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Distribution and Ratios of ^{137}Cs and K in Control and K-treated Coconut Trees at Bikini Island where Nuclear Test Fallout Occurred: Effects and Implications

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Abstract
Coconut trees growing on atolls of the Bikini Islands are on the margin of K deficiency because the concentration of exchangeable K in coral soil is very low ranging from only 20 to 80 mg kg^{-1}. When provided with additional K, coconut trees absorb large quantities of K and this uptake of K significantly alters the patterns of distribution of ^{137}Cs within the plant. Following a single K fertilization event, mean total K in trunks of K-treated trees is 5.6 times greater than in trunks of control trees. In contrast, ^{137}Cs concentration in trunks of K-treated and control trees is statistically the same while ^{137}Cs is significantly lower in edible fruits of K treated trees. Within one year after fertilization (one rainy season), K concentration in soil is back to naturally, low concentrations, however, the tissue concentrations of K in treated trees stays very high internally in the trees for years while ^{137}Cs concentration in treated trees remains very low in all tree compartments except for the trunk. Potassium fertilization did not change soil Cs availability.

Mass balance calculations suggest that the fertilization event increased above ground plant K content by at least a factor of 5 or 2.2 kg. Potassium concentrations and content were higher in all organs of K fertilized trees with the greatest increases seen in organs that receive a portion of tissue K through xylem transport (trunk, fronds and fruit husks) and lowest in organs supplied predominantly with K via the phloem (palm heart, spathe, coco meat and fluid. ^{137}Cesium concentrations and contents were dramatically lower in all organs of K treated trees with greatest proportional reductions observed in organs supplied predominantly with K via the phloem (palm heart, spathe, coco meat and fluid).

All trees remobilize both K and ^{137}Cs from fronds as they proceed toward senescence. In control trees the reduction in concentration of K and ^{137}Cs in fronds as they age is logarithmic but K remobilization is linear in K-treated trees where K concentration is high. As a result of K treatment the ^{137}Cs concentration in K-treated fronds is extremely low and constant with frond age. Fronds of K treated trees contain a greater amount of K than control tree fronds. As they fall to the ground and decay they provide a small continuing pool of K that is about 3% of the natural K in soil under the tree canopy.

Results of K and ^{137}Cs concentration and distribution in control and K-treated coconut trees suggest that the application of K reduces ^{137}Cs uptake both in the short term immediately following K fertilization and in the long term, after soil K levels have returned to normal but while plant K stores remain high. These results suggests that high internal K concentration and not high soil K is primarily responsible for long-term reduction of ^{137}Cs in edible fruits, and plays a significant role in limiting further uptake of ^{137}Cs by roots, and affects allocation of ^{137}Cs to edible fruits for years. Coconut trees are capable of luxury K accumulation when provided with excess K and in this example the additional K can effectively provide the K requirements of the plant for in excess of 10 years. The reduction of ^{137}Cs uptake lasts for at least 10 y after K is last applied and greatly reduces the estimated radiation dose to people consuming local tree foods. Effectiveness and duration of K treatment provides important assurances that reduction in ^{137}Cs is long term and the radiation dose from consuming local plant foods will remain low.

Key words: Trees; Potassium transport; ^{137}Cs transport; uptake; compartment-distribution; internal-K control; K channel; K transporters

Introduction

The United States conducted two nuclear tests at Bikini Atoll in 1946 (Crossroads series: tests Able and Baker). Testing resumed at Bikini Atoll in 1954 with the Castle series of tests, then the Redwing series in 1956, and concluded with the Hardtack I series in 1958. The primary source of contamination on Bikini Island occurred on March 1, 1954 from test Bravo that was part of the Castle series. Remaining radionuclides currently on the atoll are ^{137}Cs, ^{90}Sr, $^{239+240}$Pu, and ^{241}Am (Robison et al., 1982, 1997). About 90% of the estimated dose to people returning to live on Bikini Island comes from uptake of this aged source of ^{137}Cs by terrestrial food crops, mainly coconuts (*Cocos nucifera L.*), *Pandanus,* breadfruit, and some papaya and banana (Robison et al., 1997). Thus, radioecology studies were implemented to try to find ways to either remove or immobilize ^{137}Cs in the soil or block its uptake by terrestrial plants (Robison and Stone, 1998). Of all the various methods tested, by far the most effective technique was application of potassium (K) on the soil surface (Robison and Stone, 1992, 1998; Robison et al., 2006). Added K was subsequently dissolved by rainfall and accumulated in terrestrial plants growing on the island in competition with residual ^{137}Cs fallout contamination.

Concentration of ^{137}Cs in drinking-coconut meat is reduced to about 3 to 5 % of pretreatment concentrations when about 1000 to 2000 kg ha^{-1} of K is applied and the ^{137}Cs concentration remains at these very low concentrations for periods of up to 10 y after the last application of K (Robison et al., 2006). The mechanism of how coconut trees sustain this effect for such a long period of time even after a single application of potassium was largely unknown, and led to an interest in how much K was accumulated in the coconut trees after K treatment, how it was distributed relative to ^{137}Cs among the various parts of the tree, and how it might differ from the distribution in trees not treated with K.

A common explanation of the suppressive effect of K on uptake of ^{137}Cs is the competition between ^{137}Cs and K in the soil solution; that is, the ^{137}Cs /K ratio in soil water determines transfer K and ^{137}Cs by roots into the trees. However, natural, exchangeable K concentration in the top 10 to 15 cm of soil at Bikini ranges from 2.6 x 10 $^{-4}$ to 10^{-3} mol kg^{-1} and the ^{137}Cs concentration from 10^{-12} to 10^{-11} mol kg^{-1}, a difference of about 8 orders of magnitude (^{137}Cs/K ~ 10^{-8}). Applying 1.000 to 2,000 kg ha^{-1} of K distributed to a depth of 20 cm adds about 10^{-2} mol kg^{-1} of K. This K addition is large relative to the normal K concentrations in coral soil but it only changes the 8 order of magnitude difference in the ^{137}Cs /K ratio by about an order of magnitude (~ to 10^{-9}). This is insignificant relative to the initial ^{137}Cs /K ratio. A mechanism whereby such a slight change would drastically increase the competitive ability in soil is not yet apparent.

Other processes may be more relevant in the atoll ecosystem. In seminal experiments Epstein et al., (1963) demonstrated that cation (K, Rb, Cs) transport in plant cells consisted of a high affinity component (mechanism 1) and a low affinity component (mechanism 2). High affinity mechanisms operate primarily at low concentrations of K in solution while low affinity mechanisms operate at high concentrations of K in solution. Thus, the concentration of K in soil solution greatly impacts the mode of K and Cs uptake by plant root cells. Since that time much more detail on the physiological and molecular mechanisms involved in these two transport processes has been elucidated (Schachtman et al., 1992; Schachtman and Schroeder, 1994; Kim et al. 1998; Ashely et al., 2006; Grabov, 2007; Qi et al., 2008). The mechanisms by which Cs is taken up by plants has been reviewed by White and Broadley (2000), who suggested based upon theoretical evidence, that the voltage insensitive cation channels (VICCs) are likely the most dominant mechanisms of Cs transport. Cesium transport has now been demonstrated to occur through high affinity transport processes including KUP/HAK family (Maser et al, 2001), as well as inward rectifying (KIR) outward rectifying (KOR) and voltage insensitive channels (White and Broadley (2000).

Potassium fertilization may also directly impact Cs uptake by down-regulating the activity of K uptake process in the root membrane however this has received scant attention in perennial species. In pistachio, substantial amounts of N and K can be stored in perennial tree organs when fertilization exceeds demand

for leaf and nut production (Rosecrance et al; 1996, 1998). This luxury consumption has been shown to subsequently depress uptake of these nutrients from the soil (Youssefi et al, 2000; Rosecrance et al; 1996, 1998; Burns, 1992).

2.0 Background

2.1 Soil composition and properties

The effectiveness of K application in reducing ^{137}Cs uptake in terrestrial plants results from the very low concentration of K in atoll coral soils and lack of any significant amount of clay minerals. Detailed elemental composition of coral soil is discussed in Robison and Stone, 1992. 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. Concentration of naturally occurring total K is only 300 mg kg^{-1} at coral atolls (Fosberg and Carroll, 1965). Extractable K ranges from 20 to 80 mg kg^{-1} in the upper 30 cm of soil and diminishes rapidly below that depth (Robison and Stone, 1992; unpublished data). Organic matter is contained in the top 40 to 50 cm of soil and diminishes rapidly with depth below about 20 to 25 cm. Most all K and ^{137}Cs in the soil profile is contained in the top 40 cm of soil. The white coral sand below about 40 cm is devoid of organic matter, and has far lesser water holding capacity, and extremely lower concentrations of K and ^{137}Cs than the organic containing topsoil. Absorbing tree roots are primarily in the top 40 to 50cm of soil containing the organic matter. A few roots do extend downward into the white sand below the organic zone but they are relatively few very limited branching and insignificant root hair formationand do not reach the groundwater.

2.2 Rainfall and groundwater lens recharge

Potassium applied to the soil surface is readily leached out of the root zone of island vegetation to the ground water during periods of moderate to high rainfall (Cole et al., 1961; Stone and Robison, 2002). 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 installed below the organic zone also show the rapid flow to the ground water lens of water added to the soil column when the soil is near 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 repeatedly over a 9 y period at 50 cm intervals to a 2 m depth in the ground water lens. 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. 5m of the ground water and the entire salinity profile become fresher because of fresh water re-charge. Furthermore, ^{137}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 fresh water measurements made in the slotted wells on Bikini Island during an extended dry period from October 30, 1997 to April 1999 when recharge of the lens never occurred (Robison unpublished data). Thus, as a result of rainfall, there is a continual annual input of fresh water into the lens that contains ^{137}Cs and K that is in excess of the binding capacity of the soil organic matter.

3.0 Methods and materials

2.2 Field Methods

The four experimental sites from which these coconut trees were taken are located along the midline of Bikini Island where the ^{137}Cs concentration and organic matter in soil are highest. Two these sites are several hectares in area. Eighteen mature coconut trees 30 to 35 y in age that had been treated with K between 1985-1992 were selected from sites where trees received between 2000 and 2500 kg K per hectare in one application or in two or three split applications. They were harvested about 7 years after the last application of K. Ten control trees where additional K was unavailable (not treated with K) were selected from areas near the K experimental sites. Fronds were the first part of the trees to be collected. The sequence was: the number one frond (the youngest developed frond on the tree) growing just below the spike (the early inflorescence or flower sessile) and spathe (from which the spike arises and that shields the inflorescence), followed by frond numbers 5, 10, 15, 20, 25, 30, 35, 40, each one older than the previous one. Some trees had only enough fronds to collect frond 30 while others would go to frond 35 or 40. Fronds 35 and 40, or the last frond collected even if it were frond 30, were the driest and most senescent and were ready to fall to the ground. The mass of each frond not collected for analysis, fronds 2-4, 6-9, 11-14, etc., was determined so total radionuclide and K inventories could be developed by interpolating K and ^{137}Cs concentrations between the collected fronds that were analyzed for K and ^{137}Cs. Frond leaves were cut from the frond rib and double bagged. The large frond rib was cut into small sections and double bagged.

Next, all coconuts on the trees were collected. These included the younger developmental stage called 'drinking-coconuts' and the older stage referred to as 'copra-nuts'. Husks of each coconut were removed from around the shell and were double bagged. Coconut fluid was drained from the drinking-coconuts through an "eye" in the top of the shell into polypropylene bottles. Drinking-coconut meat that is of gelatin consistency was left in the shell and double bagged. Copra-nut shells that contain the thick, hardened, white meat were double bagged.

At this stage the tree was felled using a chain saw to cut it at the ground surface. The entire heart of palm was collected, along with the spike and spathe. The very long trunk was cut into several sections. The total mass of each section of trunk was measured. Trunk sections were very large and heavy and consequently were quartered to obtain a representative sample. Quartered trunk sections, along with the spathe, spike, fronds, frond rib, drinking-coconuts, drinking-coconut fluid, and copra-nuts were placed in large freezer vans where they were frozen on the day of collection. At the end of a mission, the freezer-vans were returned to Lawrence Livermore National Laboratory (LLNL) for subsequent processing and analysis of the samples. The median mass of the collected K-treated mature coconut trees was 969 kg wet weight and of control trees was 852 kg wet weight. Two K-treated trees that were fertilized from the time of planting to their age of about 9 y had wet weight masses of 235 kg and 416 kg.

2.3 Laboratory procedures

At LLNL drinking-coconut meat and copra-meat shells were cracked, the meat extracted and refrozen if any thawing occurred, dried to constant weight by lyophilization, and ground to fine consistency in a Waring blender. Drinking-coconut fluid was reduced by evaporation. The spathe, spike, fronds, ribs, coconut husks and shells, and large quartered sections of coconut tree trunks were oven dried at low temperature to constant weight. They were then cut into small pieces and ground to a fine consistency in a Willey mill. Dried samples and the reduced fluid were packed into 8.0-cm-diameter x 4.6-cm-high aluminum cans for gamma spectrometry analysis. Separate aliquots of the samples were saved in plastic vials for K analysis.

Analyses for ^{137}Cs were performed in the LLNL Gamma Spectroscopy Facility [GSF] (Hamilton et al., 2000) that consists of 22 high-resolution, intrinsic, germanium gamma detectors. Potassium concentrations were determined in our analytical laboratory (AL) using an ASOMA x-ray fluorescence system and an external calibration curve.

Standards and duplicate samples, blind to the analyst and each totaling 10% of the samples submitted to the GSF for ^{137}Cs analysis and the AL for K analysis, were included in each batch of 50 to 100 samples

sent to the facilities. 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 was not necessary to reanalyze any samples from these experiments.

4.0 Results and Discussion

Total mass of harvested mature coconut trees ranged from 580 kg to 1360 kg wet weight for twenty K-treated coconut trees with a median value of 969 kg and a mean and standard error of 974 ± 48. Mass of control trees ranged from 620 kg to 1024 wet weight with a median of 852 kg and a mean and standard error 816 kg ± 122. Thus, there is no statistical difference in the total mass of the K-treated and control trees at one standard error. Mean percent of the total mass of the tree in each compartment for both K-treated and control trees is listed in Table 1. Trunk (including the palm heart) and frond compartments of the trees account for about 56 % and 27% of total mass, respectively, for a total of 83 % of total tree mass. There is no statistical difference between compartment masses of the K-treated and control trees as each compartment is within one standard error of each other.

Moisture content of the coconut tree trunks (including the palm heart) and fronds of 28 trees is given in Table 2 as mean dry/wet ratios and standard errors for various sections of coconut tree trunks. Moisture content is lowest in the first 1 m of the trunk and is highest in the palm heart that is at the very top of the tree trunk. The palm heart is soft and in fact edible. The mean dry/wet ratio ranges from 0.25 to 0.32 for trunk sections. The palm heart is 0.18. The mean dry/wet ratio for all coconut fronds on all the trees ranged from 0.25 to 0.36. Dry/wet ratios for other parts of coconut trees that account for only 17% of the tree mass are: drinking coconut husks (0.14 - 0.20), drinking coconut shells (0.55 – 0.70), drinking coconut meat (0.15 - 0.40), copra meat (0.45 – 0.70), and copra husks (0.26 -0.34).

All plants growing on the atolls are on the margin of K deficiency where exchangeable K ranges from only 20 to 80 mg kg^{-1} in coral soil (Stone and Robison, 2002), and various food crops and flowers will not grow in coral soil without addition of K. Coconut trees absorb large quantities of the available K when K is supplied to the ground surface and dissolved by rainfall. A comparison of K in control and K-treated trees is shown in Figure 1. More detail on the relative distribution of K in various tree compartments is given in Table 3. A significant result is that mean K concentration in trunks of K-treated trees is 5.6 times greater than that of control trees. The total K in K-treated trees is also more than a factor of 5 greater than that of control trees This large reservoir of K in tree trunks is very important and provides the source of K that leads to long-lasting reduction of ^{137}Cs in edible fruits at the atolls 10 years or more after the last application of K (Robison et al., 2006). The final data collected for this study were taken 10 y after K was last applied. There are no immediate plans to continue to sample experimental plots on Bikini Island but such data would be valuable in helping determine the total duration of any effects and providing recommendations to resettled populations on the need to perform additional applications of K.

When additional K is available there is a significant reduction in concentration of ^{137}Cs in most coconut tree compartments. *Pandanus* fruit trees treated with K show all the same effects and results as the coconut trees (Robison et al., 2006). Mean concentration ^{137}Cs (kBq kg^{-1}) in entire coconut trees, i.e. all tree compartments, for twenty K-treated coconut trees is significantly reduced relative to ten control trees not treated with K as shown in Figure 1. Total ^{137}Cs in entire K-treated trees is about 41% of the concentration in entire control trees. Concurrently, K-treatment greatly increases the amount of K in all compartments of a tree and K concentration in entire K-treated trees is greater by a about a factor of 5 compared with entire control trees. Thus, on coral atolls, trees absorb large amounts of K when additional K is made available and that intake of K greatly reduces the amount of ^{137}Cs in K-treated trees compared to that in untreated trees. Analysis of soil prior to and subsequent to K^+ fertilization demonstrated that fertilization had no significant effect on ^{137}Cs concentrations in soils. Data in Table 4 provide a more detailed look at ^{137}Cs concentration in various compartments in both control and K-treated trees. Most significant, and especially important, is the large reduction in ^{137}Cs concentration in fruits (to 5 to 10 % of pretreatment levels) as a result K-treatment because it greatly reduces radiation dose to people eating local tree food crops. It also shows that K does not have to be re-supplied to the trees for at least 10 y and thus gives people assurance that the radiation doses from consuming the coconut meat and fluid will stay very

low for at least 10 y. This also greatly reduces the cost of remediation versus typical agriculture practice of annual or biennial applications of K. The same kind of reduction is seen in drinking-coconut fluid (also consumed by local residents) and the spike. Reduction in ^{137}Cs concentration in the spathe, heart, and husk + shell is slightly less than in coconut meat and fluid.

In K-treated trees about 60 % of K in a frond is retrieved by the tree (Figure 2) prior to its falling to the ground. Retrieval of K from fronds is linear as a function of frond age when a large amount of K has been stored in the tree. This reduction in K as fronds age is not the result of mass changes. Frond mass, on a dry weight basis, is constant from about frond 2 or 3 to frond 30 to 40 depending on the tree. This is demonstrated in Table 5 where frond mass (dry weight) for each frond on a tree is divided by the mass of frond 1 to normalize frond data for each tree for statistical analysis. The mean value for all control trees and all K-treated trees is given along with the associated standard error. Fronds of an individual tree all have the same dry mass within one standard error. The higher concentration of K in senescent fronds of K-treated trees does lead to a very small continuing annual source of K as they fall to the ground and decay. This annual pool of K is about 3% of the natural K in soil around the tree that is within the diameter of the tree canopy.

The same process operates in control trees as well (Figure 3) but concentration of K in control-tree fronds is much less than in fronds of K-treated trees. Retrieval of K from aging fronds is logarithmic when concentration of K is very low as in the case of unfertilized trees. The logarithmic curve can be interpreted as a two-component curve as shown. The first component of the curve represents the very young fronds where K is more readily retrieved. The second component of the curve represents a compartment of K in older fronds where K is retrieved less readily than from the pool of young fronds. This two-compartment structure is not observed when large quantities of K are available to the plants as seen in Figure 2.

In trunks, there is small but statistically non-significant difference between ^{137}Cs concentration of control trees and K-treated trees and the large trunk compartment accounts for about 54 % of the ^{137}Cs in a tree. Thus, while ^{137}Cs concentration in edible fruits is reduced to 5 to 10 % of pretreatment concentration, ^{137}Cs concentration in the entire tree is reduced by only about 40 % because the concentration of ^{137}Cs (and also total amount of ^{137}Cs) in the trunk is unaffected by K-treatment. Thus, added K has a pronounced effect on ^{137}Cs concentration in actively growing parts of trees but has little impact on the concentration and total inventory of ^{137}Cs that was in the tree trunk prior to application of K. Concentration of ^{137}Cs has been reduced to such a low level in fronds as a result of K treatment that it is essentially constant at about 2.5 Bq g^{-1} in all fronds regardless of age (Figure 2). However, the concentration of ^{137}Cs in control tree fronds declines with frond age and appears to be retrieved by the trees just as K is (Figure 3). This is similar to results reported by Jones et al., 1997 for retrieval of ^{137}Cs from leaves of *Eriophorum* back into the plant.

The ^{137}Cs/K ratio in various compartments of twenty K-treated coconut trees and ten control coconut trees are listed in Table 6. It is more than an order of magnitude higher in compartments of control trees than in compartments of K-treated trees. Mean ^{137}Cs/K ratio for each compartment in control trees is essentially the same. All are within one standard error of each other except the highest ratio for the "husk + shell" compartment that is driven by one value that is three times higher than all other values. This is reflected in the high standard error for that compartment and is probably due to an error somewhere along the labeling, processing, and analytical sequence but the source of error was not obvious so the data point is included. In contrast, ^{137}Cs/K ratios in K-treated trees are the same for all compartments (all within one standard error of each other) except the trunk that has a ratio about four times greater than the other compartments. When additional K is available in the plant there is a large reduction of ^{137}Cs in all actively growing parts of the tree relative to the largely unaffected ^{137}Cs inventory in the trunk.

Mean ^{137}Cs/K ratio in fronds for both K-treated trees and control trees is shown in Figure 4. It decreases continuously with frond age in control trees. Bunzl and Kracke (1989) also observed a decrease in the ^{137}Cs/K ratio in peatland grasses and interpreted this as K being released more readily than ^{137}Cs from the aging leaves to the soil surface. However, coconut fronds have a hard wax-like surface from the time they are frond 1 until they have progressed to the position of being the last two fronds on the tree (between frond 35 and 42 for most trees) at which time they lose the hard waxy surface, turn brown and deteriorate.

The hard wax-like surface of the fronds prior to deterioration prohibits leaching of ^{137}Cs and K from the fronds during rainfall. Less than 0.1% of ^{137}Cs in fronds is released to the water when the amount of water available is equal to the frond mass (Robison and Stone unpublished data). Therefore, in contrast to the interpretation reported by Bunzl and Kracke, 1989, it appears that the very low ^{137}Cs mass concentration of 6.9 **picograms g^{-1}** in the youngest fronds that have the highest concentration of ^{137}Cs is retrieved by the tree on a percentage basis more readily than the vastly larger concentrations (> 20 **milligrams g^{-1}**) of K in fronds thus causing the declining ratio throughout the frond life cycle.

In contrast, the ^{137}Cs/K ratio in K-treated trees appears to increase but only because of the unique situation of ^{137}Cs concentration in K-treated fronds. Mass concentrations (0.69 **picograms g^{-1}**) of ^{137}Cs are so low in fronds from K-treated trees that there is no recovery of ^{137}Cs as the fronds age. This is not a detection limit issue. The detection limit for ^{137}Cs in our analytical facility is about 10 to 30 mBq and the concentrations of ^{137}Cs involved here are counted to an accuracy of about 3%. Thus, decreasing concentration in K with frond age and the constant, low concentration of ^{137}Cs (Figure 2) gives the appearance of an increasing^{137}Cs/K ratio but the appearance is totally driven by the low, constant concentration of ^{137}Cs that is irretrievable by the plant.

This large-scale field study with trees supports the concept developed from short-term studies using two upland species of vegetation (Jones et al., 1991) and springwheat (Zhu et al., 2000a) that internal K in a plant can have as great an affect on ^{137}Cs uptake by plant roots and on plant allocation strategies of K and ^{137}Cs. The application of high levels of K likely had a direct effect on K uptake in the year of application possibly through a combination of direct ion dilution in the soil solution and competition for uptake between K and Cs through putative K transporters and channels. Plants are also known to respond rapidly to increases in internal and external K concentrations by down-regulating the expression of high affinity K transporters resulting in a proportional increase in the flux of K through low-affinity channels (White and Broadley, 2000). Since the known K transporters and channels exhibit differential sensitivity to Cs it is possible that the shift in activity of these K transporters and channels could also result in differential uptake of Cs and K. The inward rectifying K^+ channel (AKT1) is a major pathway for K transport under K-replete conditions and is thought to be impermeable to Cs (White and Broadley, 2000). This channel, however, is inhibited by Cs^+ (Bertl et al., 1997), and mutants lacking this channel do not exhibit reduced Cs uptake or accumulation suggesting that AKT1 is probably not a main Cs^+ uptake pathway (White and Broadley, 2000). Thus a shift in proportional K^+ uptake through AKT1, as a result of high external or internal K concentrations, might result in a persistent reduction in Cs uptake.

Following the year of K application, after soil K concentrations had returned to pre-treatment levels, the persistent reduction in Cs concentrations seen in annual tissues can not be explained on the basis of soil K/Cs interactions or competition for uptake at the root surface. In addition to the possible role of a shift in mechanism and selectivity of K uptake described above, the results observed here may be a result of changes in within-plant allocation of K. Previous research in tree species has indicated that uptake of potassium is reduced in trees that contain substantial stores of K (Burns, 1992; Rosecrance et al, 1996, 1998), under these circumstances stored nutrients are preferentially utilized to supply K requirements of growing tissues and new soil uptake is limited.

Calculations of K storage in coconut trees used in this experiment suggest that the single fertilization event had the potential to support many years of annual K demand. Ten years post fertilization, K treated trees contain approximately 3 kg of total aboveground K while control trees contain less than1 kg of total K. In annual tissues (fronds, spike, spath and fruits), in which K is expected to be most mobile, K treated trees contain approximately 2.2 kg of total aboveground K while control trees contain less than 0.75 kg of total K. Potassium storage in root tissues was not estimated in these studies but is known to be significant in perennial species where root stored K was 125% greater than shoot storage (Rosecrance, 1998). The K content of unfertilized trees grown under these limiting conditions, likely represents structural and non-available K. The difference between K content in control and K fertilized trees of at least 2 kg observed here, can be considered available to support the K demand of subsequent growth. In K fertilized trees an estimated 220 g of K is lost to the soil in fruit and frond removal with an unknown, but likely substantial amount of this K loss recycled through soil uptake in subsequent years. Collectively, these results suggest that the luxury accumulation of K during the single fertilization event provided sufficient K to support at

least 10 years of K demands of these trees. This estimation is likely highly conservative as it does not include root K storage and is based upon tree harvests conducted 10 years after the fertilization event.

The reduction in ratio of Cs/K in fertilized trees is likely a consequence of an immediate and persistent inhibition of soil K and Cs uptake as a result of down regulation of K uptake mechanisms caused by increased internal K accumulation. The availability of stored K also alters tissue K and Cs allocation patterns. Organs that are predominantly supplied with nutrients through phloem transport (coco fluid, coco meat, heart) exhibit the lowest Cs/K ratio as a result of a nearly explicit dependence upon remobilized (phloem transported) K, while tissue that transpire have a proportionally higher provision of K through xylem supply and may obtain a limited amount of contain Cs obtained from the soil in addition to remobilized K. The sensitivity and specificity of phloem K loading mechanisms to Cs is unknown.

The trunk tissue retains a proportionally higher Cs/K ratio as it consists of tissues that were developed prior to the initiation of the experiment under low natural soil K conditions and consequential uptake of Cs through K transport mechanisms. The slightly lower Cs concentration in trunks of K treated trees is likely a result of dilution in new trunk tissue formed over the ten year experimental period.

Data from experiments where only a portion (one half or less) of the root system of coconut trees growing in atoll soil were fertilized at the same kg K m^{-2} that was applied to the entire root zone of the other trees led to a reduction of ^{137}Cs in coconuts that was about 90% percent of that observed when the entire root system received K (Stone and Robison, 2002). Similar results were obtained in a controlled field experiment using corn (Zea *mays*) cv Silver Queen and sorghum (*sorghum bicolor)*. For these plants the kg K m^{-2} was applied over the entire root zone and for comparison the same amount of kg K m^{-2} over only half of the root zone. The reduction in ^{137}Cs in the plants was the same for both applications again suggesting that the internal supply of K is the critical factor regulating plant uptake of ^{137}Cs (Stone and Robison, 2002).

Based on data in this manuscript from sacrificed coconut trees, and previous studies discussed above, an alternative hypothesis appropriate for atoll soils lacking silicate clays is that K concentration within plants controls ^{137}Cs uptake. If so, then uniform distribution of applied K to the soil surface would be unnecessary to achieve large reductions of ^{137}Cs in plants. Thus, K can be applied in every other row of trees thereby reducing by one half the required K and the area that must be covered. This will significantly reduce cost of remediation for the island.

5.0 Conclusions

Potassium inventory in K-treated coconut trees is greater by a factor of 5.6 than K inventory in trunks of control trees and is a critical factor in the long-term reduction of ^{137}Cs observed in edible fruits at atolls. This greatly reduces the estimated radiation dose to people who would resettle the islands and consume local tree food crops. Reduced concentration of ^{137}Cs in edible fruits lasts for at least 10 y after the last application of K. This provides important assurances that reduction in ^{137}Cs is long term and that the radiation dose from consuming local plant foods will remain low upon resettlement. Based on these data, K needs to be applied no more frequently than about every 10 y which greatly reduces the cost of remediation versus annual or biennial applications of K that is common in many agricultural practices.

This large-scale field study with coconut trees supports the concept that internal K in trees greatly influences ^{137}Cs uptake by plant roots, and allocation strategies of K and ^{137}Cs within the plant. This is consistent with the hypothesis by Jones et al. 1991 and Zhu et al. 2000a, that internal concentration of K in plants is as important as the K concentration and the ^{137}Cs /K ratio in the soil when it comes to regulating uptake of ^{137}Cs by roots and allocation strategies for K and ^{137}Cs within the plant.

Based on data presented here, and data for coconut trees, sorghum and sweet corn from previous studies where the ^{137}Cs concentration in the plants was the same whether K was applied over the entire root zone or over only half of the root zone, suggests that the internal supply of K is the critical factor regulating plant uptake of ^{137}Cs rather than the proportion of the root system in K-treated soil. If so, then uniform distribution of K over the entire root-zone would not be necessary to achieve large reductions of ^{137}Cs in plants. The application of high levels of K likely had a direct effect on K uptake in the year of application possibly through a combination of direct ion dilution in the soil solution and competition for uptake between K and Cs through putative K transporters and channels. Plants are also known to respond rapidly to increases in internal and external K concentrations by down-regulating the expression of high affinity K transporters resulting in a proportional increase in the flux of K through low-affinity channels (White and Broadley, 2000). Since the known K transporters and channels exhibit differential sensitivity to Cs it is possible that the shift in activity of these K transporters and channels could also result in differential uptake of Cs and K. The inward rectifying K+ channel (AKT1) is a major pathway for K transport under K replete conditions and is thought to be impermeable to Cs (White and Broadley, 2000). This channel, however, is inhibited by Cs^+ (Bertl et al., 1997), and mutants lacking this channel do not exhibit reduced Cs uptake or accumulation suggesting that AKT1 is probably not a main Cs+ uptake pathway (White and Broadley, 2000). Thus a shift in proportional K+ uptake through AKT1,as a result of high external or internal K concentrations, might result in a persistent reduction in Cs uptake.

Following the year of K application, after soil K concentrations had returned to pre-treatment levels, the persistent reduction in Cs concentrations seen in annual tissues can not be explained on the basis of soil K/Cs interactions or competition for uptake at the root surface. In addition to the possible role of a shift in mechanism and selectivity of K uptake described above, the observed effects may be a result of changes in within-plant allocation of K. Previous research in tree species has indicated that uptake of potassium is reduced in trees that contain substantial stores of K (Burns, 1992; Rosecrance et al, 1996, 1998), under these circumstances stored nutrients are preferentially utilized to supply K requirements of growing tissues and new soil uptake is limited.

Calculations of K storage in coconut trees used in this experiment suggest that the single fertilization event had the potential to support many years of annual K demand. Ten years post fertilization, K treated trees contain approximately 3 kg of total aboveground K while control trees contain less than1 kg of total K. In annual tissues (fronds, spike, spath and fruits), in which K is expected to be most mobile, K treated trees contain approximately 2.2 kg of total aboveground K while control trees contain less than 0.75 kg of total K. Potassium storage in root tissues was not estimated in these studies but is known to be significant in perennial species where root stored K was 125% greater than shoot storage (Rosecrance, 1998). The K content of unfertilized trees grown under these limiting conditions, likely represents structural and non-available K. The difference between K content in control and K fertilized trees of at least 2 kg observed here, can be considered available to support the K demand of subsequent growth. In K fertilized trees an estimated 220 g of K is lost to the soil in fruit and frond removal with an unknown, but likely substantial

amount of this K loss recycled through soil uptake in subsequent years. Collectively, these results suggest that the luxury accumulation of K during the single fertilization event provided sufficient K to support at least 10 years of K demands of these trees. This estimation is likely highly conservative as it does not include root K storage and is based upon tree harvests conducted 10 years after the fertilization event.

The reduction in ratio of Cs/K in fertilized trees is likely a consequence of an immediate and persistent inhibition of soil K and Cs uptake as a result of down regulation of K uptake mechanisms caused by increased internal K accumulation. The availability of stored K also alters tissue K and Cs allocation patterns. Organs that are predominantly supplied with nutrients through phloem transport (coco fluid, coco meat, heart) exhibit the lowest Cs/K ratio as a result of a nearly explicit dependence upon remobilized (phloem transported) K, while tissue that transpire have a proportionally higher provision of K through xylem supply and may obtain a limited amount of contain Cs obtained from the soil in addition to remobilized K. The sensitivity and specificity of phloem K loading mechanisms to Cs is unknown.

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Figure legends

Figure 1. Mean concentration of K and ^{137}Cs in the entire above ground portion of twenty K-treated coconut trees and ten control coconut trees (no added K).

Figure 2. Mean concentration of ^{137}Cs and K in fronds from K-treated trees. Error bars are 1 standard error.

Figure 3. Mean concentration of K and ^{137}Cs in fronds from ten control trees. Data are separated into two linear components for both K and ^{137}Cs.

Figure 4. Mean ^{137}Cs/K ratio in fronds of control and K-treated coconut trees. Error bars are 1 standard error.

Table 1. The mean percent of the total mass (wet wt.) of the coconut tree in each compartment

Coconut Tree Compartment	Percent of Total Mass			
	K-treated trees		Control trees	
	Mean	Stderr	Mean	Stderr
Trunk	55	1.4	51	3.1
Fronds	26	1.3	28	2.5
Husk +Shell	9.4	1.3	11	1.8
Heart	3.2	0.3	2.7	0.32
Flower + spathe	2.6	0.2	2.8	0.5
DCF[a]	1.7	0.3	1.7	0.29
Spike	1.6	0.1	1.7	0.2
DCM[b]	1.0	0.2	1.0	0.2
Total	101		100	

a = drinking coconut fluid
b = drinking coconut meat

Table 2. Dry/wet ratio for twenty-eight coconut tree-trunk sections

Coconut tree trunk section	Dry/wet ratio Mean	No.	Stderr
0.0 m to 1.0 m	0.30	26	0.02
1.0 m to 2.0 m	0.32	26	0.02
2.0 m to 3.0 m	0.30	26	0.02
3.0 m to 4.0 m	0.28	25	0.02
4.0 m to 5.0 m	0.28	19	0.01
5.0 m to 6.0 m	0.26	18	0.01
6.0 m to 7.0 m	0.27	13	0.01
7.0 m to 8.0 m	0.25	8	0.01
8.0 m to 9 0 m	0.25	1	
9.0 m to 10 m	0.26	1	
Palm Heart	0.18	26	0.005

Table 3. Mean K concentration in various compartments of 10 control and 20 K-treated coconut trees

	K-treated trees K		Control trees K		Ratio K-treated to
	g kg^{-1}	Stderr	g kg^{-1}	Stderr	Control
Trunk	1.8	0.1	0.32	0.03	5.6
Heart	3.8	0.1	1.0	0.1	3.8
Spathe	5.1	0.3	1.8	0.2	2.8
Spike	4.9	0.1	2.1	0.2	2.3
Fronds	4.5	0.2	0.85	0.1	5.3
Coco meat	4.0	0.1	2.8	0.1	1.4
Coco fluid	2.0	0.04	1.1	0.1	1.8
Husk+shell	3.8	0.2	0.84	0.1	4.5

Table 4. Mean ^{137}Cs concentration in various compartments of 10 control and 20 K-treated coconut trees

	K-treated trees ^{137}Cs kBq kg^{-1}	Stderr	Control trees ^{137}Cs kBq kg^{-1}	Stderr	Ratio K-treated to Control
Trunk	0.71	0.09	1.0	0.12	0.70
Heart	0.44	0.05	2.8	0.31	0.16
Spathe	0.75	0.15	5.5	0.69	0.14
Spike	0.52	0.07	5.7	0.64	0.09
Fronds	0.72	0.10	2.1	0.24	0.34
Coco meat	0.52	0.07	6.2	0.67	0.08
Coco fluid	0.19	0.03	2.2	0.41	0.09
Husk+shell	0.58	0.08	4.4	0.45	0.13

Table 5. The mean normalized[a] mass of the fronds from each coconut tree in the control and K-treated groups

Frond No.	K-treated trees Normalized Mean	Stderr	Control Trees Normalized Mean	Stderr
1	1.0		1.0	
2	1.1	0.03	1.1	0.03
3	1.2	0.07	1.2	0.03
4	1.2	0.04	1.2	
5	1.2	0.05	1.3	0.06
10	1.3	0.07	1.4	0.06
15	1.3	0.07	1.4	0.06
20	1.4	0.07	1.4	0.06
25	1.4	0.07	1.4	0.1
30	1.3	0.07	1.4	0.06
35	1.4	0.05	1.3	0.06
40	1.3	0.05		

[a]Mass of each tree frond was divided by the mass of frond 1 for that tree

Table 6. Mean ^{137}Cs/K ratios in various compartments of 10 control and 20 K-treated coconut trees

	K-treated trees ^{137}Cs/K ratios kBq kg^{-1}/g kg^{-1}		Control trees ^{137}Cs/K ratios kBq kg^{-1}/g kg^{-1}	
	Mean	**Stderr**	**Mean**	**Stderr**
Trunk	0.42	0.05	3.5	0.6
Heart	0.11	0.01	3.0	0.6
Spike	0.11	0.01	2.9	0.5
Spathe	0.15	0.04	3.6	0.6
Fronds	0.17	0.03	2.5	0.4
C. meat	0.13	0.02	2.2	0.3
C. fluid	0.09	0.02	2.2	0.5
Husk+shell	0.16	0.02	5.7	1.0