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The effective and environmental half-life of ^{137}Cs at former U.S. nuclear test sites in the Marshall Islands

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Introduction

The United States conducted atmospheric nuclear weapons testing from 1946 to 1958 at Bikini and Enewetak Atolls in the northern Marshall Islands. The nuclear weapons testing program produced widespread fallout contamination over these test sites as well as neighboring atolls to the east of Bikini. Radioactive debris deposited in soil or concentrated in lagoon sediment formed a reservoir and source-term for re-mobilization of long-lived radionuclides into the environment. Over the past three decades, researchers from the Lawrence Livermore National Laboratory (LLNL) have been evaluating radiological conditions on affected atolls and using the data to develop dose assessments for resettled and resettling populations (Robison et al., 1987, 1994, 1997a,b, 1999). These prospective dose assessments are based on monitoring data developed for external radiation, terrestrial foods, groundwater, marine foods and inhalation exposure pathways. The resultant modeled present-day and future doses are dominated by external and internal exposure to cesium-137 (^{137}Cs). Other radionuclides of potential radiological significance include strontium-90 (^{90}Sr), plutonium isotopes ($^{239+240}$Pu) and americium-241 (^{241}Am). For Bikini Island (a former residence island on Bikini Atoll), ^{137}Cs contributes as much as 85 to 90% of the estimated dose through ingestion and up to another 10% via external exposure to ^{137}Cs (^{137m}Ba) gamma radiation. Of interest, is the fact that coral soils provide conditions where ^{137}Cs is preferentially taken up from the root zone of plants and concentrated in subsistence crops such as coconuts, *Pandanus* and breadfruit, and is the most important factor in controlling the long-term environmental and radiological consequences of the nuclear test program in the Marshall Islands.

Background

The integrated 30–, 50– and 70–y predictive dose estimates for living conditions on the islands have considered only the radiological decay (λ_{decay}) of radionuclides as a loss mechanism. However, elevated concentrations of ^{137}Cs (as well as other radionuclides) have been observed in the fresh water portion of the groundwater lens on affected atolls over many years (Noshkin et al., 1977, 1981; Robison et al., 1988). This suggests that soil ^{137}Cs is being transported to the groundwater by the action of rainfall and that the inventory of ^{137}Cs available for uptake from the root zone of plants is being depleted at an accelerated compared with its loss from radiological decay (Robison et al., 2003). Additionally, labile soil ^{137}Cs can be bound up in geochemical components of the soil (e.g., clay minerals) and make cesium much more resistant to further remobilization or uptake into plants. The accelerated loss of ^{137}Cs for plant

uptake by the combined effects of these environmental processes, in addition to that occurring from radiological decay, have very important implications on projected doses over periods of 30–, 50– and 70–years, and also for providing guidance on the need for and cost benefits of remediation programs.

The Effective and Environmental half-life of ^{137}Cs

The availability of ^{137}Cs for plant uptake in surface soils of Bikini, Enewetak and Rongelap Atolls has been monitoring by time-dependent studies of ^{137}Cs concentrations in leaves of *Pisonia grandis*, *Guettarda specosia*, *Tournefortia argentea* (also called *Messerschmidia*), *Scaevola taccada*, and fruit from Pandanus and coconut trees (*Cocus nucifera L.*). Using empirical data from our annual or semi-annual monitoring programs and tracing the concentration of ^{137}Cs in individual plants over time, we have recently demonstrated that the environmental half-life of ^{137}Cs is more important that radiological decay in controlling the fate and distribution of ^{137}Cs in coral soils (Robison et al., 2003).

The time required for depleting by half the available labile ^{137}Cs from soil is defined as the environmental-loss half-life (ELH) (λ_{loss}). The loss rate, and that from radiological decay, constitute the effective ^{137}Cs loss-rate constant for a coral environment, i.e., $\lambda_{effective} = \lambda_{loss} + \lambda_{decay}$; and because plants will retain a steady-state inventory of ^{137}Cs based on its soil availability and recycling of leaf litter, the ^{137}Cs biological half-life or residence time in the plant is not considered to contribute to the total loss-rate constant. Therefore, the usefulness and accuracy of predictive dose assessments based on historical environmental monitoring data in the Marshall Islands will be critically dependent on the precise determination of $\lambda_{effective}$ and associated uncertainties for each atoll environment. As an example, the mean effective and environmental-loss half-life rate constants for ^{137}Cs on Bikini, Enewetak and Rongelap Atolls at the 95% confidence level are reportedly 8 to 9.8 years, and 11 to 15 years, respectively. Using the most conservative upper confidence level, the resultant 30–, 50– and 70–year integrated doses are 58, 46 and 41%, respectively, of those previously calculated based on radiological decay alone. These data and information are essential in helping the United States Department of Energy (DOE) provide accurate predictions of future doses and potential health risks associated with the resettlement of affected atolls, and helps build confidence that resettlement programs can be accomplished in a sustainable, and environmentally and public health protective manner.

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