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# Long-Term Reduction in $^{137}\text{Cs}$ Concentration in Food Crops on Coral Atolls Resulting from Potassium Treatment

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# Long –Term Reduction in $^{137}\text{Cs}$ Concentration in Food Crops on Coral Atolls Resulting from Potassium Treatment

## Abstract

Bikini Island was contaminated March 1, 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 ( $^{137}\text{Cs}$ ) transferred from soil to plants that are consumed by residents. Thus, radioecology research efforts have been focused on removing  $^{137}\text{Cs}$  from soil and/or reducing its uptake into vegetation. Most effective was addition of potassium (K) to soil that reduces  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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 2070 kg ha<sup>-1</sup> lead to a  $^{137}\text{Cs}$  concentration in drinking coconut meat that is 34, 22, 10, and about 4 % of original concentration, respectively. Concentration of  $^{137}\text{Cs}$  remains low 8 to 10 y after K is last applied. An explanation for this unexpected result is discussed.

*Keywords:* reduced  $^{137}\text{Cs}$  uptake; K effect; long-term 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, Rongelap, and Utirik 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  $^{137}\text{Cs}$  (Robison et al., 1987, 1994, 1997, 1999). The primary exposure pathway is consumption of locally grown foods that accumulate  $^{137}\text{Cs}$  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. 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  $^{137}\text{Cs}$  from soil and/or reduce its uptake into food crops. Many different remedial measures were evaluated including, soil removal, immobilization of  $^{137}\text{Cs}$  in the soil (use of clays and clinoptilolites), leaching with salt water, repeated cropping, etc. (Robison and Stone, 1998). Most effective and least destructive of all evaluated methods was applying K to the soil surface that subsequently is dissolved by rainfall and absorbed through vegetation roots. Historically fertilizers have not been used at inhabited atolls for most of the large food trees (coconut, *Pandanus*, lime, and breadfruit). Only occasionally are fertilizers used on papaya and banana trees and ornamental flowers that generally grow near the houses.

Naturally low concentrations of K and lack of any significant amount of clay minerals in coral soil is the basis for enhanced uptake of  $^{37}\text{Cs}$  into atoll plants. When additional K is available, trees readily accumulate it with a resulting effective reduction in  $^{137}\text{Cs}$  concentration in plants. This in turn leads to significant reduction of  $^{137}\text{Cs}$  in foods. Early results were very dramatic and  $^{137}\text{Cs}$  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). The range of 1000 to 2000 kg ha<sup>-1</sup> K was selected based on demonstration experiments to evaluate the effectiveness of K treatment in the atoll soil system.

Similar reductions in uptake of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  concentration in coconuts remained at reduced levels for a period of about 2.5 y after the last application of K.

Confirming reduction in  $^{137}\text{Cs}$  concentration and defining the period of time reduction lasts after K is last applied is very important. It provides assurance to returning populations that  $^{137}\text{Cs}$  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

A detailed literature survey was conducted for the 1992 publication of the effect of added K on  $^{137}\text{Cs}$  uptake in plants. The conclusion was that the findings have only limited relevance to Bikini plant-soil systems in view of the very wide ratio between available  $^{137}\text{Cs}$  ( $\sim 10^{-12}$  mol  $\text{kg}^{-1}$  soil) and K ( $\sim 10^{-4}$  mol  $\text{kg}^{-1}$  soil) at Bikini Atoll. Similarly the extensive literature on  $^{137}\text{Cs}$  retention in soils with silicate clays is largely irrelevant to atoll soils where clay minerals are essentially absent and Ca and Mg concentrations are very high. A review of the extensive literature since 1992 on  $^{137}\text{Cs}$ -K interactions relative to  $^{137}\text{Cs}$  uptake by plants (a few examples are, Zhu et al., 2000 a, b; Konoplev et al., 1993; Smolders et al., 1997 a; Roca-Jove and Vallejo-Calzada, 2000) lead to the same general conclusions because of high concentrations of clay minerals, and relatively low concentrations of Ca and organic matter, in many of the soils in these studies compared with the lack of clay minerals and very high concentration of Ca (and organic matter) in atoll soils. Jinzhou et al. (1998) make the point that in calcareous soils they studied  $^{137}\text{Cs}$  retention was largely determined by oxides and silicate clays. These materials are not present in the atoll soil system where  $^{137}\text{Cs}$  is primarily bound with organic matter. Rigol et al. (2002) came to similar conclusions on the importance of clay minerals in controlling  $^{137}\text{Cs}$  adsorption in organic soils. Smolders et al. (1997 b) did find that  $^{137}\text{Cs}$  uptake was reduced in plants grown in solution when  $\text{Ca}^{++}$  and  $\text{Mg}^{++}$  were added to the solution. This is consistent with the relatively low uptake of  $^{137}\text{Cs}$  observed in atoll soils that contain large quantities of  $\text{Ca}^{++}$  and  $\text{Mg}^{++}$ .

Detailed composition, and relative concentration of  $^{137}\text{Cs}$  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 (>30%), some magnesium carbonate, organic matter (as high as 15%), and essentially no clays. The pH ranges from about 7.5 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  $\text{kg}^{-1}$  (Fosberg and Carroll, 1965). Extractable K ranges from 20 to 80 mg  $\text{kg}^{-1}$  in the upper 25 cm of soil and diminishes

rapidly below that depth (Robison and Stone, 1992). The organic matter is contained in the top 40 to 50 cm of the soil column and diminishes rapidly with depth below about 15 to 20 cm. The white coral sand below about 50 cm is devoid of organic matter and has lesser water field capacity than the organic containing top 50 cm of soil.

Additional K applied to the soil surface of the island is readily leached out of the root zone of the island vegetation to the ground water during periods of moderate to high rainfall (Cole et al., 1961; Stone and Robison, 2002). Similar results have been obtained from column leaching studies where a typical atoll soil profile was reconstructed in 20 cm (8 inch) PVC columns. Added K was readily leached from the columns with water quantities similar to observed rainfall (Stone and Robison unpublished data). This is as expected because atoll soils are very porous, have a very high hydraulic conductivity, and even under heavy rainfall conditions water flow is vertical with essentially no lateral flow (Hunt and Peterson, 1980; Peterson and Hunt, 1981; Buddemeier and Oberdorfer, 1997). On Kwajalein Atoll about 30 to 50 % of the annual rainfall recharges the ground water lens that is 3 to 4 m below the ground surface (Hunt and Peterson 1980; Peterson and Hunt 1981). On atolls with lesser rainfall than Kwajalein the recharge is less frequent. For example, on Enjebi Island at Enewetak Atoll the estimated recharge is 0.5 m or about 33 % of the average rainfall (Oberdorfer and Buddemeier, 1988; Buddemeier, 1992; Buddemeier and Oberdorfer, 1997).

Recent experiments with very large plate lysimeters also show the rapid flow to the ground water lens of water added to the soil column when the soil is at field capacity (Robison et al., 2004). Also, 4 wells, with casings slotted from the groundwater surface to about 2 m depth in the ground water lens, were installed at four locations on Bikini Island in 1995. Salinities have been measured at 50 cm intervals to a 2 m depth in the ground water lens repeatedly over a 9 y period. Reduced salinity in the groundwater is very apparent after rainfall of a few inches when the soil is at 50 to 80 % of field capacity, a common condition during 7 or 8 months out of the year (Robison unpublished data). During periods of extended drought, usually December through March, the fresh water layer breaks down and salinity of the entire profile increases. When adequate rain arrives, the fresh water layer is reestablished in the top 0.5 m of the ground water and the entire salinity profile become fresher because of the fresh water recharge.

Furthermore,  $^{137}\text{Cs}$  has been observed in the fresh water component of the groundwater for over 30 y (Noshkin et al., 1977, Robison et al., 1988) and the mean residence time of fresh water in atoll lens is about 5 to 7 y (Oberdorfer and Buddemeier, 1988; Buddemeier, 1992; Buddemeier and Oberdorfer, 1997). Also, a mean residence time of 5.3 y was calculated from data from the slotted wells on Bikini Island during an extended dry period from October 1997 to October 1999 when recharge of the lens never occurred (Robison unpublished data). Thus, there is a continual input of soluble  $^{137}\text{Cs}$  into the lens over the years due to rainfall and subsequent recharge of the lens. Such recharge of the lens will carry along any ions (such as added K) that are soluble in soil water.

The relative uptake of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are reversed in atoll carbonate soils compared with continental, silica soils that have significant quantities of clay minerals (Robison et al., 2000). The median concentration ratios (CR) of  $^{137}\text{Cs}$  in atoll plants ranges from 0.8 to 36 ( $\text{Bq/kg } ^{137}\text{Cs}$  in plants /  $\text{Bq kg } ^{137}\text{Cs}$  in soil) due to lack of clay minerals and binding of  $^{137}\text{Cs}$  with the organic moiety of the soil, whereas, it is relatively low in continental, silica soils (range 0.005 to 0.5). The  $^{90}\text{Sr}$  CR on the other hand is very low in coral soil (0.006 to 1.0) due to the abundance of Ca (and Mg) while in silica soils it is higher (range 0.02 to 3.0). Although the soil system at the atolls is unique, results from the studies at the atolls are applicable at other island groups that consist primarily of calcareous soil and at certain locations on the continents where high concentrations of  $\text{CaCO}_3$  soils are present.

### **3. Methods and Materials**

#### *3.1 Field Experiment Design*

##### *3.1.1 Coconuts*

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

##### *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  $^{137}\text{Cs}$  on Bikini Island, 2.6 to 3.7  $\text{Bq kg}^{-1}$ , 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-month 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). Thus, in 1985 there were six different treatments (Control; 1260 K; 2520 K; N-P; N-P +1260 K; N-P +2520 K) replicated in the two blocks. 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 months from time of initial collection.

In November 1987 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). As a result of these additional applications of K, there are eleven different treatments in the final analysis [the controls in each block are combined as one treatment]. 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  $^{137}\text{Cs}$ . 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  $300 \text{ kg ha}^{-1}$ . Thus, reduction of  $^{137}\text{Cs}$  in coconuts resulted from K in the N-P treatment although the lesser amount of K produced less of a reduction in  $^{137}\text{Cs}$  concentration than did larger quantities of K. These additional quantities ( $300 \text{ kg ha}^{-1}$ ) are included in Table 1.

#### *Experiment 2 (CLC)*

A one ha area adjacent to Experiment 1 (KNPK) was cleared about one 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 ( $76.5 \text{ m} \times 68 \text{ m}$ ). 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  $9 \times 8$ -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  $660 \text{ kg K ha}^{-1}$  (August 1988),  $660 \text{ kg K ha}^{-1}$  (December 1992), and a final  $750 \text{ kg K ha}^{-1}$  (November 1993). All were one-time applications.

#### *Experiment 3 (B1 and B4 well sites)*

Areas for these two sites were cleared in the mid 1980's and some extended clearing occurred in early 1990. They are located about 600 m from each other in the coconut grove. Four interior trees in each  $6 \times 6$ -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  $9044 \text{ kg K ha}^{-1}$  was distributed over the B1 plot and  $4344 \text{ kg K ha}^{-1}$  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.

#### *3.1.2 Pandanus*

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. The site around the tree had been cleared several years before the beginning of the experiment. Samples were collected before fertilizer applications began and during subsequent missions when *Pandanus* fruit were available on the tree.

### 3.2 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 months 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.

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

### 3.3 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. The mean standard deviation on 117 individual  $^{137}\text{Cs}$  measurements is 1.6%. This is similar for all measurements discussed in this paper. 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. The difference in replicate samples was less than 5%; 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.

## 4. Results

### 4.1 Coconut experiment number 1

Results from the eleven final treatments in Experiment 1 are shown in Figure 3. The standard error for the mean  $^{137}\text{Cs}$  value for each collection of samples from trees in each category that had multiple treatments of K ranged from 3.2 to 7.6%. 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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  visible in controls 9 months 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. However, it was not observed at sites that had been cleared and maintained for 2 y prior to treating the coconut trees with K and collecting samples (see Coconut experiment number 3).

One row of trees (row I block 2) that was intended to receive only an N-P treatment actually received 300 kg K ha<sup>-1</sup> from K inadvertently added by the supplier 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. Trees in row P of block 1 (dotted circles in Figure 3) also were treated initially with N-P (and therefore 300 kg ha<sup>-1</sup> of K) but received an additional 1260 kg ha<sup>-1</sup>K at 2.5 y as a result of the partial-root expansion experiment and subsequently a second and third application of 1260 kg K ha<sup>-1</sup>. Concentration of <sup>137</sup>Cs stabilized at about 3% of the pretreatment level in this row as it did in other trees receiving multiple applications. Rows M and S of block I also received 1260K as a result of the partial root expansion.

Samples collected 9 months after K was first applied represented a 3 to 9 months 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, occurred 18 months after first application of K which is about 6 months after the last of 4 initial treatments in 1985. 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 (row C, block 2) 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 the initial <sup>137</sup>Cs concentration) than rows of trees receiving all three treatments (about 3% of the initial <sup>137</sup>Cs concentration). The low <sup>137</sup>Cs concentration in coconuts has persisted for 13 y after the last application of K for the row receiving two treatments (open circles in Figure 3) and for 10 y since the last K treatment for rows receiving three K treatments.

The single row of trees (row C block 1) 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 the initial concentration for 11 y.

#### 4.2 Coconut experiment number 2

Results for experiment 2 are shown in Figure 4. The standard error for the mean <sup>137</sup>Cs value of the 18 trees in the control group for each collection of samples ranged from 3.2 to 7.6%; the range was 3.5 to 7.7% for the 18 fertilized trees. 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 months after K application) with the lowest concentration occurring after about one 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 destruction of highly absorbing surface roots that later reestablish in the surface soil containing the highest concentrations of <sup>137</sup>Cs, and equilibration with the redistributed surface soil associated with clearing of the site. The increase after about 2 y is probably due to less competition for the available <sup>137</sup>Cs as a result of clearing of the very dense vegetation and large *Scaevola* and *Messerschmidia* trees. It stabilized about 2 y after clearing. The gradual decline thereafter in <sup>137</sup>Cs concentration in control trees is due to radiological decay of <sup>137</sup>Cs ( $T_{1/2} = 30.1$  y) and loss of <sup>137</sup>Cs from soil when rainfall is adequate to saturate the soil and cause recharge of the ground-water lens. Loss of <sup>137</sup>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. 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  $^{137}\text{Cs}$  from soil to the ground-water lens as a result of trees preferentially accumulating K (and not  $^{137}\text{Cs}$ ) thereby leading to a greater loss of  $^{137}\text{Cs}$  when rainfall is adequate to produce recharge of the groundwater lens. That part of the experiment is not reported here but results do provide additional data on reduction of  $^{137}\text{Cs}$  in coconuts as a result of added K and duration of reduction after last application of K.

Rapid decline in  $^{137}\text{Cs}$  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. 4.4

#### 4.4 *Pandanus*

One *Pandanus* tree shows the same significant reduction in  $^{137}\text{Cs}$  concentration as a result of K treatment (Figure 6) as do coconuts and has maintained this low concentration for 5.5 y since K was last applied.

#### 4.5 Fractional $^{137}\text{Cs}$ reduction as a function of total applied K

Fractional reduction in the original  $^{137}\text{Cs}$  concentration in coconut trees as a function of total applied K is shown in Figure 7. 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 2070 kg K ha<sup>-1</sup> lead to a  $^{137}\text{Cs}$  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 is no further decrease and all results are between 3 and 5 % of the original concentration.

## 5. 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 300 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 330 kg K ha<sup>-1</sup> was not adequate to supply all of the K that trees are capable of accumulating. Treatment with 1000, and 2000 kg ha<sup>-1</sup> reduced the  $^{137}\text{Cs}$  concentration to about 3% of pretreatment levels. Thus, after the first application of 1000 to 2000 kg ha<sup>-1</sup> of K, the  $^{137}\text{Cs}$  concentration ratio (C.R.) decreased in one year from the

initial value of about 30 to about 3. Concentration of  $^{137}\text{Cs}$  in soil is unchanged over this time period. With additional K treatment the C.R. for  $^{137}\text{Cs}$  further declined to about 1.5; again with insignificant change in  $^{137}\text{Cs}$  concentration in soil. Similar results are observed for coconut experiment 2.

The most important feature of the very significant reduction in  $^{137}\text{Cs}$  concentration in food crops as a result of K treatment is the long period of time that  $^{137}\text{Cs}$  remains at these low levels after K was last applied. This long-term reduction of  $^{137}\text{Cs}$  was unexpected but important because it will greatly reduce the radiation dose people will receive upon resettling the atoll. 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 ground-water 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  $^{137}\text{Cs}$  concentrations obtained as a result of K treatment still persist.

Similar results have been obtained from coconut trees on Enjebi Island at Enewetak Atoll where the  $^{137}\text{Cs}$  concentration in drinking coconuts has remained at a very low level for 6 y since the last treatment with potassium.

Mechanisms by which plants might control uptake of  $^{137}\text{Cs}$  and K to produce rapid and significant decline after K treatment is of interest. The most common explanation is that the suppressive effect is due to competition between  $^{137}\text{Cs}$  and K in the soil solution; that is, the  $^{137}\text{Cs}/\text{K}$  ratio in soil water determines transfer by roots and trees. However, natural, exchangeable K concentration in the top 10 to 15 cm of soil at Bikini ranges from  $2.6 \times 10^{-4}$  to  $10^{-3}$  mol  $\text{kg}^{-1}$  and  $^{137}\text{Cs}$  from  $10^{-12}$  to  $10^{-11}$  mol  $\text{kg}^{-1}$ , a difference of about 8 orders of magnitude ( $^{137}\text{Cs}/\text{K} = 10^{-8}$ ). Applying 1,000  $\text{kg ha}^{-1}$  of K distributed to a depth of 20 cm adds about  $10^{-2}$  mol  $\text{kg}^{-1}$  of K. This K addition is very large relative to the normal K concentrations but it only changes the 8 order difference in the  $^{137}\text{Cs}/\text{K}$  ratio by about an order of magnitude (to  $10^{-9}$ ). This is insignificant relative to the initial  $^{137}\text{Cs}/\text{K}$  ratio. A mechanism whereby such a change would drastically increase the competitive ability in soil is not yet apparent.

Elevated concentration of internal tree-K can prevent uptake and influx of  $^{137}\text{Cs}$  into plants as described by Zhu et al. (2000 a and b). Reduced  $^{137}\text{Cs}$  concentration in coconuts and annual crops (corn and

sorghum) at the atolls, as a result of treating only a portion of the root system with K, are similar to reductions when the entire root system is treated with K. When various methods were used to treat different portions of the root system with K,  $^{137}\text{Cs}$  uptake was determined not by the proportion of the untreated root system but by the amount of K absorbed by the plant (Robison and Stone, 1992; Stone and Robison, 2002, experiments 2, 4, 6 and summary). These results support the hypothesis that in soils lacking silica clays a critical factor regulating plant uptake and influx of  $^{137}\text{Cs}$  is the concentration of K in the plant rather than the proportion of the root system in K-treated soil.

Thus, an explanation for the long duration of the reduced  $^{137}\text{Cs}$  concentrations in the plants comes from analysis of K in entire coconut trees that were sacrificed 1 to 8 y (most in the 5 to 8 y range) after the last application of K. Results indicate that when additional K is available trees accumulate large quantities of K. Amounts of K in entire fertilized coconut trees (20 trees) are greater by a factor of 4.8 than amounts of K in entire unfertilized trees (10 trees),  $3.9 \text{ g kg}^{-1}$  vs.  $0.65 \text{ g kg}^{-1}$ , respectively. The K ratio between fertilized and unfertilized trees in the two major storage compartments, trunk and fronds is 5.6 and 5.3, respectively (Robison and Stone unpublished data). This high internal concentration of K can alter the uptake of  $^{137}\text{Cs}$  from the soil and be transported to fruits for many years. These large-scale field tests support the hypothesis that uptake of  $^{137}\text{Cs}$  from soil is ultimately regulated more by internal K content than the  $^{137}\text{Cs}/\text{K}$  ratio in soil that is important initially.

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  $^{137}\text{Cs}$  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.

The field capacity of the coral soil is between 20 and 25 % [v/v] (Stone and Robison unpublished data). Other than during the driest months (December through April) the soil ranges between 50 and 80 % of field capacity. Consequently a field capacity of 20 % would require a rainfall of between 4 cm (1.6 inches) and 10 cm (3.9 inches) to create recharge of the lens and transport applied K out of the root zone to the ground water. For a field capacity of 25 % the corresponding numbers would be 5 cm (2 inches) to 12.5 cm (4.9 inches). Thus, because of the high porosity of atoll coral soil as discussed in the “Background” section of the paper, and the standing field capacity of the soil during these experiments,



significant rainfall after application of K to the soil surface can alter the “effective” amount of K available to the plants.

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, row C, block II). 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 inches), 30.5 cm (12 inches), and 35.7 cm (14 inches), respectively. Consequently, there was more than adequate rainfall to leach an unknown, but 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 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 inches) in February, 3.4 cm (1.3 inches) in March, and 6.6 cm (2.6 inches) 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, for the 3 following increments of applied K rainfall played a role. The second increment of K applied in May 1985 was followed by unusually high rainfall of 18.2 cm (7.15 inches) in May, 11.5 cm (4.5 inches) in June, and 8 cm (3.1 inches) in July. After the third increment of K was applied in August, there was 19.5 cm (7.8 inches) rainfall over the rest of the month, 20 cm (7.9 inches) in September and 17.3 cm (6.8 inches) in October. After the November 1985 application of K there was 10.8 cm (4.3 inches) and 10.9 cm (4.3 inches) in November and December, respectively. Thus, rainfall after the May, August, and November applications of K was adequate to cause transport 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 for the May, August, and November applications 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.

In addition to coconut 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 at Bikini Island 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. Based on the experiments to date, K treatment would not be required for 8 to 10 y, if one wanted to wait that long, to maintain the low

concentration of  $^{137}\text{Cs}$  in fruit trees. This certainly reduces concerns of resettling populations about applying K (or forgetting to apply K) on an annual or biannual basis. Not having to apply K annually (or biennially) significantly reduces the 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. Applying K vastly improves growth and productivity of annual plants growing in nutrient deficient coral soils and concurrently reduces  $^{137}\text{Cs}$  in a manner similar to that observed in trees.

## 7.0 Acknowledgements

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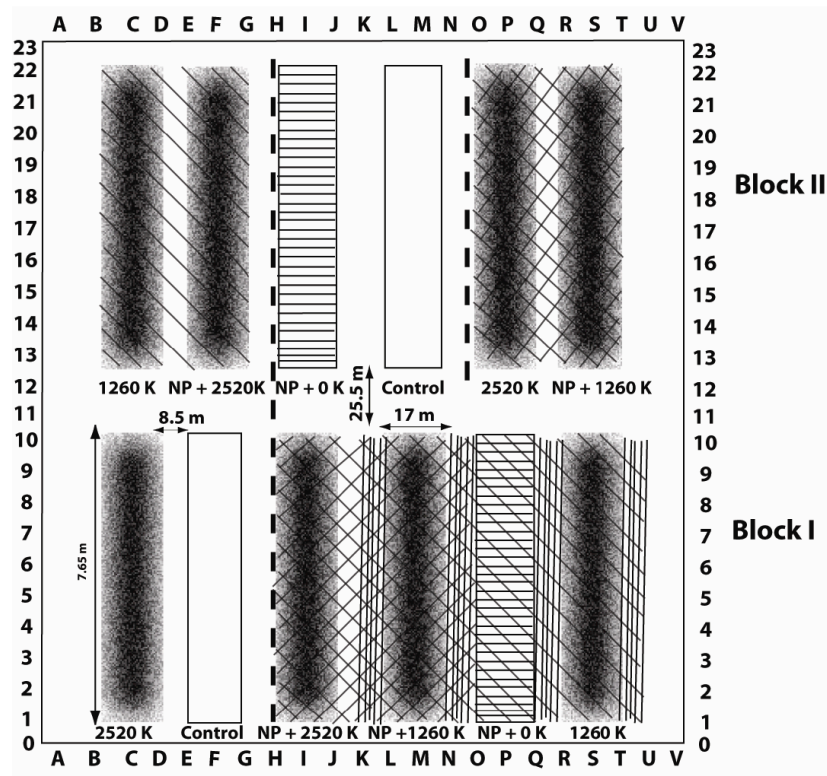
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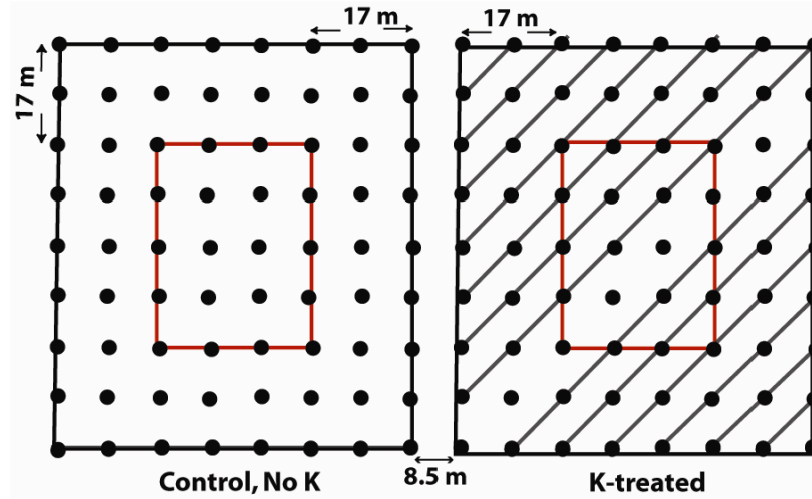
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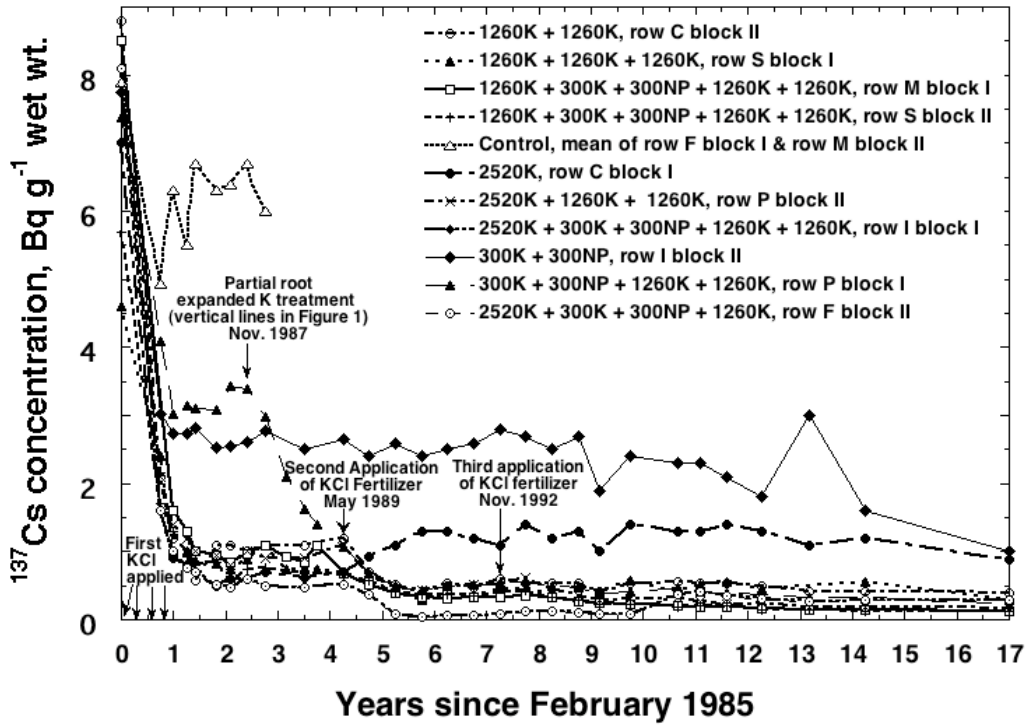
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**Figure 1.** Experiment 1 (KNPK) is a 2-Block design 22 rows x 24 rows. Numbers and letters at the front end of rows indicate initial treatment in February 1985. Each rectangle is 3 trees wide (represented by 3 alphabetic letters) and 10 trees long (1 to 10 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 I 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  $\text{kg ha}^{-1}$ , are shown at the start of each row. Vertical lines represent an expansion of K treatment ( $1260 \text{ kg ha}^{-1}$ ) in November of 1987. Diagonal lines running from upper left to lower right represent an additional application of  $1260 \text{ kg ha}^{-1}$  of K in May 1989. Diagonal lines running from lower left to upper right, that form cross-hatching with the 1987 treatment, represent an application of  $1260 \text{ kg ha}^{-1}$  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 17 y period after the start of various K treatments.



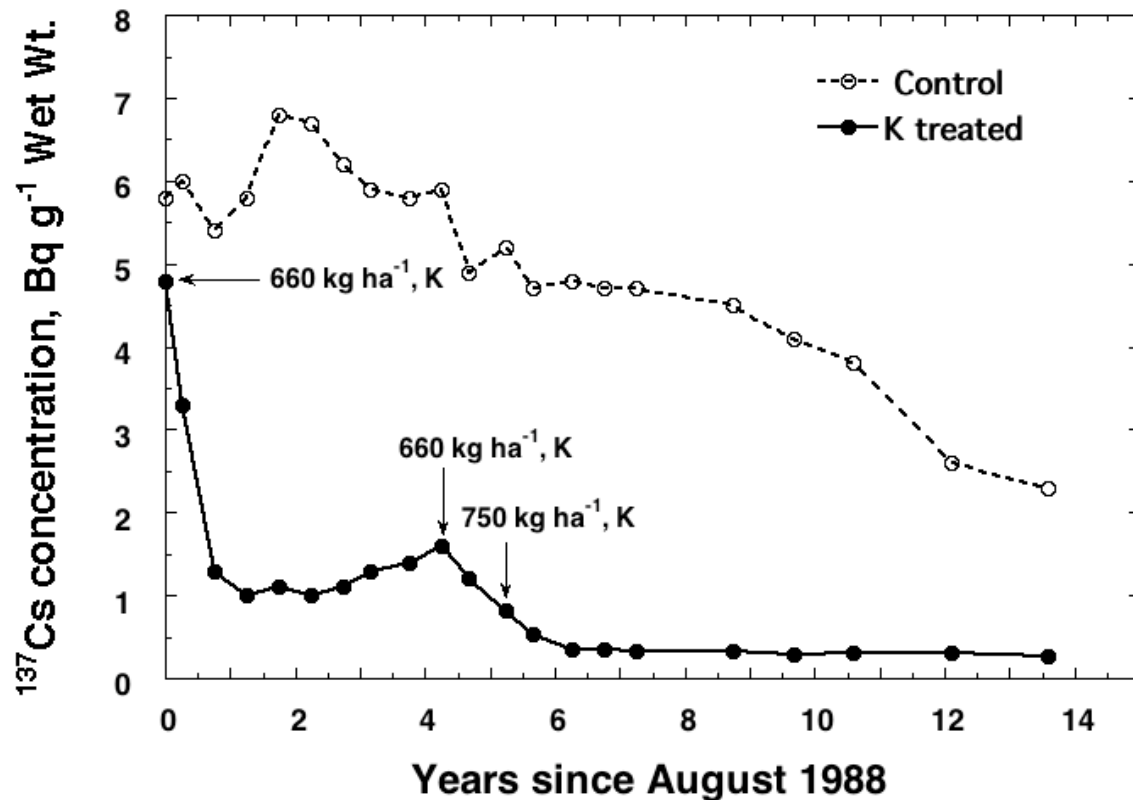
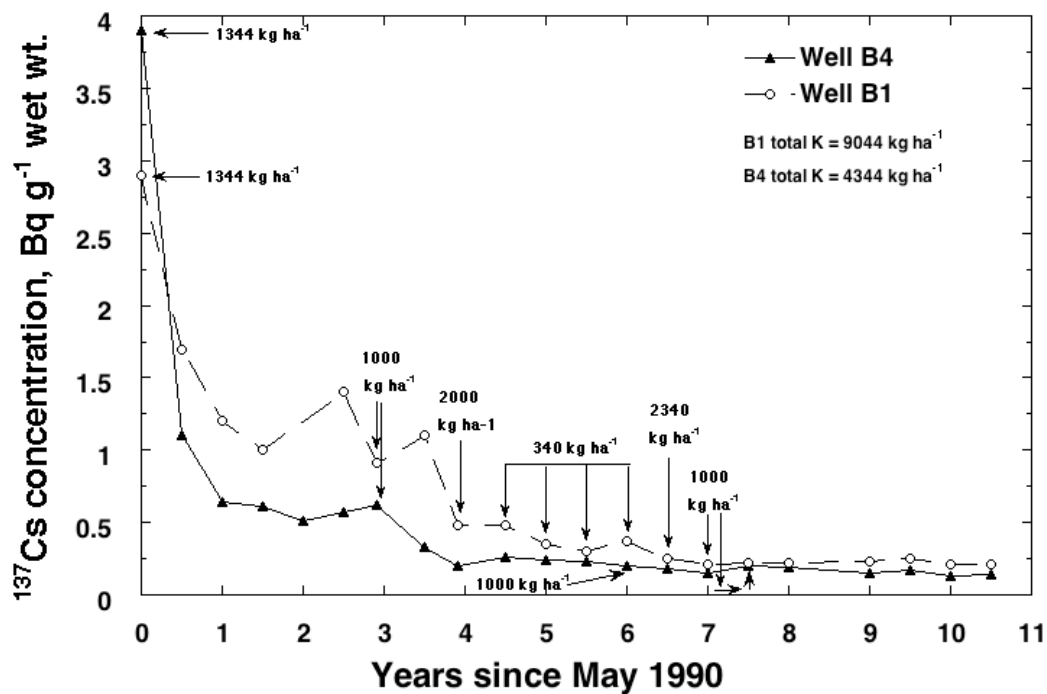
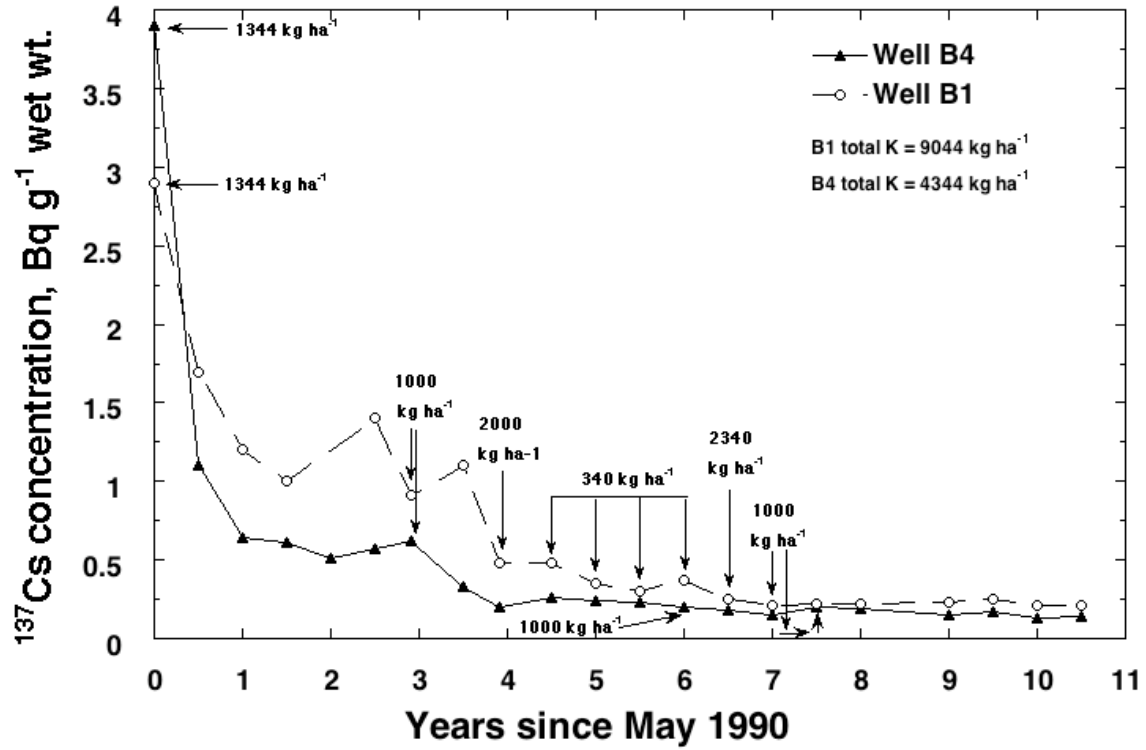
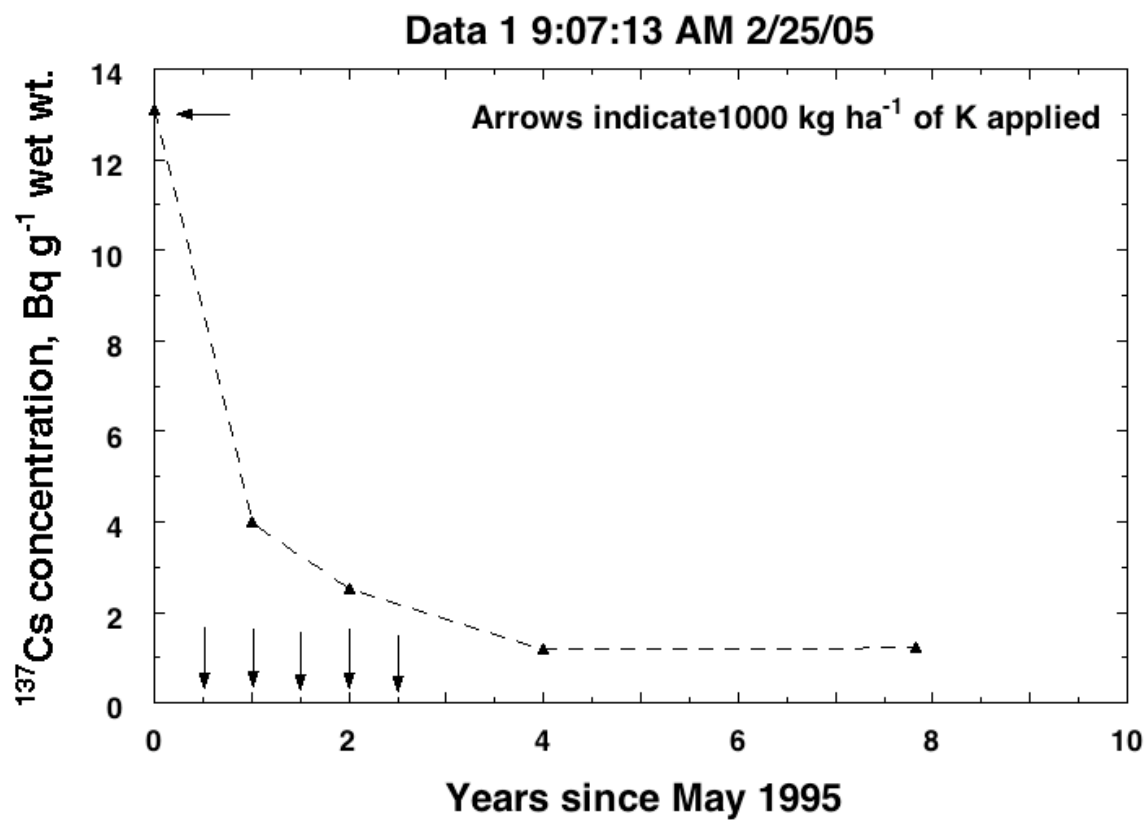


Figure 4. (Experiment 2, CLC): Concentration of  $^{137}\text{Cs}$  in drinking-coconut meat for a 13.5-y period after various K treatments.

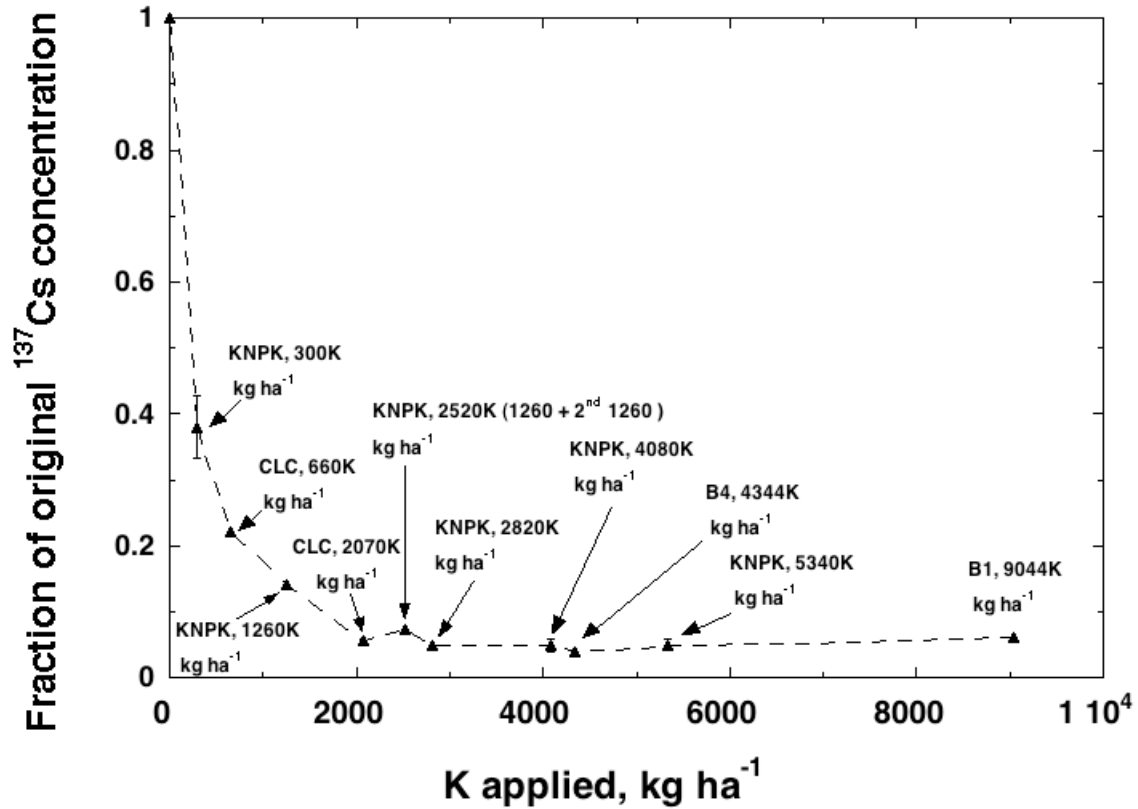




**Figure 5.** (Experiment 3, B1, B4 well sites): Concentration of  $^{137}\text{Cs}$  in drinking-coconut meat for a 10.5 y period after the start of K treatments.



**Figure 6.** (Experiment 4, *Pandanus*): Concentration of  $^{137}\text{Cs}$  in *Pandanus* for an 5.5 y period since the start of K treatments.



**Figure 7.** Reduction of coconut <sup>137</sup>Cs concentration as a function of the amount of K applied. Standard deviations are included where more than 1 row of trees received the same amount of total K even though the time distribution of K may have differed [KNPK, 300 K application (2 rows), KNPk, 4080 K application (2 rows), and KNPk, 5340 K application (2 rows)].

The mean of the last 5 years of data (11.6, 12.1, 13.1, 14, and 17 years) after the start of the experiment were used for comparison to the initial concentration (prior to treatment with K) to calculate the final reduction for the 4080 and 5340 K applications. Data points from year 1 through year 2.8 (7 data points), prior to the partial root expansion application of K, were used for the 300 K application.

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

<b>Block I (trees 1 through 10)</b>					
<b>kg ha<sup>-1</sup></b>					
<b>Row</b>	<b>Feb. 1985</b>	<b>Nov. 1987</b>	<b>May 1989</b>	<b>Nov. 1992</b>	<b>Total K</b>
C	2520K	0	0	0	2520
F	0	0	0	0	0
I	2520K +300K + 300NP	0	1260K	1260K	5040 <sup>a</sup>
M	1260K +300K +300NP	Expanded (1260K)	1260K	1260K	4080 <sup>a</sup>
P	300NP		1260K	0	1560 <sup>a</sup>
S	1260K	Expanded (1260K)	1260K	0	2520
<b>Block II (trees 13 through 22)</b>					
<b>kg ha<sup>-1</sup></b>					
<b>Row</b>	<b>Feb. 1985, K</b>	<b>Nov. 1987, K</b>	<b>May 1989, K</b>	<b>Nov. 1992, K</b>	<b>Total K</b>
C	1260K	0	1260K	0	2520
F	2520K + 300K + 300NP	0	1260K	0	4080 <sup>a</sup>
I	300NP	0	0	0	300 <sup>a</sup>
M	0	0	0	0	0
P	2520K	0	1260K	1260K	5040
S	1260K + 300K +300NP	0	1260K	1260K	4080 <sup>a</sup>

<sup>a</sup> includes 300 kg K ha<sup>-1</sup> from the contaminated NP.

