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A COMPARATIVE STUDY ON ^{137}Cs TRANSFER FROM SOIL TO VEGETATION IN THE MARSHALL ISLANDS

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ABSTRACT

The degree of ^{137}Cs root uptake into tropical fruits, vegetables and native foliage from coral atoll soils in the Marshall Islands has been quantified using soil-to-plant transfer factors. Transfer factor values for different plants on Bikini Island (Bikini Atoll) range between 0.2 and 144 and, in general, are considerably higher than those observed in continental soils. In this paper we will attempt to summarize available data on soil-to-plant transfer factors and examine some of the fundamental processes controlling the mobility and availability of ^{137}Cs in coral atoll soils.

1. INTRODUCTION

Bikini and Enewetak Atolls form part of the Western (Ralik) island chain of the Marshall Islands in the north equatorial Pacific Ocean and are former U.S. nuclear test sites (1946-58). Nuclear weapons detonations occurring in the near surface environment of coral atoll islands or over shallow water produced close-in radioactive fallout leading to contamination of islands/atolls within the northern Marshall Islands above that expected from global fallout deposition (Robison 1997a, Simon 1997). The most notable event of the entire Pacific test series was the BRAVO shot of 1 March 1954. BRAVO had an estimated explosive yield of 15 MT and produced significant localized inputs of radioactive fallout over much of the northern Marshall Islands. An extensive compilation of scientific papers dealing with the history, radiological monitoring, dose estimates, health effects, environmental studies, and related papers were recently published in a special issue of *Health Physics* on “Consequences of Nuclear Testing in the Marshall Islands” (*Health Physics*, Vol. 73).

The Lawrence Livermore National Laboratory (LLNL) has conducted a high-profile radioecology and dose assessment program in the Marshall Islands since 1973. Environmental characterization and radionuclide monitoring are used as a basis for improving estimates of the potential radiological dose at Bikini, Enewetak, Rongelap, Utirik, and other atolls. Since the early 1970s, we have collected over fifty thousand environmental samples (including soils, sediments, vegetation, water and aerosols samples) for detailed radiometric analysis, and to study rates of radionuclide uptake, redistribution, resuspension, and removal in the atoll ecosystem. ^{137}Cs contribute up to 95% of the estimated dose via ingestion (Robison 1997b, Robison 1982). The major dietary pathway for ^{137}Cs uptake is through consumption of coconut and other locally grown food crops such as *Pandanus*, breadfruit and papaya. Consequently, our research studies have focused on the need to quantify and develop a fundamental understanding of ^{137}Cs transfer from soil into coconuts and other tropical fruits, and to examine potential remedial measures to reduce doses to resident (returning) populations. This paper provides a comparative assessment of ^{137}Cs uptake in vegetation through a "BRAVO" contamination gradient extending from Bikini Atoll to the east over Rongelap, Rongerik, and Utirik Atolls. The uptake of radionuclides by plants from soils is described by a concentration ratio (CR), or soil-to-plant transfer factor, defined by the radionuclide concentration in vegetation divided by the radionuclide concentration in soils (in Bq kg^{-1} dry weight plant to Bq kg^{-1} dry weight soil normalized to a depth of 40 cm). We consider successively the range of CR values observed between different plants (largely food crop products), the spatial variation of CR values between atolls, and environmental processes controlling the mobility and availability of ^{137}Cs in coral atoll soils.

2. MATERIALS AND METHODS

2.1 Study Area

The Marshall Islands consist of two archipelagic island chains of 30 atolls and some 1,152 separate islands. Bikini and Enewetak Atolls form part of the Western (Ralik) island chain and are former U.S. nuclear test sites. During the years 1946 through 1958, a total of 66 nuclear devices were denoted in the near surface atoll environment having a combined explosive yield of approximately 108 Mt TNT equivalent. The most notable event of the entire Pacific test series and the largest U.S. thermonuclear device ever denoted was the castle-BRAVO experimental shot of 1 March 1954 at Bikini Atoll. BRAVO delivered a greater than predicted explosive yield of approximately 15 MT TNT equivalent and produced high levels of radioactive fallout over much of the northern Marshall Islands (Fig. 1).

Marshall Islands soils are composed almost entirely of Ca-Mg-Sr carbonate with varying amounts of organic matter and essentially little or no aliminosilicate material. Due to the absence of clays, the cation exchange capacity of soil is directly related to the organic content (Walker 1997). Soil organic matter is derived largely from the steady contribution of litter from surrounding vegetation. This is a critical process in maintaining fertility and stability of coral atoll soils. Coconut palms (*Cocos nucifera*) are the most common tree and provide a variety of prepared foods in Marshallese culture. Other important native food crop plants and tropical fruits include breadfruit (*Actocarpus altilis*), Pandanus (*Pandanus spp*.), papaya (*Carica papaya*), and arrowroot (*Tacca leontopetaloides*).

2.2 Field Sampling and Analytical Methods

The Lawrence Livermore National Laboratory (LLNL) has conducted field sampling expeditions to the Marshall Islands on a quarterly and/or semiannual basis since the 1970s. Field collections were designed to take representative samples of locally grown foods, tropical fruits and native foliage (e.g., Scaevola, Pisonia and Messerschmedia leaves) along with associated soil through the root zone of most of the sampled plants. All samples collected in the field were stored frozen and returned to LLNL for processing and radiometric analysis.

Dried, homogenized samples were prepared and analyzed in tuna cans on a gamma spectrometer coupled to a DEC VAXStation operating under Canberra/Nuclear Data systems data acquisition and reduction software. ^{137}Cs was quantified using the gamma photon peak at 661 keV. All samples of soil and vegetation were counted until a minimum of 5% precision was obtained on ^{137}Cs or for a maximum of 1000 minutes. Detector calibrations were performed using

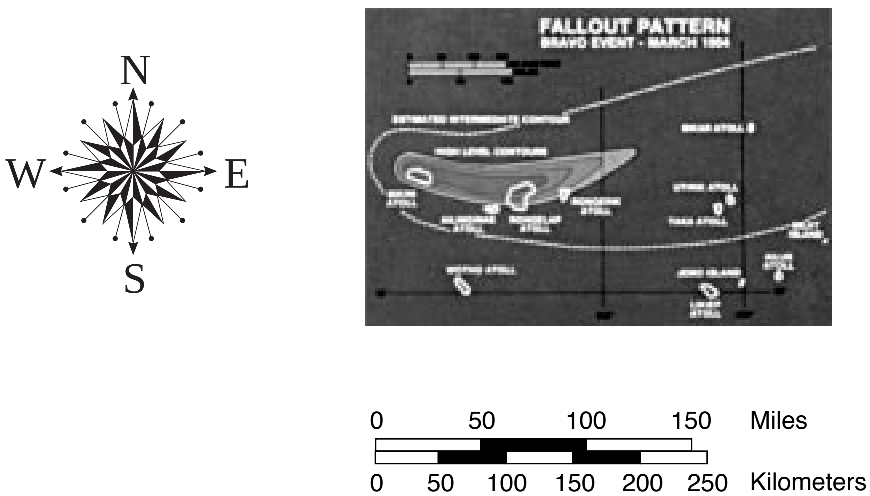

Fig. 1 Northern Marshall Islands Aerial Radiation Survey
(Fallout Pattern-Bravo Event of 1 March 1954)

NIST traceable standards. Acceptance data were evaluated on the basis of established quality control criteria for standards and duplicate analyses.

3. RESULTS AND DISCUSSION

The activity concentration of ^{137}Cs in soils from the Marshall Islands show a negative exponential decrease with increasing soil depth (Fig. 2). The ^{137}Cs depth distribution is controlled my a number of important factors including the original ^{137}Cs depositional history of the island/atoll, the age/stability of the soil and litter pile, the degree of erosion and/or physical disturbance to the soil, and the relatively high cation exchange capacity of the organic-rich surface layers which appears to limit downward migration of ^{137}Cs. The total ^{137}Cs deposit on Bikini Island (Bikini Atoll) is about 80 kBq m^{-2}. This compares with an estimated ^{137}Cs global fallout deposit in the Marshall Islands of around 0.4-0.6 kBq m^{-2} (Simon 1997). We attribute much of the excess deposit as being directly attributable to close-in fallout from BRAVO. The total ^{137}Cs deposit on Kabelle Island (northern Rongelap Atoll), Rongerik Island (Rongerik Atoll) and Utirik Island (Utirik Atoll) are 32, 18 and 1.5 kBq m^{-2}, respectively. Interestingly, our results confirm that Rongelap Island located to the south of Rongelap Atoll received significantly less fallout (about 13 kBq m^{-2} of ^{137}Cs) than Kabelle Island in the northern part of the atoll as predicted from the fallout trajectory of the BRAVO event (Fig. 1). ^{137}Cs fallout deposition over Utirik Island (Utirik Atoll), about 500 km east of Bikini Atoll, still appears to be about a factor a three higher than on islands/atolls to the west of Bikini (e.g. Ujelang Island).

Soil-to-plant ^{137}Cs transfer factors for various tropical fruits and foliage from Bikini Island have been summarized in Table 1. The soil-to-plant transfer factors appear to be extremely variable—the full range of values for different plants fall between a low of 0.2 for banana to a high of 144 for Chinese cabbage. ^{137}Cs transfer factors for drinking coconut meatrange between 2.8 and 64 with a mean of 16. Variability is also observed when comparing uptake into different parts of the same plant, e.g. coconut meat versus coconut fronds. By comparison, ^{137}Cs soil

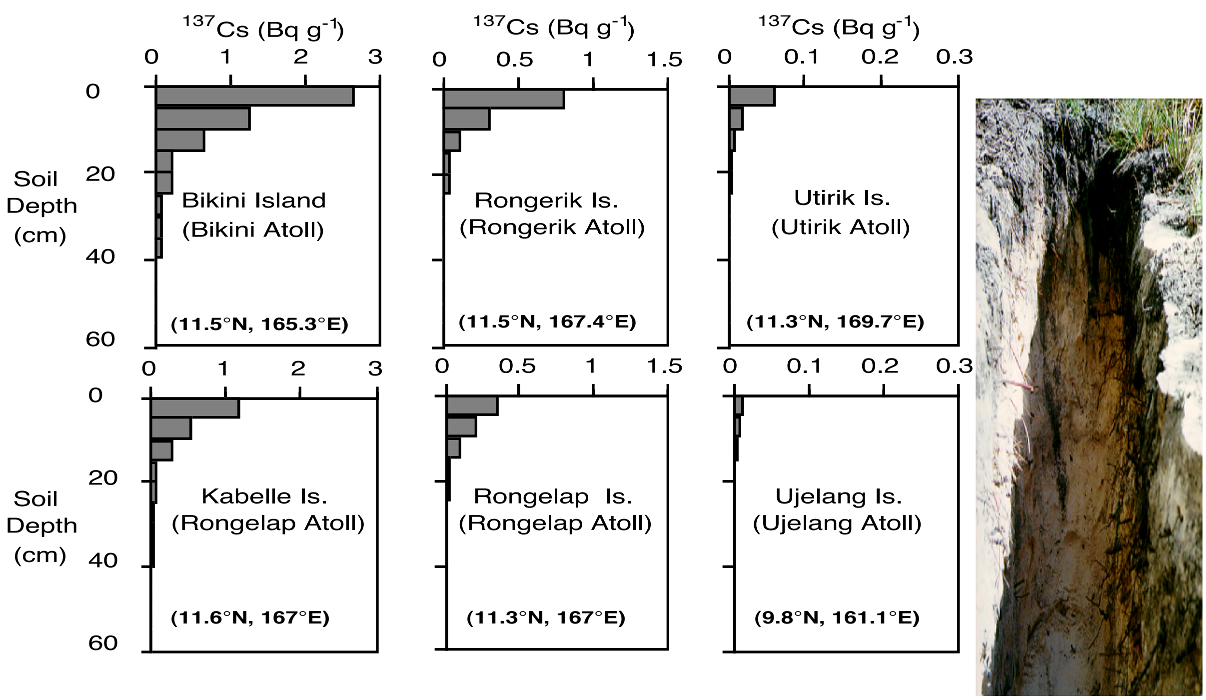

Fig. 2 ^{137}Cs soil depth-distribution profiles in northern Marshall Island soils.

concentrations (0-40 cm) over Bikini island vary by a factor of 5-6 (Table 1). Similarly, ^{137}Cs soil-to-plant transfer factors can vary up to several orders of magnitude in continental soils depending on the type, nature and fertility of the soils, the type of vegetation, and on agricultural/managementpractices (fertilization, ploughing, environmental conditions) (Frissle 1992). However, ^{137}Cs soil-to-plant transfer factors on Bikini Islands are generally much higher than those reported for temperate zones where typically values range between 0.005 and 0.5 (IAEA 1994).

We have also compared ^{137}Cs uptake in drinking coconut meat on Bikini Island with values from other islands/atolls. Soil-to-plant transfer factors on Bikini Island are considerably higher that those observed elsewhere. For example, the mean transfer factor for ^{137}Cs into drinking coconut meat on Rongelap Island (Rongelap Atoll) is about 2.4 (Table 1). This compares with a value of 16 on Bikini Island. Hence, there appears to be a largecoefficientof variability in attempting to define soil-to-plant ^{137}Cs transfer factors in coral atoll soils of the Marshall Islands. This occurs despite apparent similarities in the type, nature and development of the soils. Furthermore, the ^{137}Cs deposit on Bikini, Rongelap and Rongerik Atolls is derived largely from the BRAVO source term, and yet we see very significant differences in ^{137}Cs uptake in the same plants. What we do know is that Cs and K behave similarly, and that root uptake is enhanced in coral atoll soils because of the absence of clay minerals and the low concentration of K in the soil. The migration and availability of soil K and ^{137}Cs are strongly coupled with the development of the soil, and on the amount, type, and stability of the litter pile. The importance of K on ^{137}Cs uptake in continental crops has been well documented. We have also seen a dramatic reduction in ^{137}Cs uptake in tropical fruits (coconuts, breadfruit, *Pandanus*) after a single application of K rich fertilizer to selected experimental plots on Bikini Island (Robison 1992). Small natural differences in the concentration and availability of K in coral atoll soils may affect ^{137}Cs uptake and provide a real source of variability in soil-to-plant concentration factors (both between adjacent soils and/or across different island/atolls). If this is true, climatic and other

Table 1. ^{137}Cs soil-to-plant transfer factors on Bikini Island (Bikini Atoll)

Product	Location	^{137}Cs (Bq kg^{-1}, dry weight) Soil Median	Plant Median	^{137}Cs Transfer Factor Median	Mean	Maximum	Minimum
Drinking Coconut Meat	Bikini island	924	13537	13	16	64	2.8
	Rongelap island	127	175	1.5	2.4	18	1.4
Pandanus	Bikini Island	422	20045	35	44	82	11
	Rongelap Island	117	947	9.4	12	44	0.94
BreadfruitMeat	Bikini Island[b]	680	2217	2.9	3.4	7.4	0.44
	Rongelap Island	122	456	3.5	4.5	13	1.0
Papaya Meat	Bikini Island	1626	19959	11	22	62	2.1
Banana Meat	Bikini Island	427	487	0.78	0.71	1.2	0.27
Sorghum[a]	Bikini Island	3045	26103	8.1	10	24	0.81
Chinese Cabbage[a]	Bikini Island	1029	45364	36	41	144	2.9
Coconut Fronds	Bikini Island	859	2618	3.2	4.3	22	0.5
MesserschmidiaLeaves	Bikini Island	744	13973	4.6	13	32	4.3
ScaveolaLeaves	Bikini Island	1993	5812	8.5	12	31	1.8
GuettardaLeaves	Bikini Island	874	8611	23	23	44	1.9

[a] 0-20 cm root zone was used.
[b] Breadfruit trees were transplants from Eneu Island. Notes: Transfer factor values are expressed as Bq kg^{-1} dry weight plant to Bq kg^{-1} dry weight soil.

environmental conditions may influence ^{137}Cs uptake in plants. For example, ocean sea spray could provide an important source of available K on narrow islands with relatively short ocean breaks while the interior of larger islands will presumably be more sheltered. This would have the effect of increasing the apparent soil-to-plant transfer factors within the interiors of some of the larger islands with established vegetation—this indeed appears to be true on Bikini Islands where soil-to-plant transfer factors are high compared with those on Rongelap Island (Rongelap Atoll). Interestingly, Kabelle Island has a long extension where the potential for carry over of sea spray onto land is very high. We are presently planning a series of experiments to test this hypothesis.

There is clear evidence that nuclear weapons detonations on Bikini and Enewetak Atoll produced highly localized inputs of radioactive fallout. The ^{137}Cs soil deposit in the northern Marshall Islands decreases with increasing distance from the atolls along lines of constant latitude (e.g. 10–12°N) as might be expected from the normal direction of the equatorial tradewinds. A very strong contamination gradient exists over Bikini, Rongelap and Rongerik Atoll as a direct result of BRAVO. It is also highly probably that BRAVO also produced a contamination gradient through space and time in terms of the size and nature of radioactive particles. Hence, another important factor leading to variability in soil-to-plant transfer factors in Marshall Islands soils may associated with the geochemical behavior (e.g. availability, leachability, weathering effects) and aging of close-in fallout contamination. We anticipate the need for more detailed studies in this area in order to model the long-term behavior of ^{137}Cs and other contaminants in atoll ecosystems.

4. CONCLUSION

Soil-to-plant transfer factors for ^{137}Cs in coral atoll soils of the Marshall Islands vary over two orders of magnitude but are significantly higher than those observed in continental soils. This is attributed to the absence of clay minerals and the low concentration of available K in the soil. The migration and availability of ^{137}Cs for root uptake is strongly coupled with that of K. Important sources of variability in ^{137}Cs plant-to-soil transfer in coral atoll environments include the development of the soil, and the amount, type and stability of the litter pile. We also raise the suggestion that ocean sea spray may contribute to the pool of available K in atoll soils and therefore influence ^{137}Cs uptake into plants. Similarly, the availability of ^{137}Cs derived from close-in fallout may have changed over space and time, and depend on the size, geochemical behavior, and aging of contaminated particles in the soil.

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REFERENCES

Frissel, M. An update of the recommended soil-to-plant transfer factors, Eighth Report of the IUR Working Group on Soil-to-Plant Transfer Factors, IUR, Balen, Belgium; 1992.

International Atomic Energy Agency. Handbook of parameters values for the prediction of radionuclide transfer in temperate environments, International Union of Radiologists, IAEA, Vienna, Technical Report Series No. 364; 1994.

Robison, W.; Noshkin, V.; Conrado, C. et al. The northern Marshall Islands radiological survey: data and dose assessments. Health Phys. 73(1): 37–48; 1997a.

Robison, W.; Bogen, K.; Conrado, C. An updated dose assessment for resettlement options at Bikini Atoll—U.S. nuclear test site. Health Phys. 73(1): 100–114; 1997b.

Robison, W.; Stone, E. The effect of K on the uptake of ^{137}Cs in food crops grown on coral soils: Coconut at Bikini Atoll. Health Phys. 62: 496–511; 1992.

Robison, W.; Mount, M.; Phillips, W. et al. An updated radiological dose assessment of Bikini and Eneu Islands at Bikini Atoll. Lawrence Livermore National Laboratory, Livermore CA, UCRL-53225; 1982.

Simon. S.; Graham, J. Findings of the first comprehensive radiological monitoring program of the Republic of the Marshall Islands. Health Phys. 73(1): 66–85; 1997.

Walker, R.; Gessel, S.; Held, E. The ecosystem study on Rongelap Atoll. Health Phys. 73(1): 223–233; 1997.