



In situ measurement of cesium-137 contamination in fruits from the northern Marshall Islands

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Radioactive contamination of fruits in the northern Marshall Islands, resulting from the US nuclear weapons testing program in the 1940s and 1950s, is still a human health concern, in particular pertaining to island population resettlement and the economic benefit from farming. Over 200 fruits, primarily coconuts and pandanus, were collected on 11 islands from four atolls in the northern Marshall Islands in 2017. The energy spectra from nuclear gamma decays were measured on a research vessel for each fruit in situ. From these recordings, the level of cesium-137 (¹³⁷Cs) contamination was determined for individual fruits. Comparisons of the results are made to past studies and international food safety standards. There is a broad distribution of values, ranging from below detectable radiation levels to relatively high levels; safety concerns are largest for Bikini Island. A noticeable fraction of fruits from Bikini have significantly higher levels of ¹³⁷Cs contamination compared with those from all other measured islands.

Marshall Islands | food | radiation | cesium-137 | Bikini

The United States conducted 67 nuclear tests in the Republic of the Marshall Islands (RMI) between 1946 and 1958. These tests consisted of a large assortment of fission and fusion devices, with an enormous range of energy yields (up to 15 megatons), including 18 tests with energy yields over a megaton (1). Radioactive fallout from the testing program contaminated a huge swath of land and ocean, extending over a million square kilometers, and severely impacted the habitability of the affected atolls. Enewetak and Bikini Atolls, used as ground zero for the tests, as well as neighboring Rongelap and Utirik Atolls, were exposed to high levels of fallout and have been formally recognized as “nuclear atolls” (2, 3). See *SI Appendix, Fig. S1* for a map indicating their locations. Smaller northern atolls, such as Rongerik, Ailinginae, Ailuk, and Likiep, were also impacted (4). Inhabitants of Enewetak and Bikini Atolls were evacuated before the tests, but Rongelap and Utirik Atolls were inhabited during that time and were affected by the fallout from the largest nuclear weapon ever tested by the United States, Castle Bravo. Rongelap and Utirik inhabitants suffered severe health consequences from radiation poisoning, including death, and many of the health effects continue to affect those present on the islands at the time of the testing, as well as their descendants (5, 6).

The intervening years featured a complicated history of further evacuations, island resettlements, radiological cleanup, and even more evacuations. While Enewetak and Utirik today host populations of ~660 and ~410, respectively, Bikini and Rongelap Atolls are yet to be resettled. An in-depth understanding of the current radiological conditions is essential for informed decision making by the Marshallese people in regards to resettlement and future use of these lands, as well as to ensure the safety of the Marshallese who live in the northern atolls today.

Current external gamma radiation levels in Bikini, Enewetak, and Rongelap Atoll were assessed by our group in 2015 (7). We found that external gamma radiation levels on Bikini Island remain, on average, above 100 mrem/y, which was the total radiation exposure standard agreed upon by the United States and

the RMI governments for resettlement (8). According to our study, Rongelap Island had external gamma radiation levels below this standard, and several islands on Enewetak Atoll had levels of external gamma radiation that were indistinguishable from Majuro Island in the southern RMI, which we had designated as a control island. However, because the total radiation dose must be considered, rather than just external gamma radiation, it is critical to examine other exposure pathways, including ingestion.

Radiological contamination of the local food sources is one of the most important health hazards from the nuclear weapons testing program. Among the different radionuclides that result from a nuclear explosion, ¹³⁷Cs is most relevant to food contamination and internal exposure. For example, huge efforts have been made to regulate and reduce ¹³⁷Cs in food in Japan following the Fukushima Daiichi accident in 2011, including making over a million measurements since the accident (9, 10). Due to its extreme solubility, ¹³⁷Cs binds strongly to soil and does not travel far beneath the surface, being absorbed by trees and plants, and consequently transferred into their fruit (11). Additionally, due to its relatively long half-life of 30.1 y, ¹³⁷Cs has a tendency to bioaccumulate in plant tissue, thus prolonging exposure to radiation and becoming a long-term health concern (11). In fact, internal radiation exposure to the radionuclide ¹³⁷Cs via food ingestion has previously been estimated to contribute 85 to 90% of the total radiation exposure to the Marshallese living in the northern atolls (12). The ¹³⁷Cs nucleus forms as a nuclear-fission product

Significance

The United States performed nuclear testing on Bikini and Enewetak Atolls in the northern Marshall Islands between 1946 and 1958. Fallout from the largest test Bravo, detonated in 1954, spread over a large area, exposing to radiation not only land and ocean but also Marshallese people living in neighboring atolls, including Rongelap and Utirik. Cesium-137, present in the fallout, has a half-life of approximately 30 y and is readily absorbed by food crops, thus representing a health hazard for island inhabitants. In situ measurements of cesium-137 content were made for fruits from 11 islands on four atolls. Contamination remains above limits set by international safety standards in some measured fruits, and several islands display contamination from this human-made radionuclide.

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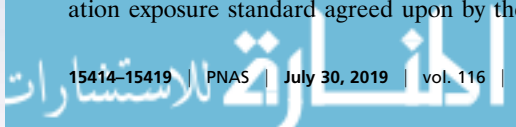
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of uranium-235 from nuclear weapon explosions, authorized discharge of nuclear waste, or accidental release from nuclear facilities (13). Thus, our study provides an indication of the lasting effects of the US nuclear weapons testing program in the RMI.

Accurate measurement of radiation in food requires sensitive radiation detectors, as well as substantial heavy shielding to differentiate fruit signal from background radiation. Several studies conducted by scientists at the Lawrence Livermore National Laboratory (LLNL), published between 1981 and 2013, report on food radiation levels in the northern Marshall Island (14–16). In these studies, fruit samples were collected on the islands and were subsequently cut and frozen aboard the ship for shipment to LLNL for analysis. Most analyzed coconut samples were composites of 4 to 8 coconuts, and the coconut meat and juice inside were separated for analysis.

In 2017, our team conducted a second research trip to the northern Marshall Islands to assess concentrations of the radionuclide ^{137}Cs in a total of 241 fruits, including coconuts and pandanus, the primary staples of the Marshallese diet (see *SI Appendix*, Fig. S3 for photographs of the fruits). The fruits were collected from 11 islands on four different atolls (Bikini, Utirik, Rongelap, and Enewetak). Unlike previous LLNL studies, we recorded spectra from a homogenous mixture of meat and juice in individual fruits aboard the research vessel shortly after collection, performing all measurements in situ. We present and discuss ^{137}Cs activity concentrations in the different samples across the islands and compare the results to various safety standards, some of which were developed in response to the Chernobyl and Fukushima accidents. There is some variation in safety standards and ^{137}Cs contamination limits for food, suggesting that the health impact of low radiation doses in food is not universally agreed upon. While translating contamination data into human health impact is still challenging (10), these data can shed some light on safety conditions as they pertain to habitation and resettlement decisions for the Marshallese people.

Results and Discussion

Raw Data. Fruits were collected from Enewetak, Ikuren, Japtan, and Medren Islands in Enewetak Atoll; Bikini and Enyu Islands in Bikini Atoll; Rongelap and Naen Islands in Rongelap Atoll; and Utirik, Aon, and Eluuk Islands in Utirik Atoll. The ^{137}Cs contamination in numerous fruits was easily identifiable from raw spectra. Fig. 1 shows characteristic spectra of fruit with (Fig. 1, *Left*) high concentration of ^{137}Cs , as indicated by a large peak at 662 keV, and (Fig. 1, *Right*) no measurable concentration of ^{137}Cs , lacking a recognizable peak at 662 keV. The peak at 1,461 keV corresponds to the signal from the potassium-40 (^{40}K) calibration ring, present inside the lead shield of the detector system during all measurements. The ^{40}K signal is used for relative calibration purposes.

Cesium-137 Levels.

Enewetak Atoll. We measured the spectra of 60 fruits from Enewetak Atoll and found no ^{137}Cs contamination in any of the samples within the sensitivity of our detectors. Our sensitivity for zero contamination was typically below 25 Bq/kg. The dominant uncertainty for fruits with low ^{137}Cs contamination comes from the uncertainty in the background subtraction. Spatial distributions of where samples were collected on various islands are shown in Fig. 2. The color coding of sample locations is described in *Comparison with International Standards*. All of the samples on Enewetak are denoted in green, indicating that the levels observed were below all standards, as reported in Table 1. Worth noting is that we only studied fruits from islands on the southern side of Enewetak Atoll. It is likely that fruits in the northern islands, such as Enjebi Island, could contain high levels of ^{137}Cs , as external gamma radiation levels in Enjebi are significantly higher than background (17). This is presumably due to radiation fallout that had either not been cleaned up in the northern Enewetak islands or had been higher in the north to begin with.

Bikini Atoll. The largest number of fruits in our study came from Bikini and Enyu Islands in Bikini Atoll, where we collected 47 fruits per island. These two islands are separated by just 20 km and had the highest levels of ^{137}Cs contamination in fruits. We found an average of 630 Bq/kg for Bikini Island, with an SD of 860 Bq/kg, as a result of several very high values (maximum of 3,770 Bq/kg). We also found an average value of 85 Bq/kg for Enyu Island, with an SD of 167 Bq/kg, also due to a few large values, including a maximal value of 1,150 Bq/kg. Fig. 3A shows the comparison of the distributions of coconuts vs. pandanus on Bikini Island. Pandanus have higher contamination levels compared with coconuts; a Wilcoxon rank sum test results in a P value of less than 0.001, suggesting that the contamination levels in the two different fruit types are significantly different. Fig. 3B shows the comparison of the distributions of fruit in Bikini and Enyu Islands; their close proximity to each other also made for an interesting comparison. One clearly sees that contamination levels are higher on Bikini Island. The difference in radiation level between these two islands is significant ($P < 0.001$).

Spatial distributions showing where samples were collected on different islands are given in Fig. 2. The color coding of sample locations is described in *Comparison with International Standards*. None of the 47 fruits on Bikini is color-coded green, meaning that they all exceed the standards set by the International Physicians for the Prevention of Nuclear War (IPPNW) and Belarus, Russia, and Ukraine standards for infant foods. Moreover, only 3 fruits out of 47 fruits were below the standards set by Japan for all foods, while all of the rest were above it. Thirteen fruits exceeded the standard set by the European Union (600 Bq/kg; orange), while five fruits had contamination levels far above even

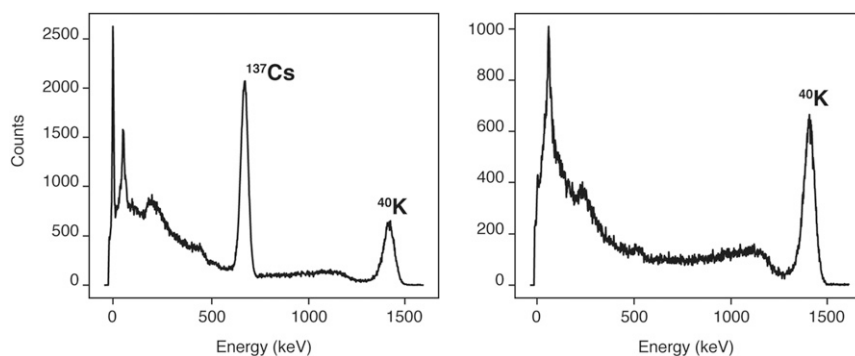


Fig. 1. Spectrum of a fruit from Bikini Island with a high concentration of ^{137}Cs (*Left*) and spectrum of a fruit from Utirik Island, with a low concentration of ^{137}Cs (*Right*). The peak at 662 keV in the Bikini fruit spectrum is a clear indication of the presence of ^{137}Cs .

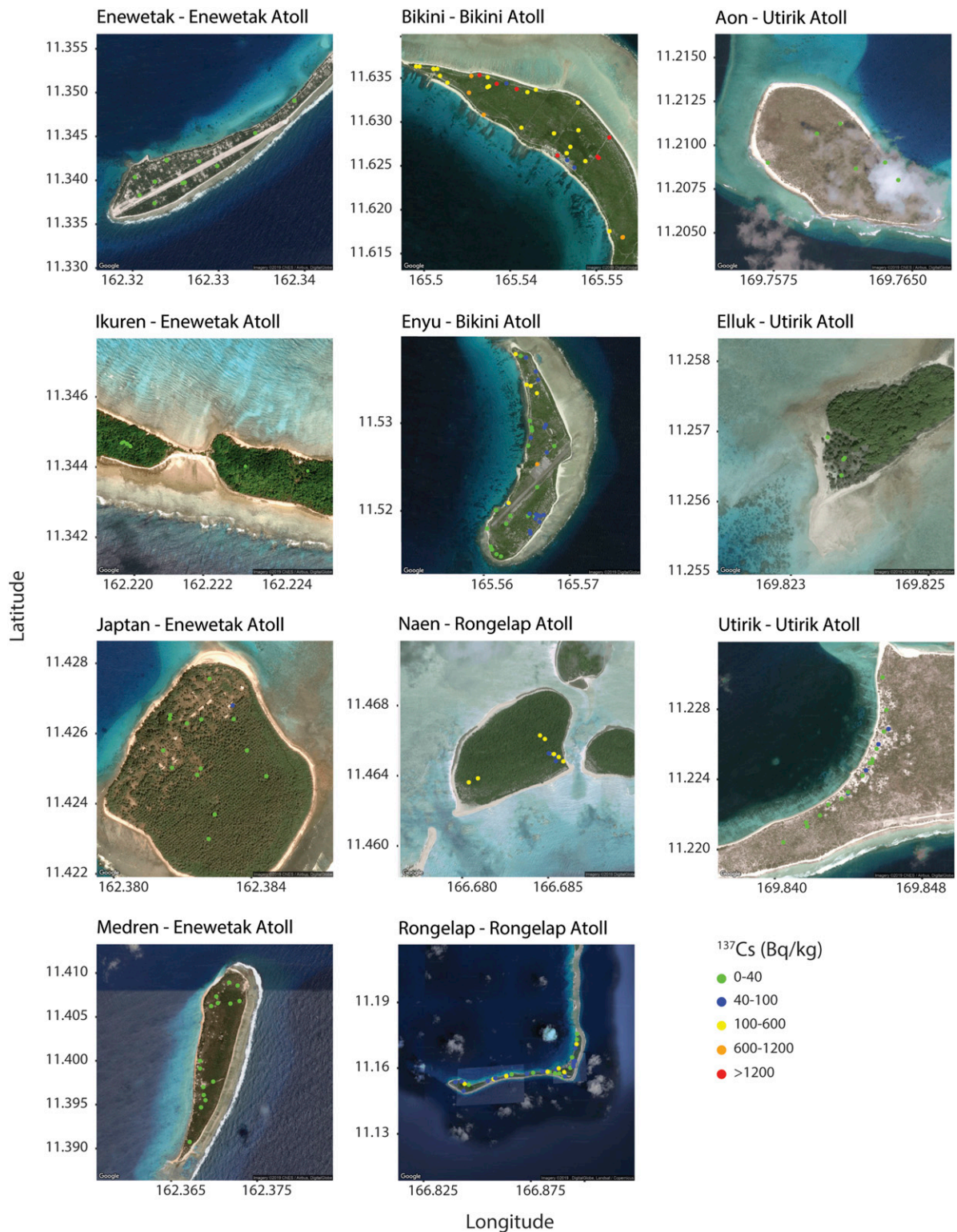


Fig. 2. Location of measured fruits on Enewetak Atoll (Left: Enewetak, Ikuren, Japtan, and Medren), Bikini Atoll (Middle: Bikini and Enyu), Rongelap Atoll (Middle: Naen and Rongelap), and Utirik Atoll (Right: Aon, Elluk, and Utirik). Color coding of each measured fruit was done according to the levels specified by international standards listed in Table 1: green (0 to 40 Bq/kg), blue (40 to 100 Bq/kg), yellow (100 to 600 Bq/kg), orange (600 to 1,200 Bq/kg), and red (above 1,200 Bq/kg), as presented in the legend.

the highest safety limits (above 1,200 Bq/kg; red). Our average value is above standards set by all countries and organizations, except for the US Food and Drug Administration (FDA) and the *Codex Alimentarius*.

Rongelap Atoll. We determined ^{137}Cs levels in 57 fruits total on Rongelap and Naen Islands and found a mean value of 67 Bq/kg with an SD of 62 Bq/kg for Rongelap Island and a maximum value of 350 Bq/kg. We found a mean value of 139 Bq/kg and SD

Table 1. International standards by country and organization, including specified food types, for ^{137}Cs contamination levels in units of becquerels per kilogram

Food	IPPNW, 1996	Belarus, current	Russia, current	Ukraine, current	Japan, current	European Union	1994 Codex	IAEA	FDA
Infant food	8	37	40–60	40	50	370	1,000	1,000	1,200
Noninfant food	16	NS	NS	NS	100	600	1,000	1,000	1,200
Milk	16	100	100	100	50	370	1,000	1,000	1,200
Fruit	16	40–100	40–120	40–70	100	600	1,000	1,000	1,200
Berries	16	185–370	160–500	500	100	600	1,000	1,000	1,200
Mushroom	16	2,500	2500	2500	100	600	1,000	1,000	1,200
Bread, cereal	16	40	40–60	20–50	100	600	1,000	1,000	1,200
Meat	16	180–500	160	200	100	600	1,000	1,000	1,200
Fish	16	150	130	150	100	600	1,000	1,000	1,200

NS, not specified.

of 105 Bq/kg for Naen Island, including a maximum value of 536 Bq/kg. As with the comparison between Bikini and Enyu islands, the difference between Rongelap and Naen Islands is statistically significant, with Naen fruits having higher contamination levels than those collected on Rongelap ($P < 0.001$). This is consistent with the observations that external gamma radiation values and radionuclide concentrations in the soil are higher in Naen than in Rongelap (17). Spatial distributions of where samples were collected on various islands are also shown in Fig. 2. Almost one-third of the fruits we sampled in Rongelap Atoll (18 out of 57 fruits) exceed the Japanese standard for all food (100 Bq/kg; yellow); 8 out of 11 samples on Naen Island are also in this range (100 Bq/kg; yellow).

One interesting determination was made on a coconut crab from Naen Island in Rongelap Atoll. *SI Appendix, Fig. S2A* shows a photo of the Marinelli beaker containing the coconut crab meat, and *SI Appendix, Fig. S2B* presents the measurement of the raw spectra. Given a very large systematic uncertainty, as the crab meat was not uniformly distributed and a calibration to the crab meat density was not performed, we do not report a quantitative value for this measurement. However, the raw spectrum clearly indicates the presence of ^{137}Cs based on a large peak at 662 keV. Our data suggest that the flora and soil contamination may lead to contamination further up the food chain.

To the extent that coconut crabs represent an additional food source to the Marshallese people, this also represents an additional exposure pathway.

Utirik Atoll. We measured the spectra of 32 fruits from Utirik Atoll and found no ^{137}Cs contamination within the sensitivity of our detectors. Utirik is farther from Enewetak and Bikini Atolls than Rongelap Atoll; it is 835 km from Enewetak and 490 km from Bikini, whereas Rongelap is 500 km from Enewetak and 155 km from Bikini. Fig. 2 presents the location of the fruit collected in Utirik Atoll. The ^{137}Cs values are all relatively low, with all samples in the green range, except for 4 samples on Utirik Island, which remain below all standards except for the IPPNW and Belarus, Russia, and Ukraine standards for infant food. Therefore, our data suggest that contamination of food by ^{137}Cs may not be a concern for people living currently in Utirik Atoll. Our sample sizes, however, are limited.

Comparison with Previous Studies. Studies from LLNL have been performed in the northern Marshall Islands for decades, including determinations of radiation levels in food (14–16). Reproducibility studies in science are a worthwhile endeavor not only to confirm what others have found in the past but also to help regain the public's trust in science (18). This is all the more

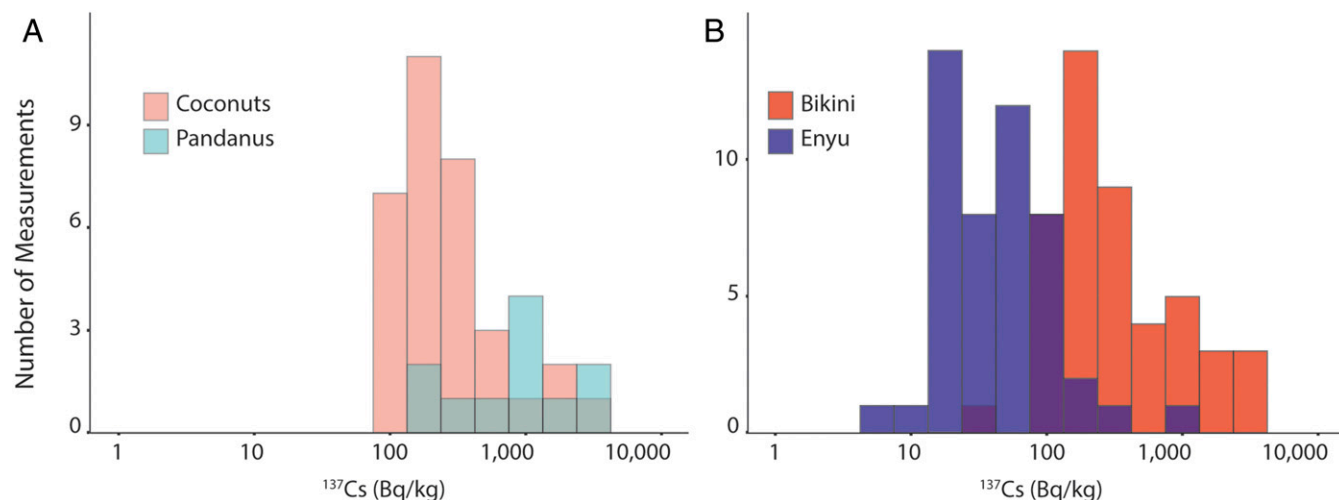


Fig. 3. (A) Comparison of the distribution of ^{137}Cs activity concentration measurements in coconuts and pandanus from Bikini Island. Contamination in pandanus is statistically significantly higher despite limited sample sizes ($P < 0.001$). **(B)** Comparison of the distribution of ^{137}Cs activity concentration measurements in fruits from Bikini Island and fruits from Enyu Island, both located in Bikini Atoll. Contamination from Bikini Island is statistically significantly higher despite limited sample sizes ($P < 0.001$).

important for the work reported here, as LLNL scientists have essentially had exclusive access to the Marshall Islands due to lack of funding for independent scientists (from the United States, RMI, or elsewhere) to pursue such research. Moreover, severe mismanagement of the nuclear testing program and its aftermath, including relocation of people onto islands that were later deemed unsafe, has given rise to a deep level of mistrust on the part of the Marshallese about LLNL reports (8, 19, 20). Therefore, our goal here is to provide an independent set of results on food radiation levels that can be compared with past studies and international standards (see *Comparison with International Standards*), while also making measurements in situ, as described earlier.

When attempting to reduce levels of ^{137}Cs contamination on Bikini Atoll, the LLNL group introduced potassium fertilizer rather than scraping the surface soil, as had been done on Enewetak Atoll. A study conducted from 1988 through 2001 suggested a decrease in ^{137}Cs for potassium-treated coconuts. The control trees, which received no treatment, fell from an average level of 5,700 Bq/kg in 1988 to an average level of 2,250 Bq/kg in 2001, and the trees with treatment dropped to an average of 240 Bq/kg (15). Our observed range of 40 to 3,770 Bq/kg is consistent with the hypothesis that the fruits we observed with the highest ^{137}Cs levels come from untreated fruits, whose contamination has gone down due to the natural decay of ^{137}Cs , whereas the fruits we observed with the lowest ^{137}Cs fruits could come from treated fruits. Given that we do not know which fruits were treated and which fruits were not treated, this is only a hypothesis. Another LLNL study, conducted in 2011, reports average Bikini ^{137}Cs values of 720 Bq/kg for coconut meat and 990 Bq/kg for pandanus (14). These values are consistent with the average values that we have found.

An assessment of Rongelap Island in 1995 using data from LLNL reports the mean coconut value that is less than 80 Bq/kg and the mean pandanus value that is less than 300 Bq/kg (8). The more recent study in 2011, mentioned above, reports an average value of 19 Bq/kg for coconut and 90 Bq/kg for pandanus (14). Our values are lower than the values reported over 20 y ago (almost a full half-life of ^{137}Cs), which is expected. Our coconuts-only average on Rongelap Island (59 Bq/kg) is almost twice as large as the more recently reported value from LLNL.

Comparison with International Standards. Table 1 summarizes a number of standards and safety limits for ^{137}Cs contamination set by different countries and organizations. Many of these standards were developed in response to the accidents in Chernobyl and Fukushima. The limits in Belarus, Russia, Ukraine, and Japan were adjusted after the Chernobyl and Fukushima accidents, respectively, and the table reflects the current permissible levels in these countries (21, 22). The table also includes limits set by the FDA in the United States (23), the European Union (21, 22), the IPPNW (24), and the International Atomic Energy Agency (IAEA) in 1996 (25). Finally, it includes the limit outlined by the 1994 Codex Alimentarius Commission (Codex), established by the Food and Agriculture Organization of the United Nations and the World Health Organization to ensure fair food trade practices (26).

We compare our results to international safety standards shown in Table 1. Ranges exist in the table as countries, such as Belarus, Russia, and Ukraine, set different limits for specific types of food; comparisons we make, where applicable, are for fruit. It should be noted, however, that none of these countries or organizations specifically set limits for fruits found in the Marshall Islands due to vastly different climates. The color-coded ranges shown in Fig. 2 reference these limits. The value of 40 Bq/kg represents the safety limits for specific fruits in Russia, Belarus, and Ukraine after the Chernobyl accident; 100 Bq/kg represents the higher limits set for fruit and the limit for fruit and berries in Japan after the Fukushima accident; 600 Bq/kg represents the fruit safety limit set

by the European Union, and 1,200 Bq/kg represents the limit for all foods, as specified by the US FDA.

Since there is a wide range of limits for what would be considered nonconsumable foods according to these different standards, numerous Marshall Islands fruits fall both above and below these stated limits. Therefore, careful attention needs to be paid when drawing conclusions on food safety. For example, the FDA food safety limits in the United States are higher than recommendations from IPPNW by nearly two orders of magnitude.

As described above, we do not find any fruits in Enewetak and Utirik with measureable contamination and therefore conclude that fruits on these atolls are below all standards. However, this is not the case for Bikini and Rongelap Atolls. For Bikini Island, the vast majority of fruit samples are above the IPPNW, Belarus, Russia, Ukraine, and Japan standards, and a few are even above the US FDA standard. Rongelap Island also has fruit above the IPPNW, Belarus, Russia, Ukraine, and Japan limits. Bikini and Rongelap Islands previously housed native populations and are yet to be resettled. Our data suggest that to address the issue of food contamination further remediation, such as by using potassium fertilizer, may be necessary to ensure that people could safely return to the islands and eat the local foods.

Conclusion

We were not able to detect any ^{137}Cs contamination in fruits from Enewetak and Utirik Atolls. However, Bikini and Rongelap Atolls have ^{137}Cs levels in fruits that exceed action limits set by IPPNW in 2016, and the governments of Russia, Ukraine, Belarus, and Japan, in response to nuclear disasters in those countries. Additionally, they exceed values found near Fukushima in February 2018 (27) and values measured from 2011 to 2015 in areas near the Chernobyl accident (28).

Both Enewetak and Utirik Atolls are inhabited, so the lack of ^{137}Cs contamination is a comforting finding for the Marshallese people living in these atolls. However, we did not do a comprehensive study of northern islands in these atolls. Especially for Enewetak, it is even expected that the northern islands will have higher radiation contamination, since that is where the majority of the nuclear tests was conducted. Although the atoll underwent a clean-up effort in 1977 through 1979, Enjebi Island in the north, for example, has gamma background contamination measurably higher than what is found in Majuro and in the southern islands on Enewetak Atoll (7, 17).

Based upon our results, we conclude that to ensure safe relocation to Bikini and Rongelap Atolls, further environmental remediation, including application of potassium fertilizer, appears to be necessary to avoid potentially harmful exposure to radiation via the indigestion pathway.

Materials and Methods

Sample Collection. Individual islands were reached by the Indies Trader *Windward* vessel, traveling over a 1-mo period in May and June of 2017. Islands within each specific atoll were chosen based on their proximity to inhabited islands, considering that inhabitants might collect fruit from those neighboring islands, also referred to by the Marshallese as “pantry islands.” The primary foods collected were coconuts and pandanus (*SI Appendix, Fig. S3*). While a few other fruits, including breadfruit, were also collected, the sample sizes were not large enough to draw conclusions and are not presented.

Fruits were collected on foot from a wide range of trees near roads, on the outskirts of islands, and near the center of islands. In addition, teams tried to collect fruits from trees far apart from each other. There were multiple samples taken from the same tree at a few islands, either due to difficulty in finding trees with accessible fruit or difficulty in navigating through the overgrown vegetation on the island. Each fruit sample collected was labeled and paired with a Garmin eTrex 10 GPS waypoint, and fruits were transported to the boat for processing and measurement.

Sample Processing and Spectral Recording. Samples were processed and analyzed in a temperature-controlled room onboard the *Windward*. Coconuts

were opened using a machete and their water was collected; pandanus keys were pried from the fruit and chopped into thin pieces. The fruit's total contents were then homogenized in a food processor. All containers and knives that came into contact with the fruit were rinsed to prevent cross-contamination. Once blended, samples were placed in a Marinelli Beaker and weighed to record the sample's "raw" weight. All samples were measured in a filled 1-L volume. Thus, for samples which were less than 1 L, water was added to fill up the Marinelli Beaker. None of the samples exceeded 1 L once homogenized. Samples were weighed and recorded as the "shake" weight.

Radiological spectra were generated using an ORTEC FoodGuard-1 sodium iodide (NaI) rapid food screening system. One-liter samples were placed inside the ORTEC lead shield, to block any potential background radiation. Count time per sample was set at 60 min. Minimum detectable concentration for ^{137}Cs in the FoodGuard-1 for a 60-min count time is quoted to be 6.0 Bq/L, although we set a far more conservative limit after the full analysis. A background radiation run was performed at each atoll to record the presence of any residual natural background radiation. Spectra were measured periodically using a microcurie ^{137}Cs source.

Analysis of the Spectra. Analysis of the spectra was performed to quantify the results and convert into units of becquerels per kilogram. Raw spectra were analyzed using Python programming code. To convert the measured spectra, (e.g., Fig. 1) to an average ^{137}Cs activity concentration in units of becquerels per kilogram, a polynomial fit was performed to determine the area under the cesium peak, including a background subtraction. The polynomial fit was performed both on the spectrum for the individual fruit and on the spectrum from a known calibration source. *SI Appendix, Fig. S4* shows an example of a fruit spectrum, including background fit for subtraction. Peaks were studied between 0 and 1,500 keV in the spectra, which corresponds to a broader range than where the ^{137}Cs peak at 662 keV is found. The area under the peak was determined in arbitrary units of counts after subtraction and this subtraction was similarly performed on a calibration source with an identical geometry, as shown in *SI Appendix, Fig. S4*.

Once the signal areas were determined, the radiation value in units of becquerels was found via the following formula:

$$\text{Radiation value} = \frac{\text{fruit counts}}{\text{calibration source counts} \times \text{source calibration constant (Bq)}}$$

The result was finally normalized to the raw weight of the fruit to determine the final result in units of becquerels per kilogram.

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