability in the uranium content of coral soil and sediment. In order to provide a more detailed and comprehensive assessment of potential sources of anthropogenic uranium at the test sites, we analyzed the uranium isotopic composition in representative soil samples from Enewetak Atoll using high-precision mass spectrometry. The results of this new study confirms the presence of an anthropogenic source of uranium in the environment based on observed enrichments in measured $^{235}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ atom ratios. The source of anthropogenic uranium on Enewetak can be attributed to be presence of residual amounts of nuclear fuel containing ^{235}U , and ^{236}U produced by neutron capture on ^{235}U . The ^{235}U and ^{236}U content of soils are enriched relative to ^{238}U in the natural environment by factors up to 2 fold, and between 10 to 1000 fold, respectively. ^{235}U and ^{236}U enrichments in soils are very heterogeneous but appear to be limited to selected locations on the northern islands of Enewetak Atoll. From a radiological perspective, the specific activity of uranium in all the analyzed soil samples was still overwhelmingly dominated by the natural uranium content of the bulk matrix. Consequently, there appears to be no immediate radiological or toxicological concern for human health associated with these findings.

In summary, we conclude that estimated residual concentrations of tracer elements and isotopes used in the U.S. nuclear test program in the Marshall Island pose no adverse effects on the health of resident populations currently living on the islands.

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References

Robison W.L., V.E. Noshkin, T.F. Hamilton, C.L. Conrado, and K.T. Bogen (2001). An assessment of the current day impact of various materials associated with the U.S. nuclear test program in the Marshall Islands, Lawrence Livermore National Laboratory (LLNL), UCRL-LR-143980, 21 pp.

UNSCEAR (2000). United Nations Scientific Committee on the Effects of Atomic Radiation, USCEAR 2000 Report to the General Assembly, with Annexes, Volume 1: Sources, United Nations, New York.

Parker R. (1967). Composition of the earth's crust, chapter D, In: Data of Geochemistry, sixth edition (ed. M. Fleischer), Geological Survey Professional Paper 440-D, United States Government Printing Office, Washington.