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# **<sup>137</sup>Cs Inter-Plant Concentration Ratios Provide a Predictive Tool for Coral Atolls with Distinct Benefits Over Transfer Factors.**

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## **Abstract**

Inter-plant concentration ratios (IPCR), [ $\text{Bq g}^{-1} \text{ }^{137}\text{Cs}$  in coral atoll tree food-crops /  $\text{Bq g}^{-1} \text{ }^{137}\text{Cs}$  in leaves of native plant species whose roots share a common soil volume], can replace transfer factors (TF) to predict  $^{137}\text{Cs}$  concentration in tree food-crops in a contaminated area with an aged source term. The IPCR strategy has significant benefits relative to TF strategy for such purposes in the atoll ecosystem. IPCR strategy applied to specific assessments takes advantage of the fact tree roots naturally integrate  $^{137}\text{Cs}$  over large volumes of soil. Root absorption of  $^{137}\text{Cs}$  replaces large-scale, expensive soil sampling schemes to reduce variability in  $^{137}\text{Cs}$  concentration due to inhomogeneous radionuclide distribution. IPCR [drinking-coconut meat (DCM) / *Scaevola* (SCA) and *Tournefortia* (TOU) leaves (native trees growing on all atoll islands)] are log normally distributed (LND) with geometric standard deviation (GSD) = 1.85. TF for DCM from Enewetak, Eneu, Rongelap and Bikini Atolls are LND with GSD's of 3.5, 3.0, 2.7, and 2.1, respectively. TF GSD for Rongelap copra coconut meat is 2.5. IPCR of *Pandanus* fruit to SCA and TOU leaves are LND with GSD = 1.7 while TF GSD is 2.1. Because IPCR variability is much lower than TF variability, relative sampling error of an IPCR field sample mean is up 6- to 10-fold lower than that of a TF sample mean if sample sizes are small (10 to 20). Other IPCR advantages are that plant leaf samples are collected and processed in far less time with much less effort and cost than soil samples.

*Keywords.*  $^{137}\text{Cs}$ ; Inter-plant concentration ratio; transfer factors; radionuclide prediction

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## **1. Introduction**

Plant to soil radionuclide transfer factors (TF) [ $\text{Bq kg}^{-1}$  in plants/ $\text{Bq kg}^{-1}$  in soil; both in dry weight] have been used for many years to predict concentrations of radionuclides that could be expected in food crops after accidental releases of radionuclides into the environment. TF have been developed from planned

53 greenhouse, pot, and small field experiments where various radionuclides are added to the soil, and from  
54 large-scale field experiments in various environments and ecosystems that have been contaminated by  
55 nuclear test programs, reactor accidents, or other major accidents. Many reports are available that deal  
56 with TF in various types of plants, soil, and ecosystems for both radionuclides and stable elements. Only a  
57 few are referenced here as examples (Al-Oudat et al., 2006; Rahman and Voigt, 2004; Frissel, M. J., et al.,  
58 2002; Robison et al., 2000; Papastefanou et al., 1999; Sheppard and Eveden, 1990). Various summaries of  
59 TF data (and aggregated transfer factors ATF, [Bq kg<sup>-1</sup> dry weight in food/ Bq m<sup>-2</sup> dry soil]) have been  
60 published over the years (Ng et al., 1982; IAEA, 1994; International Union of Radioecology (IUR) Data  
61 Base (2007)).

62  
63 TF are very useful in radiological dose-prediction models to estimate radionuclide concentrations in food  
64 crops after a release of radionuclides to the environment with a subsequent estimate of dose to exposed  
65 populations. However, in cases where prediction of radionuclide concentration in tree food crops in large  
66 contaminated areas with an aged source term are important, the soil sampling required to develop TF  
67 values for a specific radionuclide (or stable element) for tree food crops is time consuming and expensive.  
68 Moreover, the ratio of the highest to lowest TF values (or the ratio of the 95% confidence limits) can  
69 range from a factor of 10 to a factor 100 or more (Tome et al., 2003; Carini, F., 2001; IAEA, 1994;  
70 Sheppard and Eveden, 1990; Ng et al., 1982) leading to a large variance. Median or mean values of these  
71 wide-ranging data are generally used to predict radionuclide transfer from soil to plants for predictive dose  
72 assessments. There is also a large variance associated with TF data from coral atolls in the Marshall  
73 Islands (Robison et al., 2000; Robison and Conrado, 1997).

74  
75 About 90% of the estimated radiation dose to people returning to live on islands affected by U.S.  
76 atmospheric nuclear testing at Bikini and Enewetak Atolls results from consumption of locally grown  
77 foods that contain <sup>137</sup>Cs accumulated from the contaminated soil (Robison et al., 1997). The major intake  
78 of <sup>137</sup>Cs is mostly from tree crops such as coconut (*Cocos nucifera* L.), *Pandanus* fruit, and breadfruit.  
79 Drinking-coconut meat and fluid account for a large part of the dietary intake of <sup>137</sup>Cs at the atolls.  
80 However, *Pandanus* fruit, breadfruit, and to a lesser extent papaya and banana, also contribute. TF for  
81 these and many other plant species have been determined on Bikini and Eneu Islands at Bikini Atoll, eight  
82 islands at Enewetak Atoll, and Rongelap Island at Rongelap Atoll where these crops are growing in an  
83 environment contaminated with <sup>137</sup>Cs and <sup>90</sup>Sr. The ratio of high to low values observed in the range of TF  
84 data for <sup>137</sup>Cs and <sup>90</sup>Sr in food crops at Bikini is about a factor of 20 to 30 leading to a high variance in the  
85 TF data (Robison et al., 2000; Robison and Conrado, 1997; Robison et al., 1997).

86  
87 Much of the TF variability results from non-uniform spatial distribution of <sup>137</sup>Cs that occurred at time of  
88 deposition and subsequent redistribution due to anthropogenic activities on-island since the time of  
89 deposition. This is probably the case for most large-scale contaminations after a period of time. Roots  
90 from coconut, breadfruit, and *Pandanus* trees extend radially from the trees for 24 m or more. Any  
91 reasonable soil sampling scheme that includes both areal and depth distribution of the very large root zone  
92 of large fruit trees represents a very small fraction of the contaminated soil occupied by tree roots. This  
93 directly leads to a considerable variance in the data because of the very small area sampled when  
94 collecting soil relative to the very large area (and volume) of soil occupied by tree roots. Thus, a  
95 considerable range of results in TF is unavoidable.

96  
97 The International Union of Radioecologists (IUR) recently published the results of a web-based  
98 questionnaire to identify knowledge gaps on issues related to the protection of the environment (IUR,  
99 2006). A large cross-section of researchers from around globe are agreed that one of the highest research  
100 priorities in this field is to develop a better understanding of the transfer of radionuclides in the  
101 environment and, more specifically, on the need to understand and estimate uncertainties in radionuclide  
102 TF.

103  
104 Islands at the atolls other than the main residence islands are being considered for use as either potential  
105 residence and/or "agricultural" islands. Currently these islands do not have tree food-crops growing on  
106 them and the concentrations of <sup>137</sup>Cs and <sup>90</sup>Sr in the soil are different from islands such as Bikini, Eneu, or  
107 Rongelap where food crops are available and where TF have been determined. In such specific cases it is  
108 necessary to predict the concentrations of <sup>137</sup>Cs that will occur in tree food-crops to be planted on the

109 islands, and estimate the dose to people consuming the food crops, to help with resettlement and land-use  
110 decisions. A strategy that could eliminate the cost and effort associated with the tremendous amount of  
111 soil sampling required for TF development on these islands, and at the same time reduce the variance in  
112 the resulting product, would be very useful.

113  
114 Here we present data from the Marshall Islands that demonstrate the value of basing predictions of  
115 radionuclide concentration in tree food crops on inter-plant concentration ratios (IPCR) rather than TF  
116 between soil and plant. IPCR's are determined for paired trees - one being a tree food-crop the other a  
117 native, non-food tree that grow on all the islands at the atolls - whose root systems share a common  
118 volume of soil and radionuclide source for the majority of their roots. IPCR strategy applied to specific  
119 assessments takes advantage of the fact tree roots naturally integrate  $^{137}\text{Cs}$  over large volumes of soil. Root  
120 absorption of  $^{137}\text{Cs}$  replaces large-scale, expensive soil sampling schemes to reduce variability in  $^{137}\text{Cs}$   
121 concentration due to inhomogeneous distribution of radionuclides. The IPCR methodology is not designed  
122 to replace TF in large-scale predictive models that usually begin with deposition data ( $\text{Bq m}^{-2}$ , etc.). They  
123 would not work for such an application.

124  
125 All but a few of the pairs of trees in this study were separated by distances of merely 1 or 2 meters; there  
126 were only a few paired trees separated by as much 3 to 4 meters. We have traced the roots from specific  
127 trees to determine their radial distance from the trees (24 m or more) and have observed other tree roots  
128 from the nearby-neighbor trees in the same volume of soil (coconut roots, and *Scaevola* and *Tournefortia*  
129 roots, are readily identified). So by far, most of the absorbing roots of near-neighbor trees are in a  
130 common volume of soil and radionuclide pool. The roots systems are massive and the small portion of the  
131 roots of one tree that might be outside a common volume of soil is of no consequence.

132  
133 The IPCR is defined as the ratio of  $^{137}\text{Cs}$  concentration in the fruit of a food plant species (coconut,  
134 *Pandanus*, etc.) divided by the  $^{137}\text{Cs}$  concentration in the leaves of the paired native plant species.

## 135 136 **2. Methods**

### 137 138 **2.1 Sample collection, preparation, and analysis**

139 Thousands of coconut trees, planted on 8-m grid spacing, are growing on islands at Bikini, Enewetak, and  
140 Rongelap atolls. Drinking-coconut meat (coconuts at a young stage of development that contain fluid that  
141 is drunk and a meat the consistency of gelatin that is eaten) was obtained from coconut trees as one of the  
142 food crops because it is an important part of the local-food diet and contributes significantly to the  
143 radiation dose people receive from eating locally grown foods. The older copra stage of the coconut  
144 development where the meat is a white and firm is also used for flavoring in cooking, or occasionally  
145 consumed directly, but to a much lesser extent than the drinking stage.

146  
147 *Pandanus* fruit was selected as a second food item because it is common in the diet at the atolls. *Pandanus*  
148 trees grow wild and are randomly distributed around the islands. Some have been planted in defined areas  
149 around the island. Large, ripe fruit from these trees were collected for processing and analysis.

150  
151 Native trees that grow on all islands at all atolls in the Marshall Islands, and whose root systems are of  
152 similar radial scale and depth as coconut and *Pandanus* tree roots, include *Tournefortia argentea* (also  
153 known as *Argusia* and *Messerschmidia*) and *Scaevola taccada*. Actively growing leaves (no senescent  
154 leaves) were collected from these trees for comparison with drinking-coconut meat and *Pandanus* fruit.  
155 The number of paired trees found at the atolls for use in analysis of inter-plant correlation coefficients is:  
156 17 pairs of drinking-coconut and *Scaevola* trees and 18 pairs of drinking-coconut and *Tournefortia* trees  
157 were located on Bikini Island; 24 pairs of drinking-coconut and *Scaevola* trees and 17 pairs of drinking-  
158 coconut and *Tournefortia* trees were located on islands at Enewetak Atoll; 9 pairs of drinking-coconut and  
159 *Scaevola* trees and 6 pairs drinking-coconut and *Tournefortia* trees were located on two islands at  
160 Rongelap Atoll; 3 pairs of *Pandanus* and *Scaevola* trees were found on Bikini Island, and 3 pairs on  
161 Rongelap Atoll; 3 pairs of *Pandanus* and *Tournefortia* were found on Bikini Island and 1 pair on Aej  
162 Island at Enewetak Atoll.

164 Drinking-coconut meat, copra meat, and Pandanus fruit, and associated soil samples, have been collected  
165 over many years to develop TF data for hundreds of coconut trees at Bikini, Enewetak, and Rongelap  
166 Atolls and for a number of Pandanus trees at the atolls.

167  
168 Leaf and drinking-coconut meat samples collected on the islands were double bagged in the field, placed  
169 in large Matson freezer vans shortly after collection, and shipped frozen to Lawrence Livermore National  
170 Laboratory (LLNL). At LLNL vegetation samples were lyophilized to constant dry weight, ground to fine  
171 consistency, and packed in steel cans (233 cm<sup>3</sup>) for analysis of <sup>137</sup>Cs by gamma spectroscopy. Soil profile  
172 samples were collected in increments of 0-5 cm, 5-10 cm, 10-15 cm, 15-25 cm, and 25-40 cm from the  
173 sidewalls of trenches dug radially from the tree trunk for several meters. This provided a method for  
174 collecting uncontaminated samples to determine the distribution of <sup>137</sup>Cs with depth (significant for  
175 purposes of remediation) as well as the necessary data for TF calculations. Most all atoll tree roots  
176 involved in water and electrolyte absorption are in the top 40 cm of soil that contains nearly all organic  
177 material in the soil profile. There is very little organic matter from 40 cm to the water table at about 3 to  
178 3.5 m depth. Soil samples were oven dried at LLNL at low temperature to constant weight, then ball-  
179 milled to a fine powder and packed in steel cans (233 cm<sup>3</sup>) for analysis by gamma spectroscopy. The  
180 gamma spectroscopy facility (GSF) consists of 22 high-resolution, solid-state, intrinsic germanium  
181 detectors and the spectra are analyzed using the Canberra GENIE system. Details on calibration of the  
182 detectors were reported previously (Brunk, 1995; Hamilton et al., 2000).

183  
184 A set of samples submitted to the GSF included blind-duplicate samples and standard samples each  
185 totaling 10% of the number of submitted samples. Results for the standards had to be within 10% of the  
186 accepted standard value or re-analysis was required. Similarly, the blind-duplicate samples had to be  
187 within 10% of each other or else this triggered a re-analysis. No re-analyses were required in these data.

## 188 2.2 Statistical Analyses

189  
190 Measured coconut:leaf <sup>137</sup>Cs-concentration ratios  $R_{k,j,i}$  involving two leaf types ( $k = 1,2$ , denoting  
191 *Scaevola* and *Tournefortia*, respectively) each compared to corresponding samples of drinking-coconut  
192 meat from nearby coconuts gathered on eight different islands ( $j = 1, \dots, 8$ ), were analyzed for  
193 homogeneity. They were characterized statistically using  $n_{k,j}$  to denote the number of ratios measured for  
194 each leaf-type and island. Island-specific sets of  $\ln(\text{ratio})$  values were (if  $n_{k,j} > 3$ ) each assessed for  
195 consistency with a normal distribution using the Shapiro-Wilk test (Royston, 1992). Sample geometric  
196 mean (GM) and geometric standard deviation (GSD) values were calculated for each of these data sets,  
197 and Student's t-test (Kendall and Stuart, 1979) was done on the hypothesis that  $\ln(\text{ratio}) = 0$  (which is  
198 equivalent to the hypothesis that  $\text{GM} = 1$ ). For each test (Shapiro-Wilk and Student's t) the corresponding  
199 leaf-type and island-specific set of up to 8 p-values reported in columns 3 or 6 of Table 1 was adjusted for  
200 multiple independent tests using Hommel's Bonferroni-type adjustment algorithm (Wright, 1992). Each  
201 combined set of values,  $\ln(R_{k,j,i})/k$  for each leaf type, was assessed for homogeneity using the  
202 nonparametric Kruskal-Wallis test (Lehman and D'Abbrera, 1975). In any set(s) of leaf-specific values  
203  $\ln(R_{k,j,i})$  determined to be significantly nonhomogeneous, the small subset values of island-specific ratios  
204 with a GM found to differ significantly from 1 were adjusted by a common factor of  $1/\text{GM}$  determined for  
205 that data set (i.e., were normalized to have a  $\text{GM} = 1$ ). This procedure maximized the statistical power of  
206 variance estimation based on all available data despite the fact that *Tournefortia* ratios from Bikini and  
207 Rongelap were found to be  $< 1$  (see Section 4). Resulting data were combined into a single set of raw  
208 (and, as just described, potentially adjusted) ratios  $R_i$  for all islands and both leaf types, where  $i = 1, \dots, n$ ,

$$209 \text{ and } n = \sum_{k=1}^2 \sum_{j=1}^8 n_{k,j}.$$

210  
211 The combined set of  $\ln(R_i)$  values was assessed for normality by Shapiro-Wilk test and homogeneity by  
212 Kruskal-Wallis test, as measured by corresponding unadjusted p-values. Finally, parameters of a  
213 lognormal distribution fit to  $n$  combined measures  $R_i$  were then estimated by least-squares linear  
214 regression of the sorted log-ratios  $\ln(R_{(i)})$  against corresponding standard-normal (Van der Waerden)  $z$ -  
215 scores,  $z_i = F^{-1}(i/[n+1])$  for  $i = 1, \dots, n$ , where  $F^{-1}$  is the inverse cumulative standard normal distribution  
216 function. Consistency of the empirical distribution of combined  $\ln(\text{ratio})$  values with an estimated normal

217 distribution was also assessed using modified Kolmogorov-Smirnov (D), Camer-von-Mises ( $W^2$ ) and  
218 Watson ( $U^2$ ) statistics (Stephens,1970).  
219

220 The following procedure was used to compare variability among combined, adjusted sets of coconut:leaf  
221 (IPCR) ratios,  $R$ , to that among coconut:soil (TF) ratios measured on four different islands (Eneu,  
222 Enewetak, Rongelap and Bikini). A lognormal fit was obtained as described above for each island-  
223 specific set of  $\ln(\text{TF})$  values, after verifying consistency with the lognormal assumption. Some TF data for  
224 Eneu and Rongelap involved subsets of trees for which multiple measures were obtained over a period of  
225 time. Specifically, an average ( $\pm 1$  SD) of  $6 \pm 4$  measures/tree was made on each of 90 trees among 125  
226 trees studied on Eneu, and  $3.4 \pm 1.5$  measures/tree were made on each of 59 trees among 232 trees studied  
227 on Rongelap. Corresponding lognormal fits were obtained by weighted least-squares linear regression of  
228  $\ln(S_{(i)})$  on  $z_i$  (as explained above), using tree-specific sample numbers as weights. Finally, fitted  
229 regression slopes for island-specific TF data were compared to the fitted slope of the combined, adjusted  
230  $R$ -data by analysis of covariance (ANOCOVAR) for linear regression (Selvin, 1995), and corresponding  
231 variances were also compared using Bartlett's test (Snedecor and Cochran, 1989).  
232

233 Monte Carlo methods were used to compare the expected magnitudes of relative error (defined as the  
234 upper 2-tail 95% confidence limit, divided by the expected value) of sampled mean values of  $R$  vs. TF  
235 data. To do this, sample means were obtained for each of 5,000 simulated sets of  $k$  values, each sampled  
236 from a specified ( $R$ - or TF-specific) lognormal distribution, with  $k$  fixed at 10, 20 or 60.  
237

238 All calculations were done using *Mathematica* 5.2<sup>®</sup> and related software (Wolfram, 1999; Bogen, 2002).  
239

#### 240 241 4. Results 242

243 Coconut:leaf ratios of  $^{137}\text{Cs}$  concentration measured from *Scaevola* and *Tournefortia* leaves and  
244 corresponding nearby-coconut samples gathered at eight different Marshall Islands locations are  
245 summarized in Table 1. *Scaevola* ratios obtained from each island are consistent with null hypotheses  
246 concerning log-normality and  $\text{GM} = 1$  (Table 1), and combined log-ratio values for this leaf type from all  
247 islands are statistically homogeneous ( $p = 0.12$ ). *Tournefortia* ratios from each island are consistent with  
248 null hypotheses concerning log-normality and  $\text{GM} = 1$ , except with respect to the latter hypothesis for  
249 ratios from Bikini ( $p_{\text{adj}} = 2.0 \times 10^{-6}$ ) and possibly those from Rongelap ( $p_{\text{adj}} = 0.096$ ,  $p_{\text{non-adj}} = 0.016$ ) (Table  
250 1). The combined log-ratio values for *Tournefortia* from all islands are inconsistent with a null  
251 homogeneity hypothesis ( $p = 0.0071$ ), but are quite consistent with this hypothesis when ratios from  
252 Bikini and Rongelap were dropped ( $p = 0.20$ ). After the sets of *Tournefortia* ratios from Bikini and from  
253 Rongelap were each scaled by factors  $1/\text{GM}$  using the respective island-specific GM estimates listed in  
254 Table 1, and the ratios for both leaf types from all islands were then combined, the 109 combined ratios  
255 became consistent with null hypotheses concerning log-normality ( $p = 0.79$ ),  $\text{GM} = 1$  (0.24), and  $\ln$  ratio  
256 homogeneity ( $p = 0.10$ ) (Table 1).  
257

258 The lognormal model fit to the combined data (Figure 1) corresponds to the following estimated values  
259 (and corresponding 95% confidence limits) for the geometric mean and geometric standard deviation:  $\text{GM}$   
260  $= 0.935$  (0.928, 0.942),  $\text{GSD} = 1.85$  (1.83, 1.86). The empirical distribution of combined  $\ln$  ratio values  
261 was found to be consistent with the corresponding estimated normal distribution based on modified  
262 Kolmogorov-Smirnov (D), Camer-von-Mises ( $W^2$ ) and Watson ( $U^2$ ) statistics ( $p > 0.15$ , all three tests).  
263 Note that for the purpose of estimating confidence limits on GM, the t-test done on combined  $\ln$  ratio  
264 values was not as powerful statistically as the linear regression done involving these values.  
265 Consequently, while the t-test did not detect statistically significant deviation from  $\text{GM} = 1$ , the regression  
266 did.  
267

268 Results obtained imply a GSD of approximately 1.85 for the combined ratios of  $^{137}\text{Cs}$  in drinking-coconut  
269 meat to that in *Scaevola* and *Tournefortia* leaves, with *Scaevola* leaves yielding ratios found to have GM  
270 values consistent across all islands, and *Tournefortia* leaves somewhat less so.  
271

272 TF data obtained on Eneu, Enewetak, Rongelap, and Bikini are shown with corresponding lognormal fits  
273 (light lines) in Figure 2, and for comparison the combined  $R$ -data and fit (bold line) from Figure 1. Each  
274 island-specific set of  $\ln(\text{TF})$  data was found to be approximately normally distributed ( $p > 0.01$ , by  
275 Shapiro-Wilk test). The island-specific log-TF slopes (corresponding to GSD values ranging from 2.1 to  
276 3.5) and corresponding variances all differ significantly from one another ( $p < 10^{-10}$ , by ANOCOVAR),  
277 and all are significantly larger ( $p < 10^{-10}$ , by ANOCOVAR and by Bartlett's test) than that obtained based  
278 on the combined, adjusted multi-island  $R$ -data (corresponding to GSD = 1.85). Consequently, TF data  
279 gathered from all four islands indicate that TF variability is greater or substantially greater (on all island  
280 except Bikini) than leaf-based measures of  $^{137}\text{Cs}$  uptake into drinking coconut meat.

281  
282 IPCR of *Pandanus* fruit (another contributor of  $^{137}\text{Cs}$  in the diet) to *Scaevola* and *Tournefortia* leaves (10  
283 data points) also are log-normally distributed with a GM of 4.1 and GSD of 1.7. Corresponding TF data  
284 for nine *Pandanus* trees has a GM of 1.5 and GSD of 2.1, again indicating considerably greater variability  
285 using the TF rather than leaf-based approach to estimating dietary  $^{137}\text{Cs}$  based on radioecology data.

## 287 5. Discussion

288  
289 Results obtained in this study indicate substantial statistical homogeneity of leaf:coconut ratios of  $^{137}\text{Cs}$   
290 concentration measured from *Scaevola* and *Tournefortia* leaves and corresponding nearby-coconut  
291 samples gathered from three atolls and eight different islands in the Marshall islands. Only *Tournefortia*  
292 ratios obtained from Bikini and Rongelap appeared to be clearly, or possibly, sampled from populations  
293 with a GM significantly  $< 1$ . After adjusting for this source of bias, the combined set of 109 measures  
294 involving two leaf types from a total of eight islands were found to be statistically homogeneous and log-  
295 normally distributed with a GM of 0.935 and a GSD of 1.85. By the method of moments (Aitchison and  
296 Brown, 1957), it is readily shown that such a lognormal distribution has an expected value of  $m = 1.13$ .

297  
298 These findings support the general conclusion that  $^{137}\text{Cs}$  concentration  $C$  in drinking-coconut meat  
299 throughout the Marshall Islands may be estimated reliably by sampling  $^{137}\text{Cs}$  concentration  $S$  in a  
300 sufficient number of *Scaevola* or *Tournefortia* leaves, and multiplying it by the expected value ( $m = 1.13$ )  
301 from the IPCR data,  $C = S * 1.13$ . Although geographic variability in  $S$  is log-normally distributed with a  
302 GSD of 1.85, the sample mean of  $S$  is an unbiased estimator of the expected value of  $S$ , regardless of the  
303 number ( $s$ ) of samples of  $S$  taken. However, for small sample size  $k$ , substantial statistical uncertainty will  
304 be associated with the sample mean of  $S$ . For example, relative error of the estimated arithmetic mean of  
305  $S$  would be 4.1%, 12%, or 21% for sample sizes of  $k = 60$ ,  $k = 20$ , or  $k = 10$ , respectively. In contrast, this  
306 relative error would be as high as 68% ( $k = 20$ ) or 210% ( $k = 10$ ) using the TF method on an island like  
307 Enewetak (GSD = 3.48) which is a 6- to 10-fold greater relative error using TF as compared the IPCR  
308 based approach, if only 10 to 20 samples are taken.

309  
310 To reduce uncertainty in  $^{137}\text{Cs}$  TF for trees requires more and more soil sampling to try to cover the very  
311 large area encompassed by the absorbing root zone of large fruit trees. To do this requires an enormous  
312 amount of work, time, and expense. In the IPCR strategy the massive roots of the trees do the integrating  
313 that one is trying to accomplish with very extensive soil sampling and they do it far better than any  
314 reasonable soil-sampling scheme. This is the primary reason that the variance in the IPCR data is much  
315 lower than the TF data.

316  
317 Also, the processes of radionuclide uptake between the plant roots and the soil are very complex due to  
318 soil properties such as pH, clay content, CEC, K and Ca concentration, etc. and can greatly affect the  
319 magnitude of the TF. Thus, a slight difference in soil composition from one location to another can be a  
320 problem when TF developed in one location is applied in another location. As a result, a lot of time-  
321 consuming work is required to collect soil samples in every new location. The IPCR strategy does not deal  
322 with those issues in any direct way. That of course is the benefit of the strategy – it simply reflects the end  
323 result of all those complex processes. Roots of the two different types of trees are looking at the same soil.  
324 Even though the detailed process of  $^{137}\text{Cs}$  (or any other radionuclide) uptake by the roots may differ in  
325 some ways between the two species of trees, it is reflected in the ratio of the  $^{137}\text{Cs}$  concentration in the  
326 food and non-food trees. The  $^{137}\text{Cs}$  concentration in each of the trees reflects the end result of whatever



327 processes lead to the uptake into the plants and would be consistent relative to each other no matter the  
328 location.

329  
330 Other advantages of the IPCR strategy are that it is much less time consuming, less effort, and less costly  
331 to generate the required data compared with soil sampling required to develop TF. Leaf samples are vastly  
332 easier and far quicker to collect than soil samples. Moreover, weight of leaf samples required is but a tiny  
333 fraction of the weight of required soil so they are much easier to handle and transportation costs are  
334 greatly reduced. Finally, because processing of leaf samples for submission to the analytical laboratories is  
335 simpler and faster than for soil, data are more rapidly attained.

336  
337 Annual and integral dose estimates for people consuming tree food- crops (coconuts and *Pandanus* fruit)  
338 that contain  $^{137}\text{Cs}$  can be made for a specific island based on a sampling and analysis of  $^{137}\text{Cs}$  in *Scaevola*  
339 and/or *Tournefortia* leaves from that island. Observed variance in the IPCR data for  $^{137}\text{Cs}$  concentration in  
340 both drinking coconuts and *Pandanus* fruit presented in this paper can be used to generate the uncertainty  
341 and individual variability in annual and integral doses (Robison et al., 1997; Bogen et al., 1997). This  
342 proposed application of IPCR data, particularly for small sample sizes, would result in dose estimates with  
343 less uncertainty than what would be generated using TF data.

344  
345  
346 As with TF data, the IPCR cannot be used immediately after deposition but only after a reasonable steady  
347 state has been developed between the radionuclide deposition on the soil and uptake by plants of the  
348 radionuclide in question. But that is the case when interest is in the longer-term consequences of a major  
349 deposition of radionuclides in the environment such as in the Marshall Islands. Also, the IPCR's are not  
350 designed to replace TF in large-scale predictive models that usually begin with deposition data ( $\text{Bq m}^{-2}$ ,  
351 etc.). They would not work for such an application.

352  
353 As a result of the Chernobyl accident (and other sources of contamination) there may be contaminated  
354 areas around the world where the IPCR method could be used to more easily develop predictions of  
355 radionuclide concentrations in tree food-crops in alternate locations rather than develop TF data. The only  
356 requirement is that roots of the native, non-food tree have the same general distribution of roots, aerally  
357 and with depth, as the food trees. The advantages are significant if the situation is appropriate.

## 358 359 **6. Conclusions**

360  
361 Inter-plant concentration ratios (IPCR) can be used to predict radionuclide concentrations in tree food  
362 crops from one location to another at coral atolls. The massive roots of the trees do the integrating in the  
363 IPCR strategy that one is trying to accomplish with very extensive soil sampling in the TF method in order  
364 to reduce uncertainty (in IPCR and TF) due to the inhomogeneous distribution of radionuclides in the soil.  
365 Also, any difference in uptake of  $^{137}\text{Cs}$  due to soil composition from one location to another is reflected in  
366 a similar way in both the food trees and the native trees. Variance in IPCR data is one half to one third that  
367 for corresponding transfer factor (TF) data developed in the same atoll ecosystem. Other IPCR method  
368 advantages are that leaf samples can be collected and processed in far less time with much less effort and  
369 cost than soil samples. Thus, final data can be obtained more efficiently and rapidly. This approach has  
370 important implications in providing guidance for resettlement and/or use of former test sites in the  
371 northern Marshall Islands and may be applicable to other radioactively contaminated regions or accident  
372 sites where tree food-crops are important in the diet.

## 373 374 375 376 **Acknowledgment**

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472 **Figure 1.** Lognormal fit to 109 combined measures of coconut:leaf  $^{137}\text{Cs}$  ratio (ICPR) for *Scaevola* and  
473 *Tournefortia* leaves and corresponding nearby-coconut samples, gathered on eight different islands. Data  
474 were adjusted for suspected downward bias in 18 and 3 *Tournefortia* ratios sampled on Bikini and  
475 Rongelap, respectively. The sorted ratios  $R_{(i)}$  and corresponding standard-normal scores,  $z_i = F^{-1}(i/[n+1])$   
476 for  $i = 1, \dots, n$  where  $F^{-1}$  is the inverse cumulative standard normal distribution function, are plotted (open  
477 points) together with the linear fit of  $\ln(R_{(i)})$  on  $z_i$  (line) and corresponding squared coefficient of  
478 correlation ( $R^2$ ). The model-parameter estimates ( $\pm 1$  SE),  $m = -0.067 \pm 0.004$  and  $s = 0.614 \pm 0.004$ ,  
479 correspond to an estimated geometric mean  $\text{GM} = 0.935$  and geometric standard deviation  $\text{GSD} = 1.85$ ,  
480 where  $m$  and  $s$  here denote  $\log(\text{GM})$  and  $\log(\text{GSD})$ , respectively.  
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483 **Figure 2.** Lognormal fits to island-specific sets of values of coconut:soil  $^{137}\text{Cs}$  ratio (TF) measured for  $n$   
484 different trees on three islands (Eneu, Enewetak and Rongelap), compared to coconut:leaf  $^{137}\text{Cs}$  ratio  
485 (ICPR) values and corresponding lognormal fit (bold line) obtained for multiple islands (from Figure 1).  
486 Corresponding island-specific model-parameter estimates ( $\pm 1$  SE) are:  $m = 2.19 \pm 0.013$ ,  $s = 1.25 \pm 0.013$ ,  
487  $\text{GSD} = 3.48$ ,  $R^2 = 0.988$ ,  $n = 125$  (Eneu);  $m = 2.10 \pm 0.014$ ,  $s = 1.11 \pm 0.015$ ,  $\text{GSD} = 3.03$ ,  $R^2 = 0.980$ ,  $n =$   
488  $115$  (Enewetak); and  $m = 0.423 \pm 0.006$ ,  $s = 1.01 \pm 0.006$ ,  $\text{GSD} = 2.73$ ,  $R^2 = 0.992$ ,  $n = 232$  (Ronelap).  
489 See Figure 1 legend for notation.  
490

491 **Figure 3.** Lognormal fits to island-specific sets of values of coconut:soil  $^{137}\text{Cs}$  ratio (TF) measured for  $n$   
492 different trees on Bikini, compared to coconut:leaf  $^{137}\text{Cs}$  ratio (ICPR) values and corresponding lognormal  
493 fit (bold line) obtained for multiple islands (from Figure 1). Corresponding Bikini-specific model-  
494 parameter estimates ( $\pm 1$  SE) are:  $m = 2.51 \pm 0.008$ ,  $s = 0.742 \pm 0.008$ ,  $\text{GSD} = 2.10$ ,  $R^2 = 0.987$ ,  $n = 106$   
495 (Bikini). Note that the Bikini TF slope is significantly greater than the multi-island IPCR slope ( $F_{1,211} =$   
496  $190.0$ ,  $p \approx 0$ , by ANOCOVAR). See Figure 1 legend for notation.  
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**Table 1.** Summary of coconut:leaf (IPCR) ratios of <sup>137</sup>Cs concentration measured in the Marshall Islands.

$k =$ Leaf type $j =$ Island		Number of measured ratios, $n_{k,j}$	Normality <sup>a</sup> of log-ratios, $p_{adj}$	Geometric mean, GM	Geometric standard deviation, GSD	Likelihood <sup>a</sup> that GM = 1, $p_{adj}$
<i>1. Scaevola</i>						
1	Aej	6	0.99	0.899	1.91	0.97
2	Aomen	8	0.90	0.978	2.40	0.97
3	Bijire	8	0.99	1.02	1.79	0.97
4	Bikini	17	0.99	0.429	1.66	0.97
5	Kabelle	4	0.99	0.655	1.25	0.97
6	Lojwa	4	0.96	1.21	1.93	0.97
7	Lujor	6	0.99	0.549	1.71	0.97
8	Rongelap	7	0.99	0.330	1.28	0.97
<i>2. Tournefortia</i>						
1	Aej	5	0.32	0.899	1.91	0.97
2	Aomen	3	—	0.978	2.40	0.97
3	Bijire	8	0.054	1.02	1.79	0.97
4	Bikini	18	0.32	0.429 <sup>b</sup>	1.66	2.0×10 <sup>-6</sup>
5	Kabelle	3	—	0.655	1.25	0.20
6	Lojwa	3	—	1.21	1.93	0.97
7	Lujor	6	0.32	0.549	1.71	0.21
8	Rongelap	3	—	0.330 <sup>b</sup>	1.28	0.096
<i>Both leaf types</i>						
1-8	All	109	0.79	0.935 <sup>b</sup>	1.85	0.24

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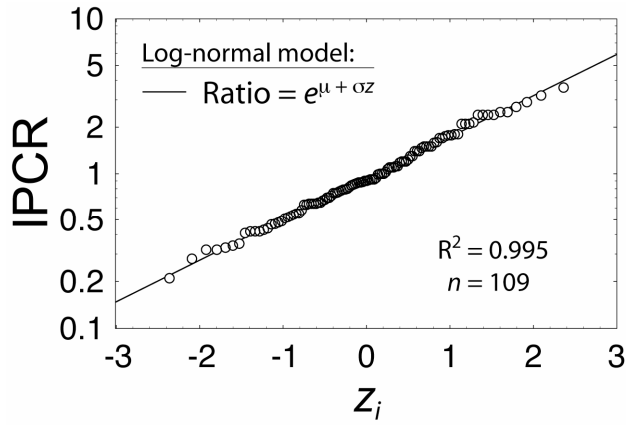
<sup>a</sup> P-values are listed for the Shapiro-Wilk test for normality and the Student's t- test hypothesis that  $\ln(\text{ratio}) = 0$ , for leaf-type- and island-specific sets of  $\ln(\text{ratio})$ . Each leaf-type and island-specific p-value was adjusted for multiple independent tests involving the same leaf type. Corresponding unadjusted p-values are shown for tests on combined data for both leaf types from all islands.

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<sup>b</sup> *Tournefortia* ratios for Bikini and Rongelap were each normalized by the factor  $1/\text{GM}$ , where GM is island specific, to adjust for suspected downward bias pertaining to measures from those islands as indicated by low values of  $p_{adj}$  from corresponding t-tests described above.

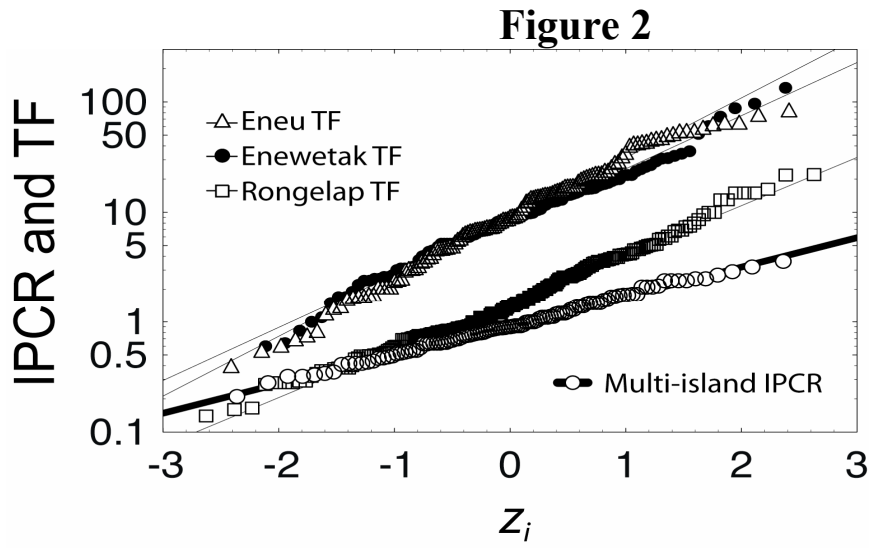
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**Figure 1**



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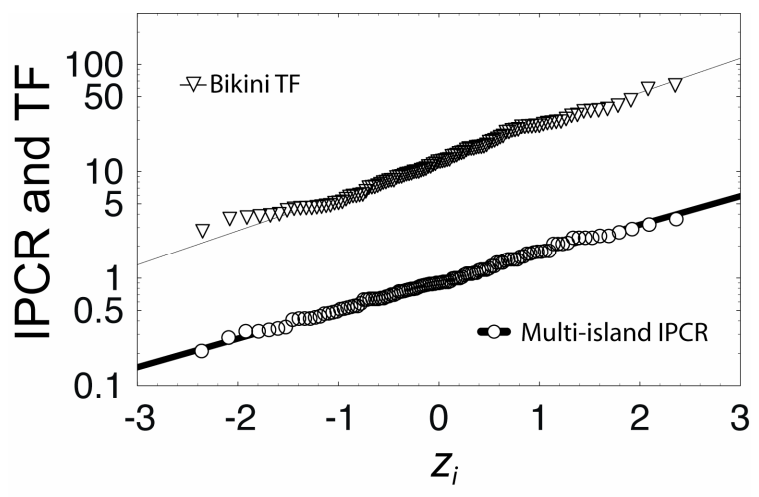
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**Figure 3**



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