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# **Long –Term Reduction in <sup>137</sup>Cs Concentration in Food Crops on Coral Atolls Resulting from Potassium Treatment**

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### Abstract

Bikini Island was contaminated March1, 1954 by the Bravo detonation (U.S nuclear test series, Castle) at Bikini Atoll. About 90% of the estimated dose from nuclear fallout to potential island residents is from cesium-137 (137Cs) transferred from soil to plants that are consumed by residents. Thus, radioecology research efforts have been focused on removing 137Cs from soil and/or reducing its uptake into vegetation. Most effective was addition of potassium (K) to soil that reduces 137Cs concentration in fruits to 3-5% of pretreatment concentrations. Initial observations indicated this low concentration continued for some time after K was last applied. Long-term studies were designed to evaluate this persistence in more detail because it is very important to provide assurance to returning populations that 137Cs concentrations in food (and, therefore, radiation dose) will remain low for extended periods, even if K is not applied annually or biennially. Potassium applied at 300, 660, 1260, and 1970 kg ha<sup>-1</sup> lead to a 137Cs concentration in drinking coconut meat that is 34, 22, 10, and about 4 % of original concentration, respectively. Concentration of 137Cs remains low 8 to 10 y after K is last applied. An explanation for this unexpected result is discussed. *Keywords*: 137Cs, transport, K effect; reduction; duration of effect; dose impact; Marshall Islands

### 1.0 Introduction

U.S. nuclear testing in the Marshall Islands from 1946 to 1958, and especially the Bravo test at Bikini Atoll in 1954, contaminated Bikini Island with local fallout. Dose assessments for Bikini, Enewetak and Rongelap Atolls, demonstrate that the highest contribution to estimated dose from nuclear device fallout for people living, or returning to live, at the atolls is from <sup>137</sup>Cs (Robison et al., 1987, 1994, 1997,

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1999). The primary exposure pathway is consumption of locally grown foods that accumulate <sup>137</sup>Cs through the root system and deposit it in fruits, grains or leaves (Robison and Stone, 1992). About 90% of total estimated dose results from this process (Robison et al., 1997). The diet model, that includes locally grown and imported foods, is a very important part of dose assessments and is discussed in detail by Robison and Sun (1997).

Consequently, radioecology research efforts were focused on ways to either remove <sup>137</sup>Cs from soil and/or reduce its uptake into food crops (Robison and Stone, 1998). Most effective of all evaluated methods was applying K to the soil surface that subsequently is dissolved by rainfall and absorbed through vegetation root systems. Naturally low concentrations of K and lack of any significant amount of clay minerals in coral soil is the basis for enhanced uptake of <sup>37</sup>Cs into atoll plants. When additional K is available, however, trees readily accumulate it with a resulting effective reduction in <sup>137</sup>Cs concentration in plants. This in turn leads to significant reduction of <sup>137</sup>Cs in foods. Early results were very dramatic and <sup>137</sup>Cs concentration in coconuts was reduced to 5 to 10 % of pretreatment concentrations when K was applied at 1000 to 2000 kg ha<sup>-1</sup> (Robison and Stone, 1992). Similar reductions in uptake of <sup>137</sup>Cs by annual crops treated with K were observed in other experiments (Stone and Robison, 2002). Results of Experiment IV in the 1992 paper also indicated that <sup>137</sup>Cs concentration in coconuts remained at reduced levels for a period of about 2.5 y after the last application of K.

Confirming reduction in <sup>137</sup>Cs concentration and defining the period of time reduction lasts after K is last applied is very important. It provides assurance to returning populations that <sup>137</sup>Cs concentration will remain low in locally grown foods for extended periods even if K is not applied on an annual or biennial schedule. It would be hard to overstate the importance of this assurance to returning populations at the atolls. Moreover, frequency with which K treatment is required also greatly influences cost of remediation over many years. Continuing results obtained from Experiment IV in the 1992 paper, and four other independent experiments, are presented here to evaluate long-term effectiveness of K applied to food crops at former nuclear test sites.

### 2. Background

Detailed composition, and relative concentration of <sup>137</sup>Cs and K, of coral soil at atolls are discussed in Robison and Stone, 1992. Briefly, atoll soils are of marine origin and consist primarily of

calcium carbonate, some magnesium carbonate, organic matter (as high as 15%), and essentially no clays. The pH ranges from about 7.8 to 9.0 in water slurries. Concentrations of phosphorus (P) are quite variable and are as high as 1.5% in some locations because of aged guano deposits from nesting birds. Concentration of naturally occurring K is very low; the average for several atolls is 300 mg kg<sup>-1</sup> (Fosberg and Carroll, 1965). Extractable K ranges from 20 to 80 mg kg<sup>-1</sup> in the upper 25 cm of soil and diminishes rapidly below that depth (Robison and Stone, 1992). Uptake of <sup>137</sup>Cs and <sup>90</sup>Sr are very different in atoll carbonate soils compared with continental, silica soils that have significant quantities of clay minerals (Robison et al., 2000). Cesium-137 is bound primarily in the organic moiety of coral soil and is relatively available for uptake by plant roots.

### 3. Methods and Materials

### 3.1 Field procedures

Three experiments described here are based on results using coconut (Cocos *nucifera L.*) trees from various locations within the island-wide coconut grove planted between 1970 -1972 on Bikini Island. Drinking-coconuts are the standard sample because meat and fluid from this stage of coconut development are common dietary items and account for a significant portion of the <sup>137</sup>Cs intake. Time from development of inflorescence (flower) to drinking stage is about 9 mo. Nuts reach mature copra stage in about 12 to 13 mo. Thus, drinking coconuts collected prior to 9 mo after application of K have not had extra K available the entire development cycle; only flower stage nuts at, or shortly after, application of K have full benefit of added K.

Dense understory of scrub brush, vines, and volunteer coconuts within experimental areas in the coconut grove were cleared to begin each experiment. Subsequently, these areas were periodically mowed, or cleared by hand, to provide easy access for collecting samples of coconuts. Potassium was applied either as a full fertilizer - nitrogen, phosphorous, potassium (N, P, K) - or as coarse-crystal potassium chloride (KCl).

Drinking -coconut samples were collected during quarterly or semi-annual field trips. Initial samples were collected from each experiment before application of any K fertilizer. One sample from a tree consists of 5 to 7 drinking coconuts. Coconuts are husked, nuts are then punctured to collect fluid (liquid

endosperm) that is commonly drunk, and gelatin-like meat characteristic of drinking coconuts, remains in coconut shell. In the absence of drinking coconuts, mature copra-stage coconuts are collected. The ratio of <sup>137</sup>Cs concentration in copra-nut meat vs. drinking-coconut meat is 1.6.

Two other demonstrations involved 1 *Pandanus* tree and 3 closely spaced breadfruit trees located outside the coconut grove. The area around the trees was mowed to keep the grass and scrub brush under control. Potassium was applied as KCl. Samples consisted of 3 to 8 fruits depending on size.

All samples were frozen in the field within 3 h of collection and returned to Lawrence Livermore National Laboratory (LLNL) in Matson freezer vans.

### 3.2 Laboratory procedures

Drinking-coconut meat was extracted from cracked coconut shells; skin of breadfruit was removed before processing the sample; and juice in Pandanus fruits was squeezed under high pressure from the individual keys. All samples were dried to constant weight by lyophilization and ground to fine consistency in a Waring blender. Fluid volume was reduced by evaporation. Dried coconut meat and reduced fluid were packed into 8.0-cm diameter x 4.6-cm-high aluminum cans for gamma spectrometry analysis. Gamma analyses were performed in the LLNL Gamma Facility [GF] (Hamilton et al., 2000) that consists of 22 high-resolution, intrinsic, germanium gamma detectors. Standards and blind-duplicates, each totaling 10% of the samples submitted, were included in each batch of 50 to 100 samples sent to the GF. If results for standards were not within 10% of the known value reanalysis was required. Blind duplicates had to be within 10% of each other or that also triggered reanalysis. It has not been necessary to do reanalysis on any samples from these experiments. All values are converted to a wet weight basis throughout the paper unless specified otherwise.

# 3. 3 Field Experiment Design

### 3.3.1 Coconuts

Coconut experiments are identified by both number (1, 2, 3) and by island location (KNPK, CLC, B1 & B4 well sites).

Experiment 1 (KNPK)

The first experiment of three to be discussed was begun in 1985. An area of more than 4 ha within the coconut grove was cleared to begin the study. This experiment was listed as Experiment IV in the preliminary report in 1992 (Robison and Stone, 1992) and is located in an area of the coconut grove having the highest average surface-soil concentration of <sup>137</sup> Cs on Bikini Island, 2.6 to 3.7 Bq kg<sup>-1</sup>, as determined by a 1976 ground survey and 1978 aerial radiological survey (Gudiksen et al., 1976, Tipton and Meibaum, 1981). It is a 2-block design (Figure 1) with blocks separated by 25.5 m. Both blocks have six 17 x 94-m plots that contain three rows of trees. The center row in each plot was sampled. Plots are separated by one 8.5 m untreated strip. Initial factorial design in each block included three amounts of K (0, 1260 and 2520 kg ha<sup>-1</sup>), with and without the addition of a combined N-P application.

Initial application of fertilizer in 1985 was applied in four equal increments at 3 mo intervals. Initial (1985) applications of K (both alone and in combination with N-P) are shown by solid gray/black, N-P treatments (no K) by horizontal lines, and controls (no treatments) by a solid border with no internal markings (Figure 1). Fertilizers were distributed with a battery-driven centrifugal spreader mounted on the back of a truck. Several passes were made to distribute all the fertilizer and this helped provide a more uniform distribution of K. Fertilizer was applied 8.5 m on each side of the middle row of trees in each treated plot, i.e. to trunks of the neighboring rows of trees. Thus, sampled rows were treated on both sides while immediate neighbor trees were fertilized on only one side. This experiment lasted 33 mo from time of initial collection.

In November1987 the treatment area for 1260 kg ha<sup>-1</sup> rows and N-P + 1260 kg ha<sup>-1</sup> row in Block I, was expanded by adding a one-time application of 1260 kg ha<sup>-1</sup> to areas shown by vertical lines in Figure 1. In May 1989, plots in blocks I and II, identified by diagonal lines running from upper left to lower right in Figure 1, were treated with a single application of 1260 kg ha<sup>-1</sup> of K. Final K treatment to this experiment occurred November 1992 when a single application of 1260 kg ha<sup>-1</sup> was applied to plots in blocks I and II that are identified by diagonal lines running from lower left to upper right that in turn form a cross-hatching with the diagonal lines representing the 1989 K application (Figure 1). A summary of treatments is listed in Table 1. Subsoil lines (vertical dashed lines in Figure 1) were dug the length of row H and O (block II only) using a bulldozer and a large subsoil bar that cut roots of trees to a depth of 3 feet to prevent roots of trees in control plots and N-P-only plots in blocks I and II from getting into fertilized areas.

Previously published data (Robison and Stone, 1992) showed a decrease in <sup>137</sup>Cs concentration in rows treated with N-P. At that time we speculated which element might be causing the effect and really wondered why either of them should cause any effect at all. Another small experiment was implemented that included control trees in which coconut trees were treated with N and P independently. No effect was observed on the uptake of <sup>137</sup>Cs. Commercial N-P previously used for large-scale experiments was then analyzed for K. It contained K in such quantities that when N-P was distributed in the original experiment it provided a K concentration of 300 kg ha<sup>-1</sup>. Thus, reduction in concentration of <sup>137</sup>Cs in coconuts resulted from K in the N-P although the lesser amount of K produced less of a reduction in <sup>137</sup>Cs concentration than did larger quantities of K. These additional quantities (300 kg ha<sup>-1</sup>) are included in Table 1.

Experiment 2 (CLC)

A 1 ha area adjacent to Experiment 1 (KNPK) was cleared about 1 y later to establish Experiment 2. It is a 2-block design separated by 8.5 m. with one block receiving K treatment and the other a control receiving no K treatment. Each block of coconut trees is 9 rows long and 8 rows wide (76.5 m x 68 m). Sampled trees are interior 20 trees in both 72-tree plots as shown in Figure 2. Distance between sampled trees in the two blocks is 42.5 m. Potassium in the K-treated block was distributed over the entire 9 x 8-tree area, shown by diagonal lines, using a large spreader run off the power-take-off (PTO) of a small tractor. Initial samples were collected from all trees before K was applied. Potassium treatments were 660 kg K ha<sup>-1</sup> (August 1988), 660 kg K ha<sup>-1</sup> (December 1992), and a final 750 kg K ha<sup>-1</sup> (November 1993). All were one-time applications.

Experiment 3 (B1 and B4 well sites)

Areas for these two sites were cleared in early 1990. They are located about 600 m from each other in the coconut grove. Four interior trees in each 6 x 6-tree plot were selected for long-term sampling. Potassium was distributed over the entire 36-tree plots using a large spreader run off the PTO of a small tractor. Four ground-water wells are in the center of each of these plots. Initial samples (time zero) were collected prior to distribution of K at both sites and serve as the only designed controls. A total of 9044 kg K ha<sup>-1</sup> was distributed over the B1 plot and 4344 kg K ha<sup>-1</sup> over the B4 plot. These sites were originally designed for an entirely different purpose but they are also relevant in the context of this paper.

### 3.3.2 Pandanus and breadfruits

The *Pandanus* tree received six 1000 kg K ha<sup>-1</sup> treatments over 2.5 y that were spread in a 12 m radius from the trunk of the tree. Breadfruit trees received about 5900 kg K ha<sup>-1</sup> over a 14-y period spread in a 20 m radius around the trees.

### 4. Results

### 4.1 Coconut experiment number 1

Results from Experiment 1 are shown in Figure 3. Control data are shown for the first 3 y of the experiment. Control rows were then inadvertently contaminated with K when their tree roots invaded adjacent K-treated areas prior to establishing the subsoil-lines. Consequently, trees showed a gradual decline in <sup>137</sup>Cs concentration although it was still well above treated rows. The first 3 y of uncontaminated control data are sufficient to show the vast difference between treated and untreated trees. Moreover, control trees in Experiment 2 (discussed below), as well as control trees scattered around the island (Robison et al., 2003), serve as surrogate controls. The abrupt decrease of <sup>137</sup>Cs visible in controls 9 mo after initial treatment followed by an appreciable recovery, is attributed to a "clearing effect" produced by destruction of some surface roots, localized shifting of the uppermost layer of soil, and off-site removal of some surface soil mixed with vegetation at the time of clearing. This phenomenon is observed in all experiments where major clearing of understory is required.

Samples collected 9 mo after K was first applied represented a 3 to 9 mo response to only 3 of 4 scheduled fertilizer applications. Minimum <sup>137</sup>Cs concentration (i.e. maximum decrease) for trees treated with 1260 kg K ha<sup>-1</sup>, or more, occurred18 mo after first application of K which is about 6 mo after the last of 4 initial treatments. Cesium-137 concentration remained constant for the next 2.5 y (4.5 y point, Figure 3) at which time a second treatment of 1260 kg ha<sup>-1</sup> of K was applied. A further decrease in <sup>137</sup>Cs concentration occurred during the following year. This newly established <sup>137</sup>Cs concentration remained constant for 3 y at which time a third treatment of 1260 kg K ha<sup>-1</sup> was applied. Once again another slight decrease occurred in <sup>137</sup>Cs concentration. The row of trees receiving two 1260 kg K ha<sup>-1</sup> treatments (represented by open circles in Figure 3), but not the third treatment, stabilized at a higher <sup>137</sup>Cs concentration (about 8% of pretreatment <sup>137</sup>Cs concentration) than 4 rows of trees receiving all three treatments (about 3% of pretreatment <sup>137</sup>Cs concentration). The low <sup>137</sup>Cs concentration in coconuts has

persisted for 10 y after the last application of K for the row receiving two treatments (open circles in Figure 1) and for 7 y since the last K treatment for 4 rows receiving 3 K treatments.

The single row of trees receiving only one 2520 kg K ha<sup>-1</sup> application in 1985 (open squares Figure 3) began to show a slight increase in <sup>137</sup>Cs concentration in coconuts after a little over 4 y (shortly after the second application of K to other rows). The increase continued for about 1.5 y and then stabilized at 17% of pretreatment concentration for 8 y.

One row of trees that was intended to receive only an N-P treatment actually received 300 kg K ha<sup>-1</sup> from K contamination in commercially obtained N-P. Results of this limited treatment with K are represented by "x" in Figure 3. Cesium-137 concentration in coconuts stabilized at about 34% of pretreatment concentration for a period of 11 y. Another row of trees treated initially with N-P (and therefore 300 kg ha<sup>-1</sup> of K) received additional K at 2.5 y as a result of the partial-root expansion experiment and then received second and third applications of 1260 kg K ha<sup>-1</sup>. Cesium-137 stabilized at about 3% of pretreatment concentration as did other trees receiving multiple applications 4.2 Coconut experiment number 2

Results for experiment 2 are shown in Figure 4. Potassium treatments were applied in a single application. After K was applied there was a rapid decrease in <sup>137</sup>Cs concentration in coconuts (samples collected 3 and 9 mo after K application) with the lowest concentration occurring after about 1 y. It began to increase about 2 y later, and continued for two more years until another application of 660 kg K ha<sup>-1</sup> was applied. Another significant decrease in <sup>137</sup>Cs concentration followed. A little over a year later another 750 kg K ha<sup>-1</sup> was applied. Cesium-137 concentration in coconuts has remained at 5 to 6% of pretreatment concentrations for a period of 8.5 y since K was last applied.

Cesium-137 concentration in control trees once again fluctuated because of disturbance of surface roots and redistribution of some surface soil associated with clearing of the site. It stabilized about 2 y after clearing. The gradual decline thereafter in  $^{137}$ Cs concentration in control trees is due to radiological decay of  $^{137}$ Cs ( $T_{1/2} = 30.1$  y) and loss of  $^{137}$ Cs from soil when rainfall is adequate to saturate the soil and cause recharge of the ground-water lens. Loss of  $^{137}$ Cs by this environmental process is more rapid than loss from radiological decay (Robison et al., 2003) and has important implications on performing accurate dose assessments associated with resettlement.

## 4.3 Coconut experiment number 3

Results for the two well sites are shown in Figure 5. Data Samples taken prior to K treatment (time zero) are the only direct controls for these sites. However, controls from other experiments such as Experiment 2 above and individual control trees around the island serve as surrogate controls (Robison and Stone, 1992; Robison et al., 2003). Large amounts of K were distributed on these two sites to determine if a continuing source of K would accelerate loss of <sup>137</sup>Cs from soil to the ground-water lens. That part of the experiment is not reported here but results do provide additional data on reduction of <sup>137</sup>Cs in coconuts as a result of added K and duration of reduction after last application of K.

Rapid decline in <sup>137</sup>Cs concentration observed in other experiments is seen once again in this experiment. Cesium-137 concentrations have remained at the lowest value, which for the 2 sites is 4% and 8% of pretreatment values, for 3.5 y since K was last applied. Further sampling may show a low concentration of <sup>137</sup>Cs well beyond the currently observed 4 y.

### 4.4 Other trees

One *Pandanus* tree and 3 closely spaced breadfruit trees that were treated with K show the same significant reduction in <sup>137</sup>Cs as do coconuts and have maintained this low concentration for 5.5 y and 8 y, respectively, since K was last applied. Future sampling may show an even longer period than the currently observed 5.5 and 8 years.

4.6 Fractional <sup>137</sup>Cs reduction as a function of total applied K

Fractional reduction in the original <sup>137</sup>Cs concentration in coconut trees as a function of total applied K is shown in Figure 6. A total application of 300 kg K ha<sup>-1</sup>, 660 kg K ha<sup>-1</sup>, 1260 kg K ha<sup>-1</sup>, and 1970 kg K ha<sup>-1</sup> lead to a <sup>137</sup>Cs concentration in drinking coconut meat that is 34 %, 22 %, 10 %, and about 4 % of the original concentration, respectively. Beyond about 2000 kg K ha<sup>-1</sup>, even up to 9044 kg K ha<sup>-1</sup>, there in no further decrease and all results are between 3 and 5 % of the original concentration.

### 5.0 Discussion

Cesium-137 concentration is significantly reduced in fruits of trees after initial treatment with K.

In Experiment 1, trees in row I (Block II) that received 330 kg K ha<sup>-1</sup> K via N-P treatment stabilized at

about 34% of pretreatment concentrations (represented by "x" in Figure 3). This demonstrates that 300 kg K ha<sup>-1</sup> was not adequate to supply all of the K that trees are capable of accumulating. Cesium-137 concentrations dropped to between 8 and 16% of pretreatment concentrations in 1 y for all trees receiving 1260 kg K ha<sup>-1</sup> or 2520 kg K ha<sup>-1</sup>. <sup>137</sup>Cs concentration remained at this level for about 3 y at which time K was again applied creating a further decline in <sup>137</sup>Cs concentration to 6 to 8% of pretreatment concentrations. It remained at this level for 3 years when a third treatment of K was applied. This produced the final drop in <sup>137</sup>Cs concentration to about 3% of pretreatment levels. It has remained at this level for 8 y since K was last applied.

A good example of the effect of heavy rainfall after application of K to soil is apparent in trees that received 2520 kg ha<sup>-1</sup> total K in two separate 1260 kg K ha<sup>-1</sup> applications (open circles in Figure 3). The second treatment was applied in May 1989 about 3.5 y after the 1985 treatments. Monthly rainfall totals in 1989 for May, June, and July were 13.7 cm (5.4 in), 30.5 cm (12 in), and 35.7 cm (14 in), respectively. Consequently, there was adequate rainfall to leach an unknown, but very likely significant, portion of applied K from soil to ground water before uptake into trees could occur. Cesium-137 concentration has remained at about 8% of pretreatment concentration for this row of trees while <sup>137</sup>Cs concentration in four remaining rows that received a third K treatment dropped to 3% of pretreatment concentration. A total of 2520 kg K ha<sup>-1</sup> added in two applications was sufficient to drop<sup>137</sup>Cs concentration to the 3% level (Figure 6) had not considerable K been lost from the second treatment due to heavy rainfall. But because K was lost from treatment 2, the third treatment still produced a further reduction in the other 4 rows of trees.

High rainfall might also explain why <sup>137</sup> Cs concentration in coconut trees didn't drop to about 3 to 4 % of pre-treatment concentration after the original (1985) 2520 kg K ha<sup>-1</sup> treatments as might be expected based on Figure 6. Rainfall after the February 1985 application of K was 3.7 cm (1.45 in) in February, 3.4 cm (1.3 in) in March, and 6.6 cm (2.6in) in April. These totals came in small amounts over many days of the month. This is adequate rainfall to dissolve the KCl but insufficient to cause movement of K beyond the tree roots. However, the story is very different for the 3 following increments of applied K. The second increment of K applied in May was followed by unusually high rainfall of 18.2 cm (7.15 in) in May, 11.5 cm (4.5 in) in June, and 8 cm (3.1 in) in July. After the third increment of K was applied in August, there

was 19.5 cm (7.8 in) rainfall over the rest of the month, 20 cm (7.9 in) in September and 17.3 cm (6.8 in) in October. After the November 1985 application of K there was 10.8 cm (4.3 in) and 10.9 cm (4.3 in) in November and December, respectively. Thus, rainfall after the May, August, and November applications of K was adequate to cause transport of a significant amount of K out of the root zone to ground water thereby reducing the effective application of K below 2520 kg K ha<sup>-1</sup>. This is true for trees receiving 1260 kg K ha<sup>-1</sup> as well.

Cs-137 concentration in trees (row C, Block I in Figure 1; open squares in Figure 3) that received only one 2520 kg K ha<sup>-1</sup> treatment of K in 1985 remained low for a period of 14 y and has stabilized at about 17% of pretreatment concentration for a period of 10 y. This row of trees would have stabilized at less than 17% if not for two important factors that reduced the effective amount of applied K. After the experiment was underway for several months, we discovered that trees from Block I border rows A and B and control row F had roots in the row C treated area and competed for the K applied to row C. Moreover, trees in row C were also affected by 1985 rainfall as described above. Thus, effective K applied to row C was less (probably significantly less) than 2520 kg K ha<sup>-1</sup>. The remaining trees, that received 1970 kg K ha<sup>-1</sup> or more, all stabilized at about 3 % to 4% of pretreatment concentrations.

An interesting feature of the very significant reduction in <sup>137</sup>Cs concentration in food crops as a result of K treatment is the unexpectedly long period of time that <sup>137</sup>Cs remains at these low levels after K was last applied. When KCl is spread on the soil surface and dissolved by rainfall, K ions become available for uptake by plants and transport to groundwater. Annual rainfall ranges from about 110 cm in a dry year to 240 cm in a wet year. Moreover, about 80% of total rainfall is delivered over a 6-mo period (June through November). Annual rainfall most years is adequate to produce a flow of fresh water to the groundwater lens on several occasions during rainy season. Applied K is readily leached to the ground water so there is very little applied K remaining in soil after one rainy season. Yet the low <sup>137</sup>Cs concentrations obtained as a result of K treatment persist.

An explanation comes from analysis of K in entire coconut trees that were sacrificed several years after the last application of K. Results indicate that when additional K is available trees accumulate large quantities of K. This provides a large reservoir of K that can be transported to fruits for many years.

Amounts of K in entire fertilized coconut trees are greater by a factor of 4.8 than amounts of K in entire

unfertilized trees (3.9 g kg<sup>-1</sup> vs. 0.65 g kg<sup>-1</sup>, respectively). The K ratio between fertilized and unfertilized trees in the two major storage compartments off trees, trunk and fronds that supply K to the fruits, is 5.6 and 5.3, respectively. This mechanism for uptake and storage of K is also active in other major food-trees on the islands. Identifying the process that creates long-term reduction in <sup>137</sup>Cs concentration in food-bearing trees is important to provide a basis for assurance that radiation dose from terrestrial foods will remain low for years after K is the last applied.

Reduction in <sup>137</sup>Cs concentration in three breadfruit trees and one *Pandanus* tree treated with K is very similar to results for coconut trees; they have remained at these low concentrations for 8 and 5.5 y, respectively, since K was last applied. This indicates the duration of the K effect is common to all the major fruit crops consumed by the population. In addition to coconut, breadfruit, and *Pandanus* tree experiments, many types of annual food crops have been grown at the island to evaluate effectiveness of K to reduce <sup>137</sup>Cs concentration should these crops ever become part of the diet. Significant reduction in <sup>137</sup>Cs concentration is observed with all tested crops when additional K is supplied. Results for some of these crops are reported in detail in Stone and Robison, 2002. Data from many other crops, evaluated over many years as demonstration trials, show similar reductions in <sup>137</sup>Cs when treated with K, but remain unpublished. Some tested crops could not be grown at all without added K and others, such as papaya and banana that will grow, show much greater productivity with added K.

It is clear that treatment with K greatly reduces <sup>137</sup>Cs concentration in food-crops at the atolls. From practical experience it is prudent to split the total amount of K to be applied into at least 2 portions applied a few weeks or months apart to avoid a large loss of K during periods of heavy rainfall.

# 6.0 Conclusions

Based on data from many experiments there is every reason to have confidence that long-life fruit trees in the Marshall Islands (coconut, breadfruit, *Pandanus*, other fruit trees, etc.), when treated with proper amounts of K, will maintain a very low concentration of <sup>137</sup>Cs in edible fruits for many years. Therefore, resettling populations can be assured that radiation dose resulting from consumption of local foods containing <sup>137</sup>Cs will remain low for the same period of time. Moreover, K treatment would not be required for 8 to 10 y if one wanted to wait that long. This certainly reduces concerns about applying K (or forgetting to apply K) on an annual or biannual basis and cost of remedial action. Because of very low

natural concentration of K in atoll soil, many annual crops will require K (and N, P) treatment at time of planting and during the growth cycle just to survive. This fertilizer treatment vastly improves growth and productivity of annual plants growing in nutrient deficient coral soils and concurrently reduces <sup>137</sup>Cs in a manner similar to that observed in trees.

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### References

- Fosberg, F. R., Carroll ,D.,1965. Terrestrial sediments and soils of the northern Marshall Islands. *Atoll Res.*. *Bulll.*, **113**, 1–156.
- Gudiksen, P. H, Crites T R., Robison W. L., 1976. External Dose Estimates For Future Bikini Atoll Inhabitants, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-51879 Rev. 1.
- Hamilton, T.F. et al.,2000, The Marshall Islands Radioassy Quality Assurance Program: An Overview, *Journal of Radioanalytical and Nuclear Chemistry*, **243**(2), 415–422.
- Robison, W. L., Conrado, C. L., Phillips, W. A., 1987. *Enjebi Island Dose Assessment*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53805.
- Robison, W. L., Stone, E. L., 1992. The effect of potassium on the uptake of <sup>137</sup>Cs in food crops grown on coral soils: coconut at Bikini Atoll, *Health Physics*, **62**(6), 496–511.
- Robison, W. L., Conrado, C. L., Bogen, K. T., 1994. *An Updated Dose Assessment for Rongelap Island*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-107036.
- Robison, W. L., Sun, C., 1997. The use of comparative <sup>137</sup>Cs body burden estimates from environmental data/models and whole body counting to evaluate diet models for the ingestion pathway. *Health Physics*, **73**(1) 152–166.

- Robison, W. L., Bogen, K. T., Conrado, C. L., 1997. An updated dose assessment for resettlement options at Bikini Atoll—A U.S. nuclear test site. *Health Physics*, **73**(1), 100–114.
- Robison, W.L., Stone, E. L., 1998. The evaluation of critical pathways, radionuclides, and remedial measures for reducing the radiological dose to returning populations at a former nuclear test site,, 1998. In: *HLW, LLW, Mixed Wastes and Environmental Restoration—Working Towards a Cleaner Environment, Session 17—ER Progress in Cleaning Up Nuclear Weapons Test Sites-II*, Tucson, Arizona, March 1–5, 1998.
- Robison, W. L., Conrado, C.L., Bogen, K. T., 1999. *Utirik Atoll Dose Assessment*. Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-135953.
- Robison, W. L., Conrado, C. L., Hamilton, T. F., Stoker, A. C., 2000. The effect of carbonate soil on transport and dose estimates for long-lived radionuclides at a U. S. Pacific test site. *Journal of Radioanalytical and Nuclear Chemistry*, **243**(2), 459–465.
- Robison, W. L., Bogen, K. T., Conrado, C. L., Stoker, A. C., 2003. *The Effective and Environmental Hal- Life of* <sup>137</sup>*Cs at Coral Islands at the Former U. S. Nuclearr Test Site*, 2003. Journal of Env.

  Radioactivity, 69, 207-223.
- Stone, E. L., Robison, W. L., 2002. Effect of Potassium on Uptake of <sup>137</sup>Cs in Food Crops Grown on Coral Soils: Annual Crops at Bikini Atoll, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-147596.
- Tipton, W. J., Meibaum, R. A., 1981. *An Aerial Radiological and Photographic Survey of Eleven Atolls and Two Islands within the Northern Marshall Islands*, EG&G, Las Vegas, NV, EGG-1183-1758.

Figure 1. Experiment 1 (KNPK) is a 2-Block design 22 rows x 24 rows. Numbers and letters at the front end of rows indicate initial treatment in February1985. Each rectangle is 3 trees wide (represented by 3 alphabetic letters) and 10 trees long (1 to10 and 13 to 22 in Blocks I and II, respectively). K-treated rows (rows C and S and C and P in blocks I and II) and the NP+K-treated rows (rows I and M and F and S in Blocks Í and II) are identified by rectangles of solid, light gray color. NP-only treated rows (row P and I in Blocks I and II) are identified by rectangles with horizontal lines. Control rows (no K or NP treatment) are identified by blank rectangles (rows F and M in blocks I and II). Original amounts of K or NP added in 1985, in kg ha<sup>-1</sup>, are shown at the start of each row. Vertical lines represent an expansion of K treatment (1260 kg ha<sup>-1</sup>) in November of 1987. Diagonal lines running from upper left to lower right represent an additional application of 1260 kg ha<sup>-1</sup> of K in May1989. Diagonal lines running from lower left to upper right, that form cross-hatching with the 1987 treatment, represent an application of 1260 kg ha<sup>-1</sup> in November 1992 (rows H to N and O to U in Blocks I and II).

**Figure 2.** Experiment 2 (CLC) is a 2-Block design with significant separation between K-treated and control blocks. The 20 interior trees (4 x 5 trees outlined in the center of each block) were sampled within the 72-tree plots. Coconut trees are on 8.5 m spacing.

**Figure 3.** (Experiment 1, KNPK): Concentration of <sup>137</sup>Cs in drinking-coconut meat for a 14-y period after various K treatments.

**Figure 4.** (Experiment 2, CLC): Concentration of <sup>137</sup>Cs in drinking-coconut meat for a 13.5-y period after various K treatments.

**Figure 5.** (Experiment 3, B1, B4 well sites): Concentration of <sup>137</sup>Cs in drinking-coconut meat for a 10.5-y period after various K treatments.

**Figure 6**. Reduction of coconut <sup>137</sup>Cs concentration as a function of the amount of K applied.

**Table 1.** Summary of experiment 1 (KNPK) potassium treatments to Blocks 1 and 2.

Block I (trees1 thru 10) kg ha <sup>-1</sup>					
Row	Feb. 1985, K	Nov. 1987, K	May 1989, K	Nov. 1992, K	Total K
C	2520	0	0	0	2520
F	0	0	0	0	0
I	N-P+2520	0	1260	1260	5040 <sup>a</sup>
M	N-P+1260	1260	1260	1260	5040 <sup>a</sup>
P	N-P	1260	1260	0	2520 <sup>a</sup>
S	1260	1260	1260	0	3780
		Block	II (trees13 thru 22)		
Row	Feb. 1985, K	Nov. 1987, K	kg ha <sup>-1</sup> May 1989, K	Nov. 1992, K	Total K
С	1260	0	1260	0	2520
F	N-P+2520	0	1260	0	3780 <sup>a</sup>
I	N-P	0	0	0	300 <sup>a</sup>
M	0	0	0	0	0
P	2520	0	1260	1260	5040
S	N-P+1260	0	1260	1260	3780 <sup>a</sup>

 $<sup>\</sup>overline{}^{a}$  includes 300 kg K ha<sup>-1</sup> from the contaminated N-P.











