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Journal of Environmental Radioactivity 59 (2002) 223–243

JOURNAL OF
ENVIRONMENTAL
RADIOACTIVITY

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Uptake of ^{40}K and ^{137}Cs in native plants of the Marshall Islands

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Abstract

Uptake of ^{137}Cs and ^{40}K was studied in seven native plant species of the Marshall Islands. Plant and soil samples were obtained across a broad range of soil ^{137}Cs concentrations (0.08–3900 Bq/kg) and a narrower range of ^{40}K soil concentrations (2.3–55 Bq/kg), but with no systematic variation of ^{40}K relative to ^{137}Cs . Potassium-40 concentrations in plants varied little within the range of ^{40}K soil concentrations observed. Unlike the case for ^{40}K , ^{137}Cs concentrations increased in plants with increasing ^{137}Cs soil concentrations though not precisely in a proportionate manner. The best-fit relationship between soil and plant concentrations was $P = aS^b$ where a and b are regression coefficients and P and S are plant and soil concentrations, respectively. The exponent b for ^{40}K was zero, implying plant concentrations were a single value, while b for ^{137}Cs varied between 0.51 and 0.82, depending on the species. For both ^{40}K and ^{137}Cs , we observed a decreasing concentration ratio (where concentration ratio = plant concentration/soil concentration) with increasing soil concentrations. For the CR values, the best-fit relationship was of the form $\text{CR} = aS^b/S = aS^{b-1}$. For the ^{40}K CR functions, the exponent $b-1$ was close to -1 for all species. For the ^{137}Cs CR functions, the exponent $b-1$ varied from -0.19 to -0.48 . The findings presented here, as well as those by other investigators, collectively argue against the usefulness of simplistic ratio models to accurately predict uptake of either ^{40}K or ^{137}Cs in plants over wide ranges of soil concentration. Published by Elsevier Science Ltd.

Keywords: Cesium-137; Soil; Vegetation; K-40; Concentration ratio

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1. Introduction

Relationships between substrate (soil) and plant concentrations for both nutrients and contaminants have been widely studied. These studies have been used for various purposes, including the development of models for predicting plant concentrations from measurements of the soil concentration. Plant:soil ratios for nutrients, metals, and a variety of radionuclides have been empirically determined, often times from small-scale studies with the intent of generalizing the findings to all values of soil concentration. In most cases, plant:soil relationships can be described by an increase in plant concentrations with an increase in soil concentrations. A linear relationship implies a constant ratio of plant concentration to soil concentration. Plant:soil ratios, usually termed concentration ratios (CRs), have been used extensively in radiological assessment models to estimate concentrations of radionuclides in vegetation. Vegetation concentrations, in turn, are used to calculate radiation exposures and doses to people as a result of direct consumption of vegetation, or via foodchain transfer from plants to animals to man.

A number of studies, however, have shown that CR values for individual elements or radionuclides are not constant over a wide range of soil concentrations (Mengel & Kirkby, 1982; Brown, Wong, & Buhler, 1984; Sheppard & Sheppard, 1985; Simon & Ibahim, 1987; Sheppard & Evenden, 1988, 1990; Livens, Horrill, & Singleton, 1991; McGee, Johanson, Keatinge, Synnott, & Colgan, 1996). Thus, a more reliable relationship between the concentration ratio and the soil concentration is needed for estimating transport of radionuclides into vegetation and through the food chain.

Uptake of ions by plants can be influenced by many factors. For example, the availability of ions in soils can be affected by soil moisture, cation exchange capacity, pH, percent organic matter, percent nutrients in the soil, and clay content (Amphlett & McDonald, 1956; Spitsyn et al., 1958; Romney & Childress, 1965; Broseus, 1970; Essington, Fowler, & Polzer, 1981; Zach, Hawkins, & Mayoh, 1989; Schuller, Lovengreen, & Handl, 1993; Papastefanou, Manopoulou, Stoulos, & Ioannidou, 1999). The great variety of factors that can influence uptake adds considerable uncertainty to predicted values of vegetation concentrations. Locations where soil characteristics have little regional variation, as well as having a wide range of radionuclide concentrations, are ideal for studying concentration effects.

The Republic of the Marshall Islands (RMI) is one location where variations in soil quality and nutrient availability, as well as plant species heterogeneity are minimal, yet there is a wide range of ^{137}Cs soil concentrations as a result of regional fallout of debris from nuclear weapons testing. Cesium-137 concentrations in the soil range over four orders of magnitude on the various atolls as determined by the Nationwide Radiological Study (Simon & Graham 1994, 1997). Hence, the Marshall Islands provide an ideal ecological setting for a study of plant:soil concentration ratios. The original and primary purpose of this investigation was to determine concentrations of ^{137}Cs in native plants of the Marshall Islands at all the atolls and to report those values as documentation of present day contamination. Relating those findings to the concentration of ^{40}K in the soil was a secondary goal. In this paper, we report CR values for ^{40}K and ^{137}Cs in eight plant species, as well as data and

discussion to support the hypothesis that concentration ratios of ^{40}K and ^{137}Cs , at least in coral-based soil, are not invariant over a wide range of soil concentration.

2. Environment

The RMI is made up of 29 atolls and five separate reef islands. There are more than 1200 islands with a total land area of approximately 180 km². The atolls are located between 4.5 and 14.5° N latitude and between 160 and 174° E longitude. Presently, 22 of the atolls and four of the separate reef islands are inhabited.

The climate of the Marshall Islands is tropical with winds typically from the NE and an average humidity near 80%. Average annual rainfall varies considerably, with the northern atolls being considerably drier than those in the southern part of the nation. The rainfall gradient across the Marshall Islands can be described by a curvilinear relationship with latitude; northern islands often receive less than 110 cm annually and southern islands in excess of 350 cm.

Soils in the Marshall Islands are primarily calcium carbonate material that originated from oceanic organisms that build reefs on slowly sinking volcanic cones. Coral atolls and islands were formed by erosion of coral material that was deposited slightly above high tide from waves and storms. Island development proceeded as land was stabilized by terrestrial plants. The organic fraction in Marshall Islands soil has been reported to be between 1 and 15% on a dry weight basis (USDA, 1989). The remainder of the bulk soil is a CaCO₃ matrix that is nearly, if not totally, devoid of clay minerals (USDA, 1989; Kenady, 1962; Welander et al., 1966) and typically low in potassium and other nutrients. A more complete discussion of the mineral composition of carbonate soil can be found in Robison, Conrado, Hamilton, and Stoker (2000).

3. Materials and methods

3.1. Plant types

The plant species discussed here are conventionally used for two different purposes in Marshallese society: as ingredients in traditional medicines and as food (Table 1). Five species of plants used for medicinal purposes were studied; they are *Morinda citrifolia* (commonly called “Indian mulberry” in English; a small tree that can reach 8–10 m in height and has soft, greenish/yellow spherical fruit), *Polypodium scolopendria* (a fern like plant that grows on the ground and on tree trunks), *Tournefortia argentea* (a tree that grows to 5 m height or more, has large leaves and small bunches of tiny flowers and fruit; grows exclusively on hot, salty, coastal environments and is a colonizer species), *Triumfetta procumbens* (a low-growing vine that grows to lengths up to 3 m; has yellow flowers and small fruit covered with spines and is common in the coastal environment), and *Scaevola taccada* (a woody shrub that grows to heights of 3 m and produces white flowers; a colonizer plant and commonly

Table 1

Plant species (Marshallese name), usage category (F=food, M=medicinal), number of CR values determined from matched plant and soil samples, and depth of related soil samples

Plant species (Marshallese name)	Usage category	Number of CR values of ^{137}Cs	Number of CR values of ^{40}K	Soil collection depth (cm)
<i>Cocos nucifera</i> (Ni)	F	201 meat 215 milk	180 meat 195 milk	0–30
<i>Morinda citrifolia</i> (Nen)	M	53 leaf 54 fruit	50 leaf 50 fruit	0–30
<i>Pandanus</i> spp. (Bop)	F	4	3	0–30
<i>Polypodium scolopendria</i> (Kino)	M	33	32	0–10
<i>Scaevola taccada</i> (Kinnat)	M	46	45	0–30
<i>Tacca leontopetaloides</i> (Mokmok)	F	9	8	0–30
<i>Tournefortia argentea</i> (Kiden)	M	50	54	0–30
<i>Triumfetta procumbens</i> (At'at)	M	46	41	0–10

found on coastal zones). Further information on the medicinal uses of these plants can be found in Merlin, Capelle, Keene, Juvik, and Maragos (1994) and Duffy, Simon, and Whicker (1999).

The plant species that we studied that are consumed as food are *Cocos nucifera* (commonly called coconut; provides a clear liquid juice from immature fruit and a thicker white milk and dense meat from mature fruit; only data for the drinking juice is reported here), *Pandanus* spp. (a large tree that produces a large pineapple like fruit from which juice is extracted), and *Tacca leontopetaloides* (a herbaceous shrub that produces a root crop commonly called arrowroot that must be processed through several leaching steps before it is consumed).

Marshallese names for all plant species studied are noted in Table 1.

3.2. Site selection and sampling density

Locations for plant and soil sampling were generally selected in the interior of the islands, away from the beach areas that are prone to severe erosion and occasional wash-over by waves and sediments. Spatial density of plant and corresponding soil samples was determined mainly by resource allocation considerations. In particular, the number of samples obtained from each atoll was generally limited by the available time during the radiological survey of that atoll, which included many other activities. Social and/or political demands from the local population also influenced sampling density. That type of information was derived from meetings with local leaders or with the local community. In particular, *C. nucifera* was sampled more frequently than other plant species because of its common occurrence and because of its great importance to the Marshallese for providing both solid food nourishment and liquid replenishment.

Vegetation samples typically consisted of leaves or fruits from one tree or shrub or from several plants in close proximity (within 10 m). Typically, 1–2 kg of material was collected (Duffy, 1994) and prepared for transport to a laboratory in the capital

city, Majuro. Soil was collected around the root zone of the plants to a depth of 30 cm for six herbaceous species and to a depth of 10 cm for a vine and a shallow-rooted fern (see Table 1). Sampling the soil to 30 cm depth for the herbaceous species was generally sufficient for two reasons: the organic layer of soil in the Marshall Islands is shallow, and most of the ^{137}Cs is retained within the top 30 cm (Graham & Simon, 1996). Because some of the ^{137}Cs has penetrated deeper than 30 cm, and because the roots of the larger trees, such as *C. nucifera*, likely extend to greater depths, some artificial variability of the CR values is introduced by the sampling protocol. However, the CR values reported here were determined consistently within and among species.

Prior to collecting the soil, litter and fallen or fresh vegetation were removed from the surface of the soil. Three large holes were excavated and a representative sample of approximately 2 l of soil was collected by compositing soil from each hole.

Soil adjacent to *C. nucifera* trees was obtained in the field by the same methods, but fruit samples were prepared as follows. Approximately 1 l of juice was collected from green (immature) coconuts and stored in airtight containers. The juice was refrigerated or frozen, and preservative was added. The meat was removed from the fruit and double bagged for transport. All samples were transported in waterproof containers to the Majuro laboratory.

Only matched plant and soil samples were used to estimate concentration ratios. The number of samples for each plant species is given in Table 1.

3.3. Soil preparation

All soil concentrations were determined on a dry weight basis and all soil samples were dried and homogenized similarly as described here. First, the sample was placed in labeled aluminum containers lined with absorbent paper for drying. Samples were dried under high intensity heat lamps at approximately 38°C until there was less than 1% change in weight over the previous 24 h. The first and last weights were used to calculate the percent water in the soil. A two-step preparation process was used to ensure homogeneity of soil samples. First, each sample was sieved for at least 5 min using a mechanized shaker. Particles >0.85 mm were separated and mechanically crushed until less than 1% of the total sample weight was composed of particles greater than 0.85 mm. The entire sample was then thoroughly mixed in a container on a rotating ball mill. An aliquot of 500, 1000 or 2000 ml was removed and placed in a Marinelli beaker for gamma spectrometry. Soil samples of either 500 or 1000 ml volumes were counted in a 1000 ml Marinelli beaker, 12.9 cm in diameter and 15.2 cm tall. Soil samples of 2000 ml were counted in a 2000 ml Marinelli beaker, 15.7 cm in diameter and 16.5 cm tall.

3.4. Vegetation preparation

Leaf, fruit and coconut meat samples were chopped in a blender or by hand to achieve a uniform sample as described by Duffy (1994). To assist in blending, distilled water was added as needed. The samples were preserved using sodium benzoate

($\text{NaC}_7\text{H}_5\text{O}_2$). Plant samples were dried in a similar manner to that of soil as described above. All plant concentrations and CR values are reported in terms of dry weight except for coconut milk, which is reported in terms of wet weight. Vegetation samples were analyzed by gamma spectrometry in a 2000 ml Marinelli beaker.

3.5. Gamma spectrometry

Four high-purity (HPGe) detectors were used for analyzing gamma-emitting radionuclides. Two electrically cooled (liquid helium recycling) detectors were predominantly used for these measurements. These instruments were of closed-end coaxial geometry, with an efficiency of 40% (relative to a 7.5×7.5 cm NaI crystal) and a resolution of 2.1 keV (FWHM) at 1.33 MeV. In addition, two liquid nitrogen cooled HPGe detectors were also used. They were both of closed-end coaxial geometry with an efficiency of 40% (relative to a 7.5×7.5 cm NaI crystal) and a resolution of 2.0 keV (FWHM) at 1.33 MeV. Spectrum analysis was performed using a PC-based data acquisition and analysis program.

3.6. Determination of cesium-137 concentration

Radionuclide standards for determining detector efficiency at each relevant energy and for each different counting geometry were prepared so that radionuclide concentrations in soil could be estimated. The standards were prepared by thoroughly mixing uncontaminated coral sand with a commercial, liquid radionuclide solution with calibrations traceable to the National Institutes of Standards and Technology (NIST).

For vegetation analysis, water-based standards were used because the density of the plant samples was nearly equal to that of water. Efficiency curves (Bq/kg per count per second as a function of volume) for water-based standards were generated for each detector over a range of sample volumes (see Duffy, 1994). Cesium-137 concentrations (Bq/kg) in both soil and plants were calculated from the net full-energy peak counts and the measured efficiency.

Several programs and procedures ensured quality control of radioactivity measurements as follows. First, an external scientific advisory group (see Simon & Graham, 1997) critically reviewed radioactivity measurement procedures. In addition, measurement precision of the laboratory was established through a blind inter-comparison with two laboratories in the USA, one laboratory in Germany, and one in New Zealand. Finally, detector efficiencies were determined daily using the counting standards described above.

3.7. Data analysis

Individual plant concentration ratios (CR) were calculated as: $\text{CR} = P/S$, where P is the radionuclide concentration in the plant (Bq/kg dry, except for coconut milk) and S is the concentration in the corresponding soil (Bq/kg dry) obtained from the immediate root-zone of the plant. Concentration ratios for each species were summar-

ized by a mean value (geometric mean) which assumes a constant ratio model, and by determining functional relationships with soil concentration.

Statistical analyses included examination of correlation (Spearman type) between ^{40}K and ^{137}Cs concentrations in soils and plants, and computation of least-squares regressions of the ^{40}K and ^{137}Cs plant concentrations and CRs with the independent variable being the soil concentration.

4. Study findings

4.1. General findings

Cesium-137 and ^{40}K concentrations were measured in approximately 225 matched plant and soil samples. Concentrations of ^{137}Cs in the eight plant species discussed have been reported elsewhere (Duffy, 1994; Duffy et al., 1999) though concentration ratios are reported here for the first time.

Because the soil concentrations were not known at the time of sampling, it was not possible to sample plants and soils equally along the soil ^{137}Cs concentration gradient. Thus, the distribution of the soil ^{137}Cs concentrations is a result of the variation of environmental contamination in the Marshall Islands as well as a reflection of the number of samples obtained from each location.

Because the presence of ^{40}K in the soil is a result of natural processes and ^{137}Cs in soil is a result of fallout deposition from weapons testing, there would be no reason to expect the two concentrations in the soil to be correlated. That assumption was verified by exploratory data analysis (see Fig. 1).

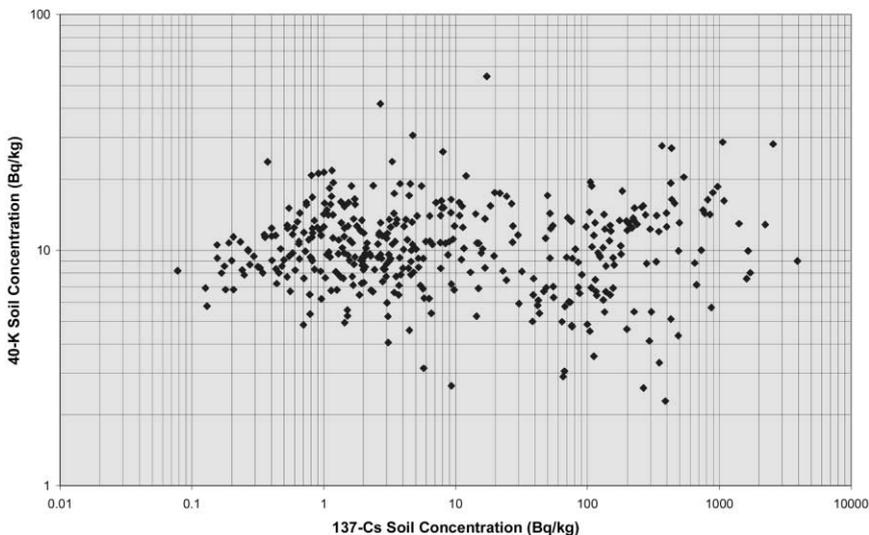


Fig. 1. Scatterplot of ^{40}K versus ^{137}Cs concentrations in soil (Bq/kg dry).

The sampling distributions of soil concentrations at each atoll and for all atolls combined, were moderately to highly skewed. Arithmetic mean values were often ten times or more greater than median values (see Table 2). Moreover, all plants were not available at all locations and at all levels of soil ^{137}Cs concentration.

4.2. Soil concentration, plant uptake and point estimates of CR for ^{40}K in *C. nucifera*

The measurements of ^{40}K in soil showed that the sample distribution was moderately skewed ($\text{CV} = 59.5\%$, $\text{GSD} = 1.64$) though the arithmetic mean soil concentration could be estimated with acceptably high precision: 10.7 ± 0.50 Bq/kg (1 SEM, $n = 200$). Concentrations ranged from 2.3 to 55 Bq/kg, though 90% of the values were in the range of 4.6–20.4 Bq/kg. These observations confirm that, like most coral atolls, the Marshall Islands are potassium deficient. Soils of volcanic origin normally contain from 40 to 1100 Bq/kg of ^{40}K (Eisenbud, 1987).

Not unexpectedly, the uptake of ^{40}K into plants was considerable. For example, coconut meat and milk had median concentrations of 635 and 73 Bq/kg, respectively, when averaged over all locations sampled. Point estimates of the CR values (geometric means) for ^{40}K in coconut meat and milk, determined from matched soil and fruit samples, were 62 ($\text{GSD} = 1.9$, $n = 180$) and 7.6 ($\text{GSD} = 1.8$, $n = 195$), respectively (see Table 3).

The ratio of highest to lowest concentrations of ^{40}K in the soil was 24, thus allowing for a limited examination of uptake of ^{40}K as a function of soil concentration. Very low Spearman correlation coefficients were determined for ^{40}K in soil and in plants: 0.10 ($p = 0.16$) for coconut meat and -0.06 ($p = 0.40$) for coconut milk. In addition, concentrations of ^{40}K in each plant species were relatively constant; see Figs. 2 and 3 for concentration data in coconut milk and meat, respectively. Hence, there was little evidence that the uptake of ^{40}K , as measured by concentrations

Table 2

Plant species, median ^{40}K concentration in soil (Bq/kg dry) and range, median, and arithmetic mean ^{137}Cs concentration in soil (Bq/kg dry) collected around the root zone of plants. Number of samples (n) is given in parentheses

Plant species	Median ^{40}K soil concentration (n)	Range of ^{137}Cs soil concentration (n)	Median ^{137}Cs soil concentration	Mean ^{137}Cs soil concentration
<i>Cocos nucifera</i>	9.2 (195)	0.16–3900 (215)	5.5	170
<i>Morinda citrifolia</i>	12 (50)	0.078–330 (54)	5.8	43
<i>Pandanus</i> spp.	6.9 (3)	890–1500 (4)	1400	1300
<i>Polypodium scolopendria</i>	15 (32)	0.66–3100 (33)	9.4	170
<i>Scaevola taccada</i>	10 (45)	ND ^a –1600 (46)	4.1	110
<i>Tacca leontopetaloides</i>	11 (8)	1.80–640 (9)	7.9	140
<i>Tournefortia argentea</i>	10 (54)	ND–1700 (50)	3.2	73
<i>Triumfetta procumbens</i>	13 (41)	0.16–1400 (46)	21	230

^a ND is not-detectable.

Table 3

Median ^{40}K concentration in individual plant species, median estimates of CR, and regression parameters for CR function ($\text{CR} = aS^{b-1}$), where S is soil concentration (Bq/kg dry). R^2 is coefficient of determination of regression

Plant species	Median concentration (Bq/kg ^a)	Median CR	a	$b-1$	R^2
<i>Cocos nucifera</i> (milk)	73	8.0	76.8	-1.03	0.60
<i>Cocos nucifera</i> (meat)	640	70	626	-0.98	0.57
<i>Morinda citrifolia</i> (fruit)	720	62	818	-1.08	0.82
<i>Morinda citrifolia</i> (leaf)	440	40	657	-1.16	0.63
<i>Pandanus</i> spp.	510	97	ns ^b	ns	ns
<i>Polypodium scolopendria</i>	1020	56	1348	-1.12	0.81
<i>Scaevola taccada</i>	530	48	273	-0.71	0.12
<i>Tacca leontopetaloides</i>	320	24	415	-1.18	0.52
<i>Tournefortia argentea</i>	530	56	528	-1.00	0.32
<i>Triumfetta procumbens</i>	570	45	654	-1.10	0.70
All plant species combined except <i>C. nucifera</i> (milk) and <i>P. scolopendria</i>	590	57	580	-1.00	0.74

^a All concentrations determined on a dry mass basis except for coconut milk.

^b ns=non-significant results because of too few samples ($n = 4$).

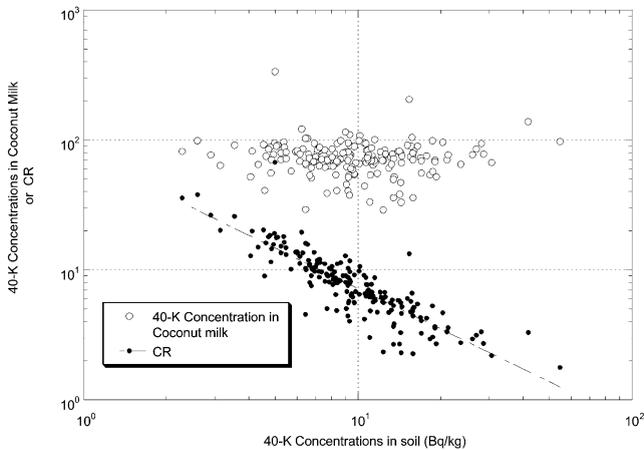


Fig. 2. ^{40}K concentrations in coconut milk (Bq/kg wet) as a function of ^{40}K soil concentration (Bq/kg dry) and CR with fitted equation, $\text{CR} = 76.8S^{-1.03}$ ($R^2 = 0.60$), where S is the soil concentration (Bq/kg dry).

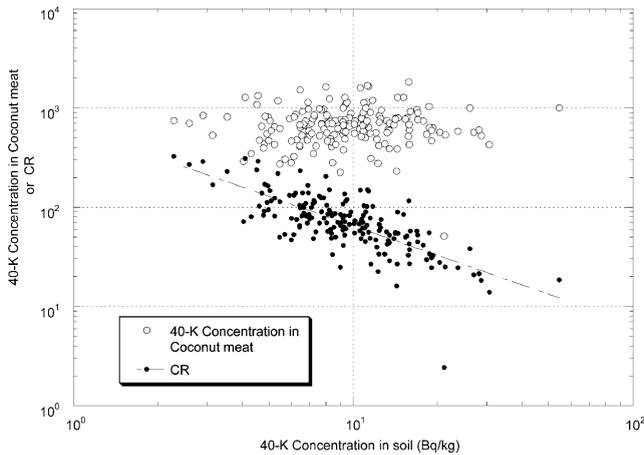


Fig. 3. ^{40}K concentrations in coconut meat (Bq/kg dry) as a function of ^{40}K soil concentration (Bq/kg dry) and CR with fitted equation, $\text{CR} = 626S^{-0.98}$ ($R^2 = 0.57$), where S is the soil concentration (Bq/kg dry).

in plant material, was related to the soil concentration. However, this finding might only be representative of the limited range of ^{40}K concentrations observed. Because there was no change in ^{40}K concentration in plants with an increase in soil concentration, we necessarily observed a trend of decreasing CR with increasing soil concentration (see Figs. 2 and 3). Functions were fit to the CR relationships as discussed in Section 4.6.

4.3. Soil concentration, plant uptake and point estimates of CR for ^{137}Cs in *C. nucifera*

Cesium-137 inventories in soil in the Marshall Islands, measured on an areal basis, range from less than 5×10^2 to more than 5×10^5 Bq/m² (Simon & Graham, 1997) while the ^{137}Cs concentration within the root-zone (0–30 cm depth) of plants sampled in this study ranged from less than 0.1 to about 4000 Bq/kg. Contamination of the soil with ^{137}Cs varies widely over the Marshall Islands primarily because of two factors: the differing distances of each of the atolls from the bomb test sites, and because the predominant wind direction from the test sites was towards a limited number of those atolls. Due to these two factors, some atolls received little or no local fallout while others received high depositions (see Simon & Graham, 1997).

The uptake of ^{137}Cs into plants appeared to be primarily determined by the local soil cesium concentration. Concentrations in soil sampled adjacent to coconut trees ranged from ~ 0.2 to nearly 4000 Bq/kg. The sampling distribution of soil concentrations was highly skewed, as evidenced by the difference between the median and means (see Table 2). The range of concentrations ranged from less than 0.5 to nearly 21,000 Bq/kg in coconut meat (dry) and from about 0.1 to 2400 Bq/kg in coconut milk (wet).

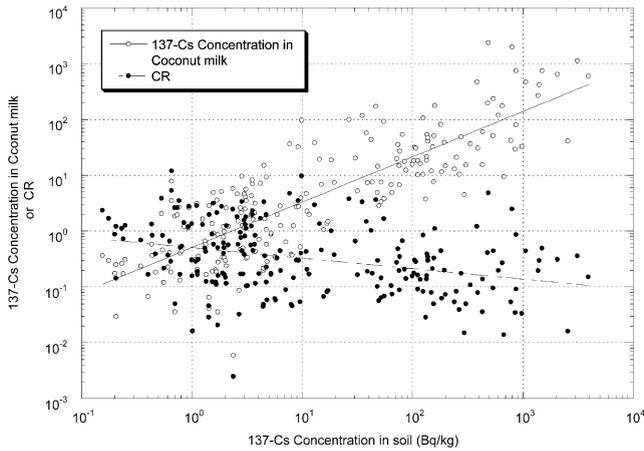


Fig. 4. ¹³⁷Cs concentrations in coconut milk (*P*, Bq/kg wet) and CR, both as a function of ¹³⁷Cs soil concentration (Bq/kg dry). *P* (coconut milk, Bq/kg) = $0.51S^{0.81}$ ($R^2 = 0.27$), CR = $0.51S^{-0.19}$ ($R^2 = 0.049$).

The local concentration of ¹³⁷Cs in soil was a moderately good indicator of the concentration found in plants, even though there was some departure from linearity. Spearman correlation coefficients between ¹³⁷Cs in soil and in coconut meat and coconut milk were 0.81 ($p < 0.001$) and 0.83 ($p < 0.001$), respectively. Point estimates of the CR (geometric means) for ¹³⁷Cs in coconut meat and milk, determined from matched soil and fruit samples, were 3.0 (GSD = 4.4, $n = 202$) and 0.32 (GSD = 4.2, $n = 215$). Moderate evidence of a decreasing trend in ¹³⁷Cs CR values with increasing soil ¹³⁷Cs concentration was also noted (see Figs. 4 and 5). This is further addressed in Section 4.6.

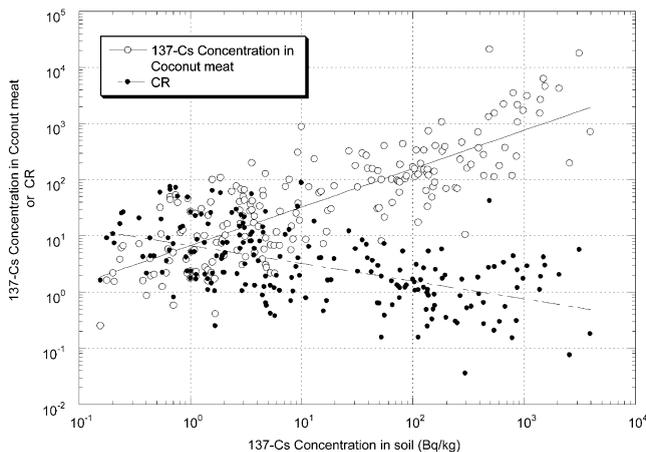


Fig. 5. ¹³⁷Cs concentrations in coconut meat (*P*, Bq/kg dry) and CR, both as a function of ¹³⁷Cs soil concentration (Bq/kg dry). *P* (coconut meat, Bq/kg) = $6.7S^{0.68}$ ($R^2 = 0.26$), CR = $6.7S^{-0.32}$ ($R^2 = 0.12$).

4.4. Interrelationship between ^{40}K and ^{137}Cs in *C. nucifera*

There was no evidence for differences in the ^{40}K soil concentration between atolls. Moreover, there was evidence of only a very weak relationship between ^{40}K in soil and ^{137}Cs uptake in plants, even though moderate applications of ^{40}K have been shown to suppress the uptake of ^{137}Cs in coral-based soil (Robison & Stone, 1992). The weak relationship between ^{40}K in soil and ^{137}Cs in plant tissues was indicated by low values of Spearman correlation coefficients. For example, the correlation coefficients between ^{40}K in soil and ^{137}Cs in coconut meat and coconut milk were -0.16 ($p = 0.034$) and -0.20 ($p = 0.0049$), respectively. Hence, within the range of our observations, ^{40}K concentrations in soil appeared to have had little or no influence on the degree of uptake of ^{137}Cs into coconuts, except that ^{137}Cs uptake is pronounced in this environment due to low overall ^{40}K soil concentrations.

The uptake of ^{40}K in all plant types relative to its concentration in soil was greater than the uptake of ^{137}Cs relative to its concentration in soil. Potassium-40 was concentrated into plant tissues from seven to ten times more than was ^{137}Cs . This conclusion can be noted by comparing CR values presented in Tables 3 and 4.

4.5. Estimation of functions for plant uptake and for CR

The data on uptake of ^{40}K and ^{137}Cs was examined in *C. nucifera* and the other plant types to look for soil concentration related trends. Because of the near constant concentration of ^{40}K in plant tissues over the range of ^{40}K soil concentration, the CR values at the highest ^{40}K soil concentrations were necessarily lower than those at the lower ^{40}K soil concentration (see Figs. 2 and 3). The best fit function to the

Table 4

Median estimates of CR values for each plant type and parameters of functions for ^{137}Cs concentrations in individual plant species ($P = aS^b$) and CR function ($\text{CR} = aS^{b-1}$), where P is plant concentration and S is soil concentration (Bq/kg dry for all plant tissues and soil except for coconut milk which was measured on a wet basis), CR is unitless. R^2 is coefficient of determination of regression for plant uptake equation

Plant species	Median CR	a	b	R^2	$b-1$
<i>Cocos nucifera</i> (milk)	0.32	0.51	0.81	0.27	-0.19
<i>Cocos nucifera</i> (meat)	3.0	6.7	0.68	0.26	-0.32
<i>Morinda citrifolia</i> (fruit)	8.7	21.9	0.66	0.80	-0.34
<i>Morinda citrifolia</i> (leaf)	5.8	14.4	0.61	0.78	-0.39
<i>Pandanus</i> spp.	9.0	39.2	0.75	0.81	-0.25
<i>Polypodium scolopendria</i>	39	92.2	0.52	0.44	-0.48
<i>Scaevola taccada</i>	8.4	14.9	0.57	0.19	-0.43
<i>Tacca leontopetaloides</i>	5.8	13.2	0.71	0.44	-0.29
<i>Tournefortia argentea</i>	6.9	14.4	0.61	0.22	-0.39
<i>Triumfetta procumbens</i>	1.1	5.3	0.67	0.40	-0.33
All plant species combined except <i>C. nucifera</i> (milk) and <i>P.</i> <i>scolopendria</i>	4.2	10.3	0.65	0.32	-0.35

^{40}K uptake values was a constant a (Bq/kg), and the best fit to the CR values was inversely proportional to the soil concentration (S) (see Table 4), i.e.,

$$\text{CR}(S) = a/S \quad (1)$$

Unlike the situation for ^{40}K , the concentrations of ^{137}Cs in the various plants increased with increasing concentration of ^{137}Cs in the soil. However, the concentration in plant tissues did not increase in a strictly proportional manner to the soil concentration. Hence, rather than modeling the plant (P) concentrations as a direct proportion of soil concentration ($P = aS$), the best-fit relationship between soil and plant concentrations was a power function of the form, $P(S) = aS^b$ where b was consistently less than unity for all plant types.

Because the plant concentrations do not strictly increase in direct proportion to the soil concentration, the CR is not accurately represented by a single value. That is, the CR can only be approximately represented as a constant equal to $\Delta P/\Delta S$, i.e., as the slope of the linear relationship between plant (P) and soil (S) concentrations. In general, the function for CR can be estimated as the ratio of the function describing the plant concentration to the soil concentration:

$$\text{CR}(S) = P(S)/S \quad (2)$$

In this case, $P(S)$ was fit by linear regression to a power function, i.e., $P = aS^b$. Hence, Eq. (1) can be rewritten as

$$\text{CR}(S) = P(S)/S = aS^b/S = aS^{b-1} \quad (3)$$

In addition to determining the CR function by Eq. (3), the function describing CR values can also be determined by regression of the CR values as a function of soil concentration. We found that the two methods produced numerically equivalent results (see Table 4). For example, the function for plant uptake of ^{137}Cs in coconut milk was P (Bq/kg) = $0.51S^{0.81}$; the function derived by Eq. (2) was $\text{CR} = 0.51S^{-0.19}$ (see Fig. 5) and was identical to that determined by regression of the CR values. Uptake and CR values in coconut meat followed similar patterns to coconut milk, but the exponent of the power function was 0.68 rather than 0.81 (see Fig. 5).

4.6. CR values for ^{40}K and ^{137}Cs in other plant types

Potassium-40 concentration ratios were compared among plant species, as were concentration ratios for ^{137}Cs . The distributions of ^{40}K CR values for all plant types, except for milk of *C. nucifera*, were nearly identical (see Fig. 6). The observation that uptake of ^{40}K did not vary over the concentration range indicates that available potassium was regulated or limited by some mechanism.

The distributions of ^{137}Cs CR values for the various plant types (see Fig. 7) were not as similar as those for ^{40}K . In particular, the median CR value for *Triumfetta procumbens* was about one-half that for coconut meat. Similarly, the CR (median) values of *Tournefortia argentea*, *M. citrifolia* fruit, *M. citrifolia* leaf, *S. taccada*,

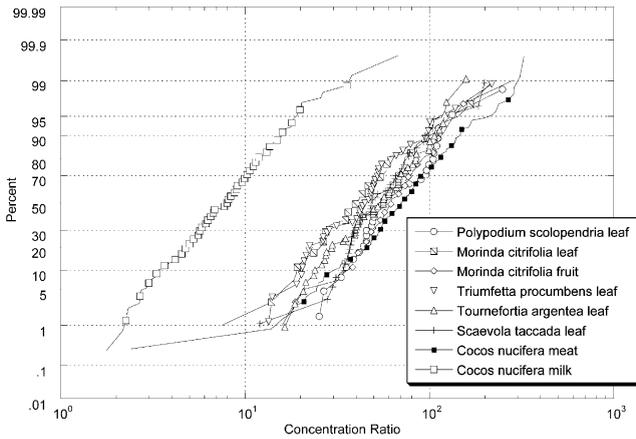


Fig. 6. Cumulative probability plot of ^{40}K CR values by plant type.

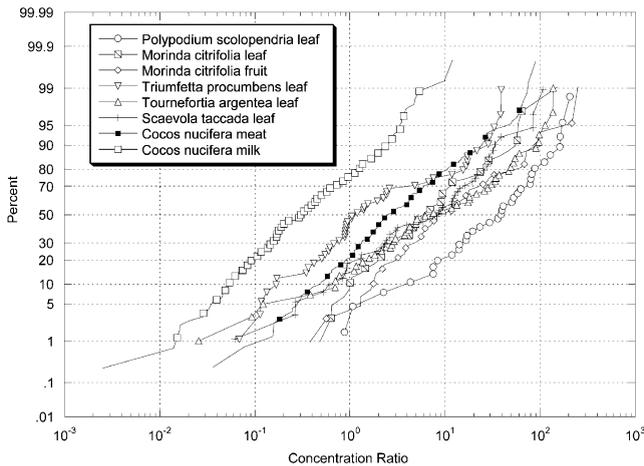


Fig. 7. Cumulative probability plot of ^{137}Cs CR values by plant type.

were about twice that of coconut meat. In one species, *P. scolopendria*, the median CR was about ten times that for coconut meat. For several of the species, there were small differences in median or arithmetic mean CR values that were statistically significant or almost so. However, in general, the median CR values could be grouped in three ranges: about 0.3 for coconut milk, about 40 for *P. scolopendria* and between 1 and 10 for the other species and plant tissues.

Because of the obvious grouping of several of the plant types into the intermediate CR range, we separately determined uptake functions and CR functions for three different plant groups: (i) coconut milk, (ii) all other plant types except coconut milk and *P. scolopendria*, and (iii) *P. scolopendria* alone. Regression values for these

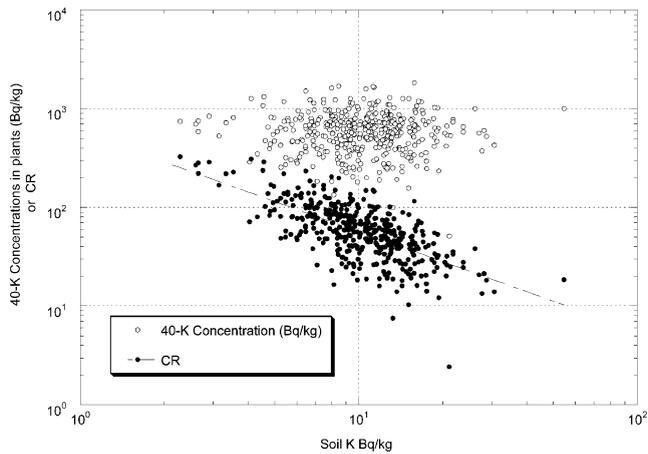


Fig. 8. ^{40}K concentrations in all plants but coconut milk and *P. scolopendria* (P , Bq/kg dry) as a function of ^{40}K soil concentration (Bq/kg dry) and CR. P (combined plants, Bq/kg)=591 (average), $\text{CR} = 581 \times S^{-1.00}$ ($R^2 = 0.74$).

groups are provided for both ^{40}K (see Table 3 and Fig. 8) and ^{137}Cs (see Table 4 and Fig. 9). The general trends were: (a) constant uptake of ^{40}K over the range of soil concentrations observed and a decreasing trend of CR with increasing soil concentration, and (b) a non-proportionate increase of ^{137}Cs in plant tissues with increasing soil concentration and a symmetrical decrease in the CR values with increasing soil concentration.

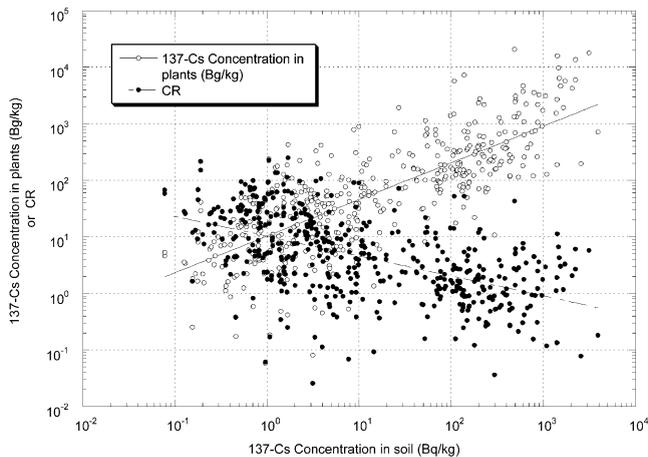


Fig. 9. ^{137}Cs concentrations in all plants but coconut milk and *P. scolopendria* (P , Bq/kg dry) as a function of ^{137}Cs soil concentration (Bq/kg dry) and CR. P (combined plants, Bq/kg) = $10.35S^{0.65}$ ($R^2 = 0.32$), $\text{CR} = 10.35S^{-0.35}$ ($R^2 = 0.15$).

5. Discussion

Other investigators have studied concentration ratios for some of the plants discussed here and their point estimates appear to be similar to ours. In particular, CRs for various plants of the Marshall Islands were reported by Robison and colleagues (Robison, Conrado, Hamilton, & Stoker, 1997; Robison et al., 2000). The number of samples analyzed in their studies was greater than in this study. However, their locations for sampling were limited to the nuclear test sites and nearby atolls. Fig. 10 provides a comparison of the range and median CR values reported by Robison et al. with those from this work for coconut meat, *Pandanus*, *T. argentea* (called *Messerschmidia* by Robison), and *S. taccada*. In general, the ranges of values reported by Robison were within the range of values reported here. This was less so for *Pandanus*, though our sample size was very small ($n = 4$).

The ranges of our CR values for *C. nucifera*, *T. argentea* and *S. taccada* are significantly wider than those reported by Robison et al. (1997, 2000), however, this is relatively easily explained. All three species are commonplace in the Marshall Islands; hence, we sampled them over a wide range of geographic locations and soil concentrations and found a wide range of CR values because of the dependence of the CR on soil concentration. The investigations of Robison et al. were conducted only at the atolls used for nuclear testing and nearby atolls. Thus, they did not sample over the same wide range of soil concentrations as was done in this work.

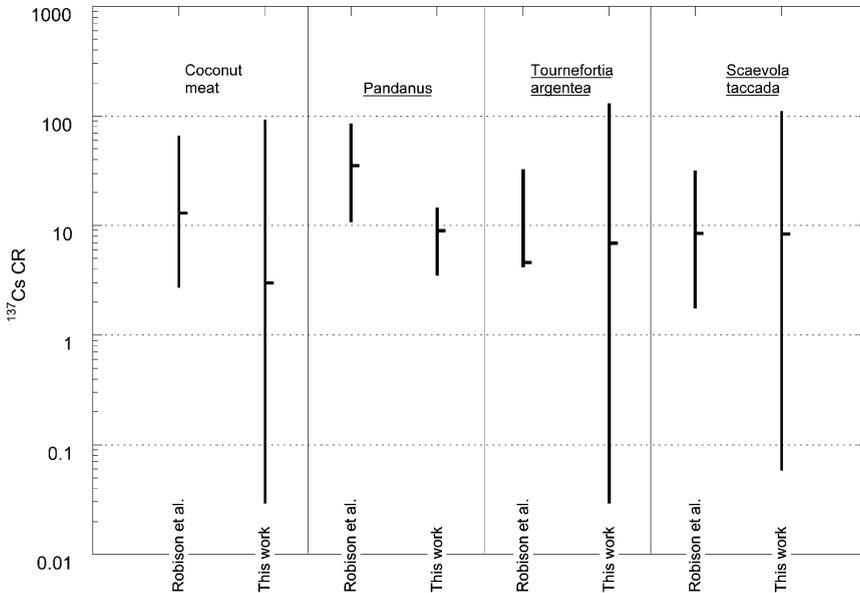


Fig. 10. Comparison of CR values from the work of Robison et al. (1997, 2000) with those reported here. The data of Robison are primarily from Bikini Island and Rongelap Island in the Marshall Islands. Vertical bars for each species represent the range (low to high) of observed values. The tick mark to the right of each bar is the sample median.

Other literature has noted trends, similar to our own, showing a dependence of concentration ratios on soil concentrations. Those various findings suggest that simple models that describe plant uptake as directly and linearly proportional to soil concentrations are not realistic and routinely fail to accurately describe environmental transfer. This section briefly summarizes some of the relevant literature on plant uptake and CRs and compares the reported data to the findings of this study. Additional literature review on ratios of plant and soil concentrations can be found in Simon and Ibahim (1987) and McGee et al. (1996).

Sheppard and Sheppard (1985) noted that CR values may be constant over several orders of magnitude, but showed that the concentration ratio did decrease as the soil concentration increased. Their explanation for the dependence of the CR on soil concentration was that ^{137}Cs might act as an essential element because soil ^{40}K is in the deficient range.

Sheppard and Evenden (1990) also noted that relationships between the concentration ratio and soil concentration were dependent on the element under study. Hence, in general, plant:soil relationships for specific elements must be determined by consideration of a mixture of elements or compounds over a broad range of soil concentrations. However, in the case of plants and soils in the Marshall Islands, the small degree of environmental heterogeneity tends to enhance such relationships and makes the isolation of K and Cs relationships a relatively simple task.

The work of Konshin (1992) provided data on uptake and CRs for ^{137}Cs in grass sampled from areas of Belarus contaminated by the Chernobyl accident. Though that work was based in volcanic soils, they also reported a sharply decreasing and non-linear trend for the CR as a function of increasing soil concentration (see Fig. 5 of Konshin). In that work, the CR was found to be nearly inversely proportional to the soil concentration from about 1000 to 9000 Bq/kg of soil ^{137}Cs . The work of Konshin did not provide any mechanistic interpretation of the change in CR with soil concentration, but it was noted that the transfer of ^{137}Cs seemed to be more efficient at low soil concentrations.

McGee et al. (1996) evaluated the assumptions and usefulness of the ratio model for purposes of radioecology and dose assessment. Their data on ^{40}K and ^{137}Cs for two plant species in Ireland also indicated decreasing trends (though not logarithmic) with increasing soil concentration. Much of their data were acquired over relatively small ranges in soil concentrations and the authors commented that the observed decrease in CR is a mathematical artifact. However, the authors also state that the trends they observed agreed with those of Simon and Ibahim (1987) and Konshin (1992).

Bunzl et al. (2000) examined the relationship between ^{137}Cs concentrations in soils and plants from alpine pastures. They found that increased ^{137}Cs plant concentrations were not significantly associated with increased soil contents. Though they did not find a correlation between ^{137}Cs contents in plants and soil, they did document a statistically significant negative correlation between ^{137}Cs concentration in plants and the ^{40}K concentration in the underlying soil.

Knatko, Ageets, Shmigelskaya, and Ivashkevich (2000) evaluated concentrations of ^{137}Cs in soil and potato in areas of Belarus contaminated by the Chernobyl acci-

dent. They found a non-proportionate increase in ^{137}Cs plant concentrations with increasing soil concentrations and a decreasing trend in the CR with increasing soil ^{137}Cs as well as with K_2O . Their data (see Fig. 4A of Knatko et al., 2000) indicated that plant concentrations of ^{137}Cs were not directly proportional to soil concentrations: $P(^{137}\text{Cs}) \propto S^{0.55}$ and the $\text{CR} \propto S^{-0.45}$. Their values are similar to those presented here for all the Marshall Islands plants combined [$P(^{137}\text{Cs}) \propto S^{0.65}$ and $\text{CR} \propto S^{-0.35}$, see Table 4 of this report].

Smolders, Van Den Brande, and Merckx (1997) studied uptake of ^{137}CS in plants potted in a wide variety of soil types, though their soils did not include typical carbonate soils of atolls with the characteristically low clay content. They concluded that plant availability of ^{137}Cs varies due to differences in, among other things, ^{137}Cs retention in soil (affecting the supply to roots) and to differences in K availability. The subsequent work of Absalom et al. (1999, 2001) was primarily focused on modeling plant uptake of radiocesium based on soil characteristics, but was based on findings that radiocesium bioavailability is strongly influenced by K status and clay content as described by Smolders et al. (1997). Because of the dependence of the models of Absalom et al. on clay content, it has minimal applicability to plants of the Marshall Islands. Nevertheless, it is worthwhile to note that their model predictions were in moderately good agreement with a constant CR model (i.e., without dependence on ^{137}Cs soil concentration). At the same time, however, a significant proportion of the variability of plant concentrations was unaccounted for. Thus, it is clear that plant uptake is still only understood to a limited degree.

Two findings of our study were notable, though neither is easy to explain. First, one species, *P. scolopendria*, exhibited significantly greater uptake of ^{40}K and ^{137}Cs . Perhaps there are differences in the metabolic requirements of this plant, however, it is difficult to understand why ^{40}K would not have been absorbed more efficiently at higher soil concentrations in preference to ^{137}Cs . Second, plant uptake of ^{137}Cs was observed not to be strictly proportional to soil concentration though there was also high scatter in the data. Reasons to explain this phenomenon cannot be easily derived from our findings. Mechanisms that might limit plant uptake could be dependent on multiple factors. Some investigators have reasoned that such behavior is restricted to potassium-deficient soil systems, but this does not appear to have a firm basis since other investigators have made similar observations with other types of soils.

It is presently not known if ^{137}Cs concentrations in plants reach a limiting or saturation value as suggested by Simon and Ibahim (1987) or whether there might be multiple inflection points in the plant uptake data, indicating multiple saturation points along the soil concentration continuum. The plant uptake data shown in Fig. 9, under careful visual examination, has a structure that might be suggestive of such an effect, though it appears very difficult to statistically determine multi-phasic trends in the face of high variability.

6. Summary and conclusions

Matched plant and soil samples for eight plant types were obtained across a widely ranging ^{137}Cs concentration gradient in the soil. Uptake of ^{137}Cs was readily observable in more than 200 plant samples obtained from the atolls of the Marshall Islands. The sampling distributions of concentration values for six plant types were nearly indistinguishable. Only the liquid fraction of coconuts and the leaves of *P. scolopendria* appeared different. In general, the uptake into *P. scolopendria* is many times higher than into the other plants and the uptake into the liquid fraction of coconuts was many times lower. This finding agrees with observations of Duffy et al. (1999) from the same islands.

Our observations also indicated that the uptake of ^{137}Cs into plants with increasing ^{137}Cs in the soil was not proportionate and best described by a power function where the exponent of the soil concentration variable was consistently less than unity. These observations have a similarity to saturation behavior, as suggested by Simon and Ibahim (1987); saturation results in a decrease in the concentration ratio with increasing soil concentrations. Regardless of whether saturation in the plant concentration and in the CR is reached, there is considerable evidence to warrant questioning the validity of the simple model that describes the uptake of essential, and perhaps non-essential, elements into plants to be in direct proportion to the soil concentration. Our findings for ^{40}K and ^{137}Cs are similar to those of numerous other authors, despite the important ecological differences between coral-based soils and soils of volcanic origin.

We have found a simple, well-fitting power function to estimate either plant concentrations ($P = aS^b$) or CR values ($\text{CR} = aS^{b-1}$) from soil concentrations. In the case of uptake of ^{137}Cs , the exponent ($b-1$) for the CR for the various plant types ranged between -0.19 and -0.48 . Exponents of that magnitude would result in a 30–67% decrease in the CR for each tenfold increase in the soil concentration.

Variations in the CR values of the magnitude noted above are important to account for in dose and risk assessment models that attempt to provide realism as opposed to conservatism. Simple CR values often fail to realistically describe the uptake of ^{137}Cs into plants, usually by overestimating it at higher soil concentrations but also possibly by underestimating it at low soil concentrations. The data provided here are primarily applicable to coral-based soil that is typically low in potassium, organic matter, clay, and many nutrients. However, publications by other authors provide additional evidence in other soil systems that agree with the basic findings presented here. Hence, dose and risk assessments for which the uptake of ^{137}Cs into plants is an important component, should either be based directly on observed plant concentrations from the site under study, or should use predicted plant concentration values only after carefully considering the applicability of the particular plant uptake model used.

Acknowledgements

This research was funded by the Government of the Republic of the Marshall Islands under Section 177 of the Compact of Free Association. We are grateful to the numerous laboratory personnel of the Nationwide Radiological Study who assisted with sample collection, preparation, and measurement.

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