



Background gamma radiation and soil activity measurements in the northern Marshall Islands

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Contributed by Malvin A. Ruderman, May 15, 2019 (sent for review March 1, 2019; reviewed by Joanna Kiryluk and Ernst Sichtermann)

We report on measurements of external gamma radiation on 9 islands in 4 atolls in the northern Marshall Islands, all of which were affected by the US nuclear testing program from 1946 to 1958 (Enjebi, Ikuren, and Japtan in Enewetak Atoll; Bikini and Enyu in Bikini Atoll; Naen in Rongelap Atoll; and Aon, Elluk, and Utirik in Utirik Atoll). We also report americium-241, cesium-137, plutonium-238, and plutonium-239,240 activity concentrations in the soil samples for 11 islands in 4 northern atolls (Enewetak, Japtan, Medren, and Runit in Enewetak Atoll; Bikini and Enyu in Bikini Atoll; Naen and Rongelap in Rongelap Atoll; and Aon, Elluk, and Utirik in Utirik Atoll) and from Majuro Island, Majuro Atoll in the southern Marshall Islands. Our results show low external gamma radiation levels on some islands in the Enewetak Atoll and Utirik Atoll, and elevated levels on Enjebi Island in the Enewetak Atoll, on Bikini Atoll, and on Naen Island in the Rongelap Atoll. We perform ordinary kriging on external gamma radiation measurements to provide interpolated maps. We find that radionuclides are absent from all Majuro soil samples, and that they are present at highest activity concentrations in samples from Runit and Enjebi islands (Enewetak Atoll), Bikini Island (Bikini Atoll), and Naen Island (Rongelap Atoll). We contextualize all results by making comparisons between islands and to various standards, as well as to regions of the world affected by nuclear accidents. We also discuss implications for informed decision-making by the Marshallese and local atoll governments and their people on issues pertaining to island resettlement.

Marshall Islands | cesium-137 | external gamma radiation | soil activity | plutonium

Three nuclear weapons were produced and used by the United States in 1945. The first was a plutonium weapon tested at the Trinity site on July 16, 1945, in New Mexico (1), and the other 2 were used in attacks on Japanese cities Hiroshima (uranium weapon) and Nagasaki (plutonium weapon) on August 6 and 9 of the same year, respectively (2). The development of these weapons represented the culmination of the war effort named the Manhattan Project. However, even as World War II came to an end with the surrender of Japan, the effort to produce nuclear weapons continued in the United States. It persisted under the name Manhattan Project through the end of 1946, whereupon the US Atomic Energy Commission was created by the Atomic Energy Act of 1946 (3).

Within less than 1 y of the Trinity test, the United States conducted 2 tests, Able and Baker, in the Bikini Atoll, which is part of the Marshall Islands in the Pacific Ocean (4). These tests began a 12-y period of nuclear testing on 2 atolls in the Marshall Islands: Bikini and Enewetak. Enewetak was the site of the first ever hydrogen bomb test (Ivy Mike in 1951) (5), and Bikini was the site of the largest hydrogen bomb the United States ever tested (Castle Bravo in 1954) (5). Although the Marshall Islands is home to ~6% of the total number of tests conducted by the United States (67 of 1,054 total tests from 1946 to 1992; the majority of US tests were performed underground), it bore the burden of more than half the total energy yield of all nuclear weapons tests

conducted by the United States (~108.5 vs. ~196 Mt total), with Bikini Atoll home to tests with a total energy yield of ~77 Mt, nearly 40% of the total energy yield of all US nuclear tests (6, 7).

Local populations living on Enewetak and Bikini before the testing began were relocated to other atolls in the region, often living in very difficult conditions on new lands (8). In addition, several other atolls received significant fallout during the testing, particularly from the 1954 Bravo test, including the populated atolls of Rongelap and Utirik (9). Populations from each of these 4 atolls (Enewetak, Bikini, Rongelap, and Utirik), which are all recognized by the United States as having been affected by nuclear testing, have had different histories during and in the aftermath of the testing. We describe these histories and current status briefly here to give context to our radiological investigations.

In Enewetak, the people were relocated to Ujelang Atoll from their islands in 1947 in preparation for the first tests, and began returning in 1980, after a massive radioactive clean-up of the atoll (10). A population of several hundred people currently lives on Enewetak Island.

Bikinians were relocated from Bikini Island in 1946 to nearby Rongerik Atoll, which quickly proved to be an inadequate new home, primarily because of the lack of sustainable food sources on the island (8). They were moved to Kwajalein Atoll for a period of a few months, and found a more permanent home on Kili Island in the southern Marshall Islands (8). Kili Island has no lagoon and continues to cause habitation hardship. Some

Significance

From 1946 to 1958, the United States tested 67 nuclear weapons in the Marshall Islands, a remote constellation of atolls in the Pacific Ocean that was then a US trust territory. Two atolls, Bikini and Enewetak, were used as ground zero for the tests, which caused unprecedented environmental contamination and, for the indigenous peoples of the islands, long-term adverse health effects. In addition to the populations of Bikini and Enewetak, the people of Rongelap and Utirik were also affected by radioactive fallout from the largest nuclear test the United States has ever conducted, the Bravo test held March 1, 1954. This article presents a picture of current radiological conditions by examining external gamma radiation and soil radionuclide activity concentrations.

Author contributions: I.N.-H., E.W.H., and M.A.R. designed research; E.W.H. performed research; M.K.I.L.A., I.N.-H., and E.W.H. analyzed data; and M.K.I.L.A., M.R.M., I.N.-H., E.W.H., and M.A.R. wrote the paper.

Reviewers: J.K., The State University of New York at Stony Brook; and E.S., Lawrence Berkeley National Laboratory.

The authors declare no conflict of interest.

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This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1903421116/-DCSupplemental.

Published online July 15, 2019.

Bikinians returned to Bikini Island in the late 1960s, only to leave by 1978, as it became clear that they were accumulating large exposures to radiation while living on the island (8). At this time, a small number of people live on Bikini Island, primarily as caretakers, doing contract work for both the US and the Marshallese governments (11).

The Rongelapese population was exposed to radiation fallout several hours after the Bravo test in 1954 (9, 12, 13). The population was relocated 3 d after the Bravo test, only to return to Rongelap 3 y later, in 1957 (9, 12, 13). The people were subsequently evacuated from Rongelap by Greenpeace in 1985 and moved to Mejato Island, where most have been living ever since (13).

Residents of Utirik Atoll were also exposed to fallout from the Bravo test, although Utirik is farther away from Bikini than Rongelap (483 vs. 152 km, respectively) (9). The Utirik population was similarly evacuated off their islands 2 d after the blast, but returned home 3 mo after Bravo (9). Utirik has been continuously populated since that time, and currently houses a population of several hundred people.

During the decades since the end of the testing in the Marshall Islands, the Atomic Energy Commission, and later the Department of Energy, carried on numerous programs and conducted dozens of studies to ascertain the radiological conditions on the islands, as well as their effect on the health of the exposed populations (4, 7, 12–20). Although individual scientists have been praised for their contributions and dedication to this work, there is a basic lack of trust in and understanding of the results on the part of the Marshallese communities. A need for independent measurements and analysis has been paramount.

In 2016, we reported on results obtained during a 2015 trip to 3 atolls (Enewetak, Bikini, and Rongelap) in the northern Marshall Islands, and compared external gamma radiation measurements in these atolls with measurements made in the capital of the Republic of Marshall Islands, Majuro, which is located in the southern Marshall Islands (21). We subsequently conducted another trip in 2017 to explore different radiation exposure pathways and answer additional questions that arose from our initial data and analysis. First, we wanted to include Utirik in our investigations, as a US-recognized affected atoll, which we were not able to visit on our first trip because of time constraints. Second, we wanted to extend our measurements on currently or previously populated islands to outer islands within affected atolls, as we learned that people frequent the unpopulated islands in search of food or other resources, in particular during feast seasons. Finally, we also sought to make measurements of concentrations of different radionuclides in the soil, including 3 isotopes of plutonium, to gain further insight into the suitability of different islands for human habitation. Although previous work suggests that accidental ingestion of plutonium in the soil would represent a small fraction of the total radiation exposure in island residents, and recommends an action limit for plutonium of 17 pCi/g (13), all nonzero concentrations of plutonium in particular must be considered and contextualized, given a widespread lack of guidance on safety levels for these isotopes.

Results and Discussion

External Gamma Radiation.

Enewetak Atoll. We performed 47 measurements of external gamma radiation on Ikuren Island, 36 measurements on Japtan Island, and 69 measurements on Enjebi Island, which is made up of 2 main parts connected by a thin strip of land. Data are presented in Fig. 1 and *SI Appendix, Table S1*. We used a Gaussian model to perform kriging interpolation of external gamma radiation values across the extent of Ikuren Island. Given the size of the island, 47 measurements were sufficient for interpolation. However, many measurements were made on the northwestern part of the island; because of dense brush, the southeastern portion of the island could not be investigated. The predicted value spread across

the island was 7.4 mrem/y (mrem/y) or 0.074 millisievert/y (mSv/y; Figs. 2 and 3). Because of the small number of measurements made relative to the size of the island, we did not perform an interpolation for Japtan Island. Enjebi Island, in northern Enewetak Atoll, had the highest external gamma radiation levels in the whole atoll, presumably because the majority of the nuclear weapon tests were performed in this region and the cleanup in the atoll had been focused on the southern islands. The mean value for Enjebi is 69 mrem/y, with a maximum of 272 mrem/y (Fig. 1 and *SI Appendix, Table S1*). An interpolated map of Enjebi is presented in Fig. 2.

Bikini Atoll. In 2015, 137 measurements of external gamma radiation on Bikini Island were taken. Subsequently, 48 additional measurements were made during the 2017 and 2018 trips to increase the island coverage, resulting in a total of 185 measurements. At this time, the areas that lack data are mostly covered by intense and overgrown vegetation, which made further investigations difficult. Our minimum and maximum values from 2015 measurements did not change in the full data set that includes additional data (11 and 648 mrem/y, respectively), but the mean for the full data set increased slightly (191 mrem/y vs. 184 mrem/y), as did the median (149 mrem/y vs. 137 mrem/y). Fig. 1 and *SI Appendix, Table S1*. The Bikini data were fit to a Gaussian model to apply kriging across the island. The interpolation still shows a trend toward higher values in the interior of the island (430 mrem/y) and lower predicted values toward the edges of the island (45 mrem/y), consistent with our measurements, and presumably as a result of the increased runoff of radionuclides due to weather and environmental conditions.

We also took 66 measurements on Enyu Island, mostly around the outer edges of the island, with significant areas of land between the 2 coasts that were not observed due to dense overgrown vegetation. The center of the island could be reached, and these values were near 20 mrem/y. The highest external gamma radiation value recorded is near the north coast of the island at 60 mrem/y, and the minimum value recorded is 7.9 mrem/y. The mean value is 20 mrem/y (Fig. 1 and *SI Appendix, Table S1*). Fitted to a Gaussian model, kriging interpolation predicted a range from 11 to 37 mrem/y, which nearly mimics the range of measured values (Fig. 2). However, there are prediction points near the southern end of the island that are at a considerable distance from the beaches, introducing considerable uncertainty. **Rongelap Atoll.** Eighteen measurements were made on Naen Island in the northern Rongelap Atoll. The measured values ranged from 34 to 543 mrem/y, with a mean value of 322 mrem/y (Fig. 1 and *SI Appendix, Table S1*). Kriging was performed across the island, and predicted values ranged from 77 to 460 mrem/y (Fig. 2). The values are similar to the trend of measured data with high values in the interior part of the island versus at the beach. Unfortunately, large sections of the island were not measured, causing some uncertainty in the interpolation to those areas. Naen has the highest average external gamma radiation levels of all of the islands we have visited during 3 trips to the northern Marshall Islands (2015, 2017, and 2018).

Utirik Atoll. Utirik Atoll was visited for the first time by our group during the 2017 boat trip. Utirik Island is populated, and adjacent islands, Elluk and Aon, represent a potential food source. Geographically, this atoll is the farthest from the nuclear test sites; nevertheless, significant radioactive fallout caused exposure to the Marshallese population of Utirik, residing on the island during the Bravo test (21). Presently, ~400 people live on Utirik Island.

We made a total of 79 measurements on Utirik Island, with observation points that were well spread out, leading to good coverage of the island. Overall, the distribution of observations fell in the narrow range between 4.4 and 12 mrem/y with the mean of the measurements at 7.9 mrem/y. Kriging was performed on a fit Gaussian model. The range of predicted values

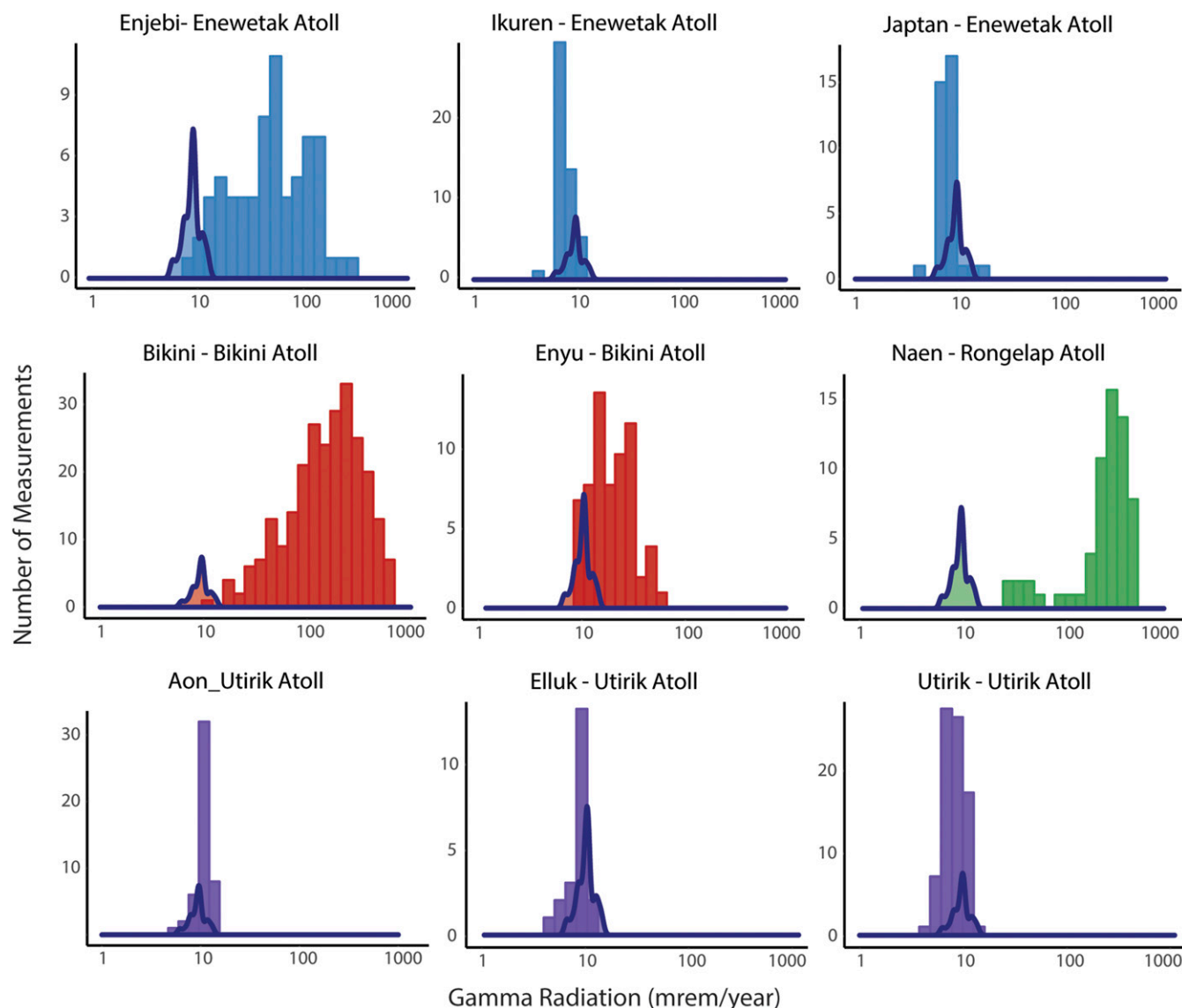


Fig. 1. Distributions of external gamma radiation on different islands in 4 atolls in the northern Marshall Islands. Blue distributions (Enewetak Atoll), Red/orange distributions (Bikini Atoll), green distribution (Rongelap Atoll), and purple distributions (Utirik Atoll). Note that the vertical axes scales differ across the islands. Superimposed on each distribution is the fitted curve (in blue) for the measured distribution of the control island, Majuro.

(5.9–11 mrem/y) was similarly flat and narrow. We made 21 measurements on Elluk, the smallest island we surveyed. Measured values were spread across the extent of the island, and ranged from 4.4 to 11 mrem/y, with a mean of 7.9 mrem/y. We fit a Gaussian semivariogram model and then used it to perform kriging across the island. Due to the small size of the island, the predicted value across the entire island was 7.8 mrem/y, which is very close to the mean value measured on the island. On Aon Island, we made 49 measurements. Similar to Utirik, the observations thoroughly covered most of the island. The values ranged from 5.3 to 15 mrem/y, with a mean of 10 mrem/y. The kriging predicted values ranged from 8.3 to 11 mrem/y (Figs. 1, 2, and 3, and *SI Appendix, Table S1*).

Comparison with Control. We compared the measured external gamma radiation levels on each island to Majuro Island, which we investigated in 2015, and designated as the control island (21). We employed upper-tailed Wilcoxon Rank-Sum tests for these comparisons. We found that 2 islands on Enewetak Atoll,

Ikuren and Japtan, did not have elevated external gamma radiation levels compared with Majuro Island, but Enjebi did ($P < 0.01$). We also found that Bikini and Enyu islands on Bikini Atoll and Naen Island on Rongelap Atoll all had external gamma radiation levels that were statistically significantly higher than those measured on Majuro Island ($P < 0.01$). For Utirik Atoll, we found that all 3 islands did not have background gamma radiation that was measurably higher than Majuro.

Comparison with Standards. The United States and the Republic of the Marshall Islands have produced a Memorandum of Understanding (MOU) regarding the resettlement of Rongelap Atoll, which as described in the introduction, is yet to be resettled (13). This agreement has a limit of 100 mrem/y for a maximally exposed resident from all exposure pathways. According to our data, both Bikini and Naen Islands exceed this limit even after accounting for natural background radiation, which we take to be equivalent to values measured on Majuro Island in the Majuro Atoll in the southern Marshall Islands, and only from exposure to external

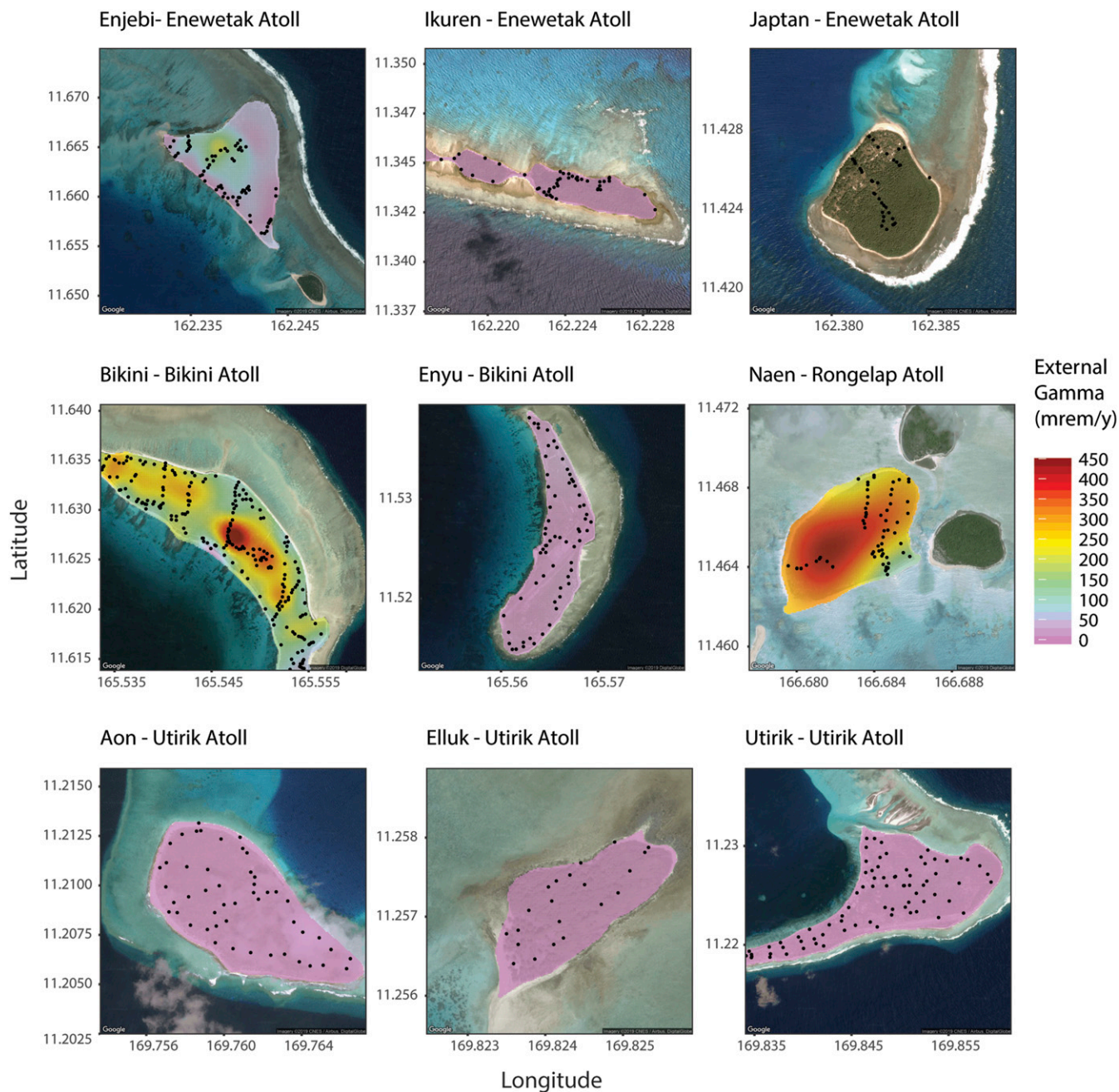


Fig. 2. Maps of 9 islands in the 4 atolls in the northern Marshall Islands, including 8 interpolated maps. The locations of measured values are denoted as black dots. The color correspondence to a predicted external gamma radiation values is the same for all eight interpolated islands (not Japtan) and is shown in the legend on the right-hand side.

gamma radiation. Although Enjebi Island does not exceed the 100 mrem/y standard just from the average external gamma radiation value, it is likely that it would if other exposure pathways were considered (e.g., food). See Fig. 4 for summary of the findings.

Radioisotope Concentrations in the Soil. We made measurements of the activity concentration of americium-241 (^{241}Am), cesium-137 (^{137}Cs), plutonium-238 (^{238}Pu), and $^{239,240}\text{Pu}$ in up to 38 soil samples: 34 from the 4 affected atolls, and 4 from Majuro Island in the southern Marshall Islands. We elaborate on measured concentrations for all the radionuclides given below, and make comparisons to standards and soil activity concentrations measured in other parts of the world.

^{241}Am . We obtained ^{241}Am activity concentrations in 30 soil samples from Enewetak, Bikini, Rongelap, and Utirik atolls. Enjebi, Runit, Bikini, and Naen islands had the highest activity concentrations. Results and uncertainties are presented in Fig. 5. All other islands had concentrations ranging from 8 to 73 Bq/kg. Locations of where soil samples were collected are shown in Fig. 6. Activity concentrations were highest in samples from Naen Island, with a mean of 1,270 Bq/kg or 34 pCi/g and a maximum value of 3,090 Bq/kg. This is a surprising finding, as Rongelap Atoll was not a site of nuclear testing, but was only exposed to fallout, primarily from the Bravo test performed in 1954. Enjebi Island also featured a single high-activity value at 1,520 Bq/kg, while the other sample was at 63 Bq/kg. The next highest results

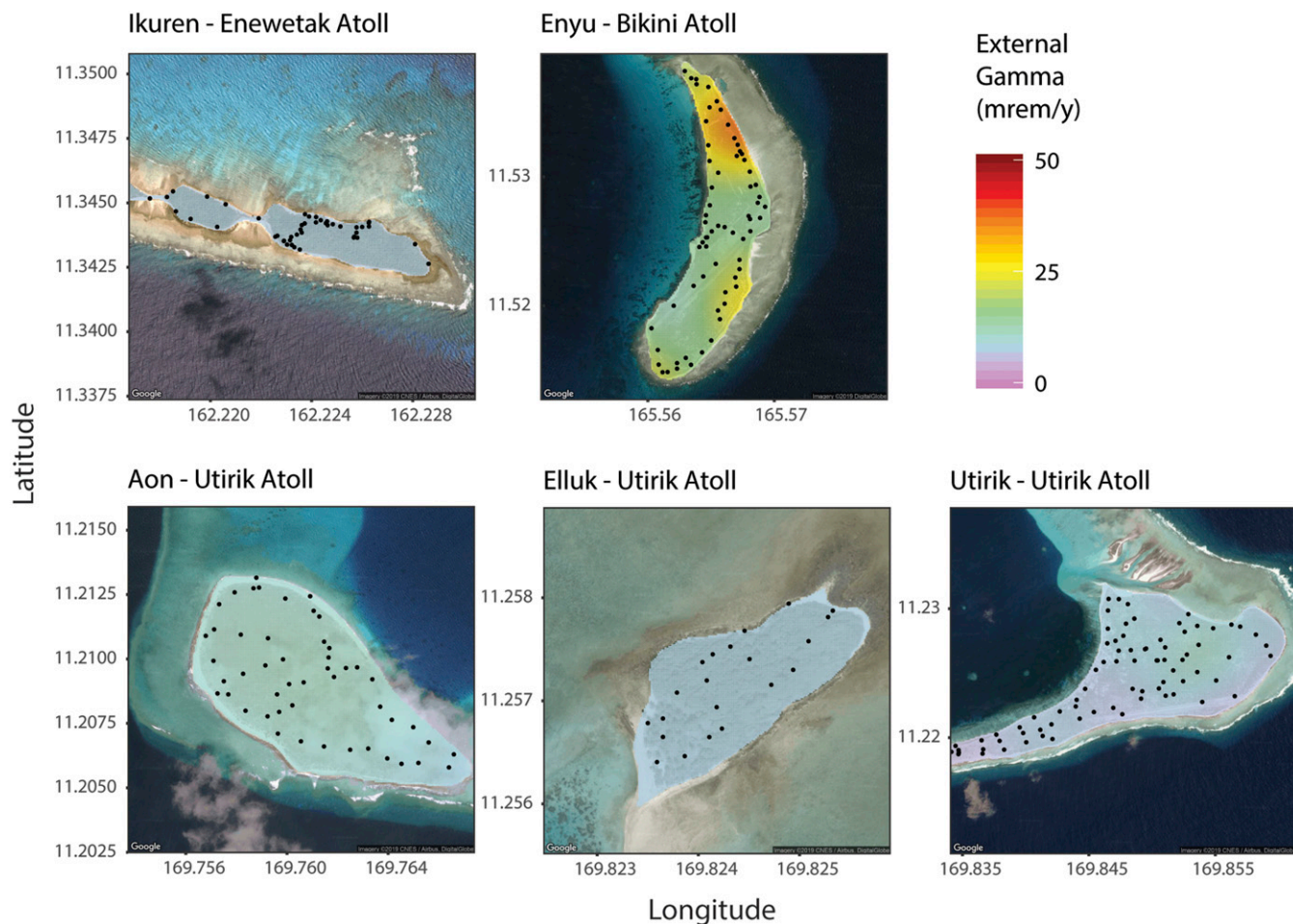


Fig. 3. Interpolated maps of 5 islands in the northern Marshall Islands with the lowest external gamma radiation levels. The maps are provided to distinguish between levels that range from near 0 to 50 mrem/y. The locations of measured values are denoted as black dots. The color correspondence to a predicted external gamma radiation values is the same for all islands and is shown in the legend on the right-hand side.

came from Runit samples, with an average of 230 Bq/kg and a maximum value of 550 Bq/kg. Our values for Bikini Island (1 sample at 80 Bq/kg and another sample at 175 Bq/kg) are consistent with previous findings (7). See Figs. 5 and 6 for data from selected samples and the sample locations.

The ^{241}Am activity concentrations in the soil were measured in 2009 (23 y after the Chernobyl disaster) in different regions of the Polesie State Radioecological Reserve (PSRER). PSRER is a region of Belarus most affected by the Chernobyl fallout, which is geographically adjacent to the Chernobyl Exclusion Zone in the Ukraine (22). Average ^{241}Am concentrations in separate plots ranged from 61.4 to 97.7 Bq/kg, which is higher or similar to concentrations from 12 northern Marshall Islands samples, and much lower than concentrations from 1 Bikini Island sample, 1 Enjebi sample, 7 Naen samples, and 5 Runit samples. In addition, ^{241}Am concentrations in the soil were measured in and near the Rocky Flats Environmental Technology Site in Colorado, which was a nuclear weapons production facility up until 1989, and which was closed due to accidental release of weapons-grade plutonium (23). Values for ^{241}Am in the soil there were rather low, and the 95% confidence interval ranged from 1.1 to 1.6 Bq/kg, which was much lower than activity concentrations in all the measured Marshall Islands samples. Finally, we are aware of only a single standard for presence of ^{241}Am in soil. It is the decontamination limit proposed by the US Nuclear Regulatory Commission, and set at 30 pCi/g or 1,110 Bq/kg (24). This limit is

exceeded by 1 Enjebi Island sample and 4 Naen Island samples. The average for Naen Island also exceeds this limit.

^{137}Cs . We obtained ^{137}Cs activity concentrations in all 38 soil samples. Enjebi, Runit, Bikini, and Naen islands have the highest concentrations, and their values and uncertainties are given in Fig. 5. All other islands have concentrations ranging from 0 to 55 Bq/kg. Fig. 6 gives the locations of the collected samples. Concentrations in Majuro samples are equal to zero within the measurement uncertainty, as are concentrations in 2 samples from Enewetak Island and 1 sample from Medren Island, both in Enewetak Atoll. Values from 5 Runit samples range from 3 to 2,790 Bq/kg, with an average activity concentration of 334 Bq/kg. ^{137}Cs measurements are low in the 2 Rongelap Island samples (13.1 and 14.5 Bq/kg), and are significantly elevated in the 2 Bikini Island samples (455.1 and 636.4 Bq/kg). ^{137}Cs concentration in Naen samples are highest, similar to the ^{241}Am measurements, with a range of 20–7,140 Bq/kg and an average of 3,090 Bq/kg. The highest concentration sample is more than an order of magnitude higher in value than the 2 Bikini samples. One of the Enjebi samples had a concentration of 2,850 Bq/kg. See Figs. 5 and 6 for data from selected samples and the sample locations.

Our values for Bikini Island are somewhat lower than values reported in Simon et al. in 1997, which would be expected given that they were obtained ~20 y later, assuming that samples were collected at similar locations (12). Samples from Pohnpei, Guam, Truk, and Palau, which ranged from 1.5 to 11 Bq/kg when

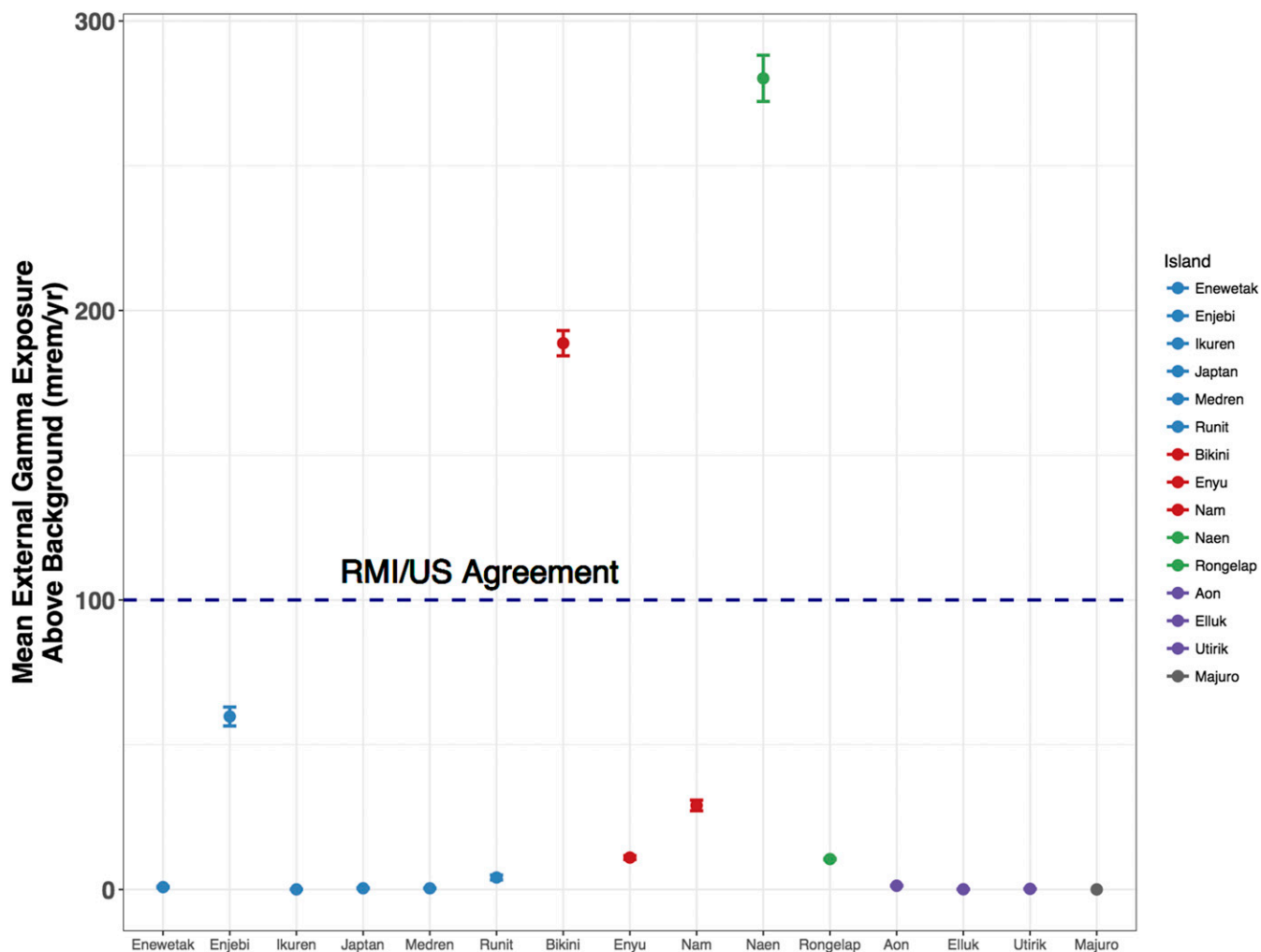


Fig. 4. Mean values of external gamma radiation on various islands in the northern Marshall Islands, adjusted for the radiation on the control island, Majuro. Error bars represent ± 1 SE. Blue denotes islands located in the Enewetak Atoll, red: Bikini Atoll, green: Rongelap Atoll; purple: Utirik Atoll. Majuro on Majuro Atoll, the control island, is denoted in gray. Dotted line denotes the United States and Republic of Marshall Islands agreed-on total radiation limit for relocation.

decay corrected to 1996, were previously reported as representative of the worldwide fallout levels for this latitude band ($5\text{--}15^\circ\text{N}$) (25). However, given the zero concentrations we found in 4 Majuro Island samples, we question such a designation at this time.

In Japan, a Food Sanitation Law sets a limit for $^{134+137}\text{Cs}$ in agricultural soils at 5,000 Bq/kg, with an implied limit for ^{137}Cs activity concentration of 2,500 Bq/kg (26). In the northern Marshall Islands, we find that most samples are below this limit. However, 3 Naen samples were near the limit, and 3 additional Naen samples, 1 Enjebi sample, and 1 Runit sample were all above the limit. The aforementioned concentrations are also all above concentrations found in soil near the Chernobyl power plant less than 10 y after the accident, which were 960 and 1,210 Bq/kg (27).

^{238}Pu and $^{239,240}\text{Pu}$. We obtained ^{238}Pu activity concentrations in all 38 soil samples. The concentrations were equal to zero within uncertainty in 26 samples. The only samples with ^{238}Pu concentrations above zero were 2 Naen Island samples (1.8 and 63 Bq/kg) and all 10 Runit Island samples (1.5–138 Bq/kg), which had a mean of 42 Bq/kg.

We obtained $^{239,240}\text{Pu}$ activity concentrations in all 38 soil samples from 13 islands, including Majuro. Majuro concentrations are equal to zero, as are concentrations in 2 samples from Enewetak, 1 sample from Medren, and 1 sample from Japtan. Lowest concentrations are found in the 2 samples from Rongelap

Island (4.5 and 6.4 Bq/kg) and 2 samples from Utirik Island (7.8 and 8.2 Bq/kg). Concentration is higher in samples from other Utirik Atoll islands; namely, 17.3 Bq/kg in a sample from Aon and 65.1 Bq/kg in a sample from Elluk. Enyu samples are also elevated at 24.8 and 30.2 Bq/kg, whereas Bikini samples are even higher at 56.2 and 210.5 Bq/kg. Naen samples have $^{239,240}\text{Pu}$ concentrations, once again higher than those from Bikini at 205–5,180 Bq/kg, with a mean of 1,640 Bq/kg, while Runit samples range from 67 to 2,340 Bq/kg, with a mean of 500 Bq/kg. One Enjebi sample has a concentration of 1,830 Bq/kg. See Figs. 5 and 6 for data from selected samples and the sample locations.

Our results for Bikini Island are consistent with previous measurements (7, 28). All nonzero values are well above the global fallout activity concentration, which was measured recently in samples from the 1970s near Fukushima, and was found to range from 0.004 to 1.5 Bq/kg (29). The $^{239,240}\text{Pu}$ levels did not increase significantly, and were found in Fukushima to be as high as 0.353 Bq/kg in the first couple of months after the accident, which is more than 10–1,000 times lower than all nonzero activity concentrations from the northern Marshall Islands samples (30). Average $^{239,240}\text{Pu}$ activity concentrations from PSRER samples were around 30 Bq/kg, which is similar to or higher than some of our samples, but 2–17 times lower than samples from Bikini, Runit, Enjebi, and Naen Islands (22).

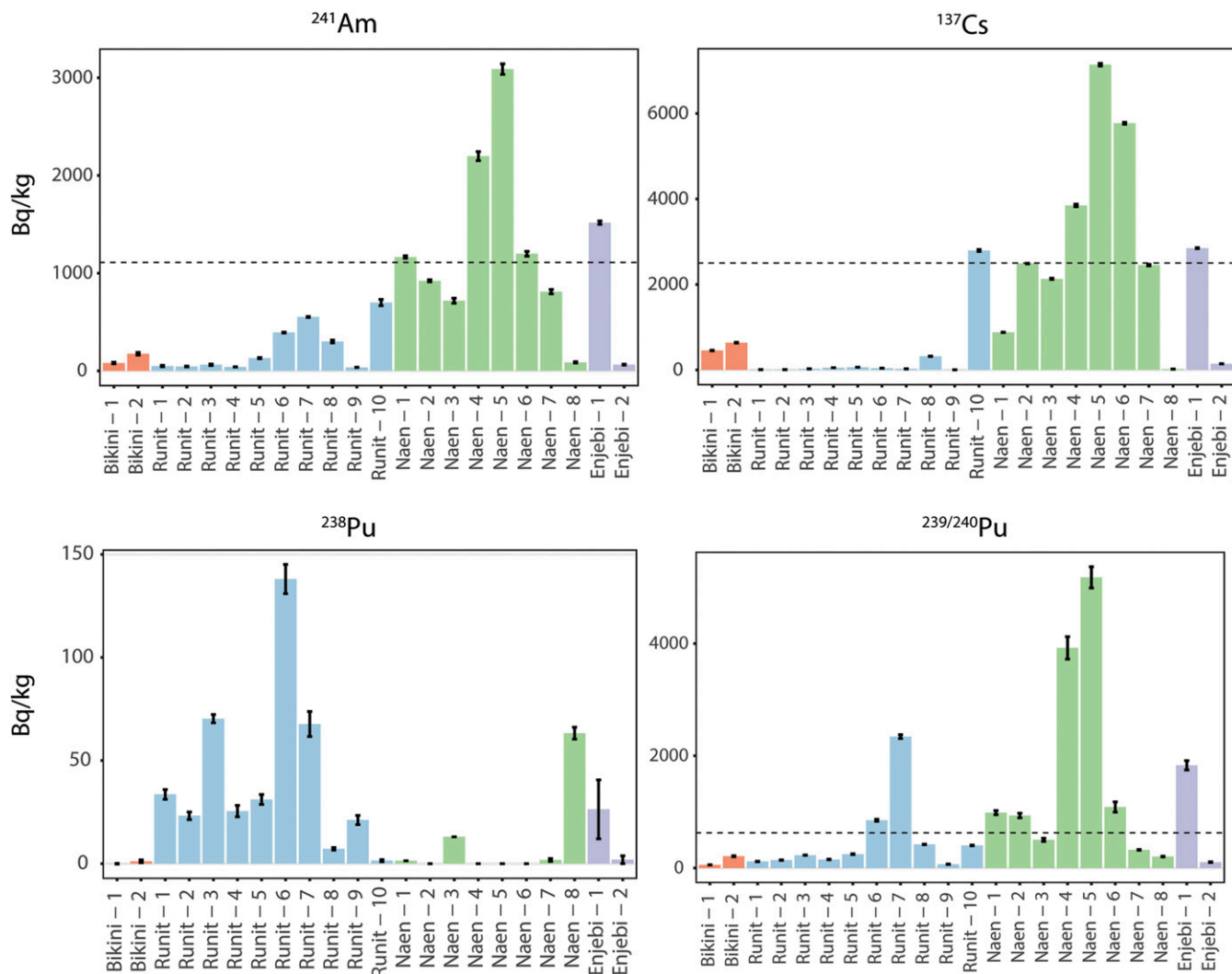


Fig. 5. Mean radioisotope activity concentrations (Bq/kg) in the soil for 20 samples with the highest concentrations from different atolls (Enjebi and Runit islands on Enewetak Atoll, Bikini Island on Bikini Atoll, and Naen Island on Rongelap Atoll). Error bars represent ± 1 SE. Note the different scales on the vertical axes for the different isotopes. In the ^{241}Am panel, the dotted line represents the decontamination limit by the US Nuclear Regulatory Commission. In the ^{137}Cs panel, the dotted line represents the Food Sanitation Law limit in Japan. In the $^{239/240}\text{Pu}$ panel, the dotted line represents the action limit for total plutonium in the soil, according to the MOU between the US and the RMI. Note that for this limit, the total plutonium, including ^{238}Pu , should be considered.

The MOU between the United States and the Government of the Republic of the Marshall Islands sets an action limit for total amount of plutonium in the soil. This action limit is set at 17 pCi/g or 629 Bq/kg (13). Two Runit samples, 1 Enjebi sample, and 5 Naen samples all exceed this limit. While many of the samples we measured fall under this action limit, this limit is also significantly higher than values for plutonium found in areas affected by the Chernobyl and Fukushima accidents. We are surprised by the lack of any further guidance on permissible plutonium levels in the soil from national governments and international organizations.

Conclusion

In this study we take a broad-brush view of radiological conditions on 4 affected atolls in the northern Marshall Islands (Enewetak, Bikini, Rongelap, and Utirik) with an emphasis on outer islands in the atolls that may not be suitable for habitation on their own, but that islanders may visit in search of food, especially in times of harvest. Here we summarize our findings for each of the 4 atolls.

Enewetak. Consistent with our previous study, in which we found low levels of external gamma radiation on Enewetak and Medren

islands, long after a large-scale clean-up effort of Enewetak Atoll, we also find low levels of external gamma radiation on both Ikuren and Japtan islands (21). However, we do find elevated gamma radiation levels on Enjebi Island in northern Enewetak. In addition, 1 of 2 soil samples we collected on Enjebi Island had high concentrations of all 5 radionuclides, including levels above safety limits for ^{137}Cs and $^{239,240}\text{Pu}$. Runit Island, which is home to a nuclear waste repository, has significant levels of all 5 radionuclide concentrations (Fig. 5).

Our results suggest that people currently living in southern Enewetak are not likely to get significant exposure to radiation from nuclear weapons testing. However, the presence of radioactive isotopes on the Runit Island is a real concern, and residents should be warned against any use of the island. Moreover, wash-off of existing isotopes off the islands into the ocean from weathering and continued sea level rise (31) continues to threaten further contaminating the lagoon and the ocean at large. Investigating radioactivity levels in the ocean near Runit Island, and in the northern Marshall Islands more generally, is of utmost importance. Furthermore, residents of southern islands in the

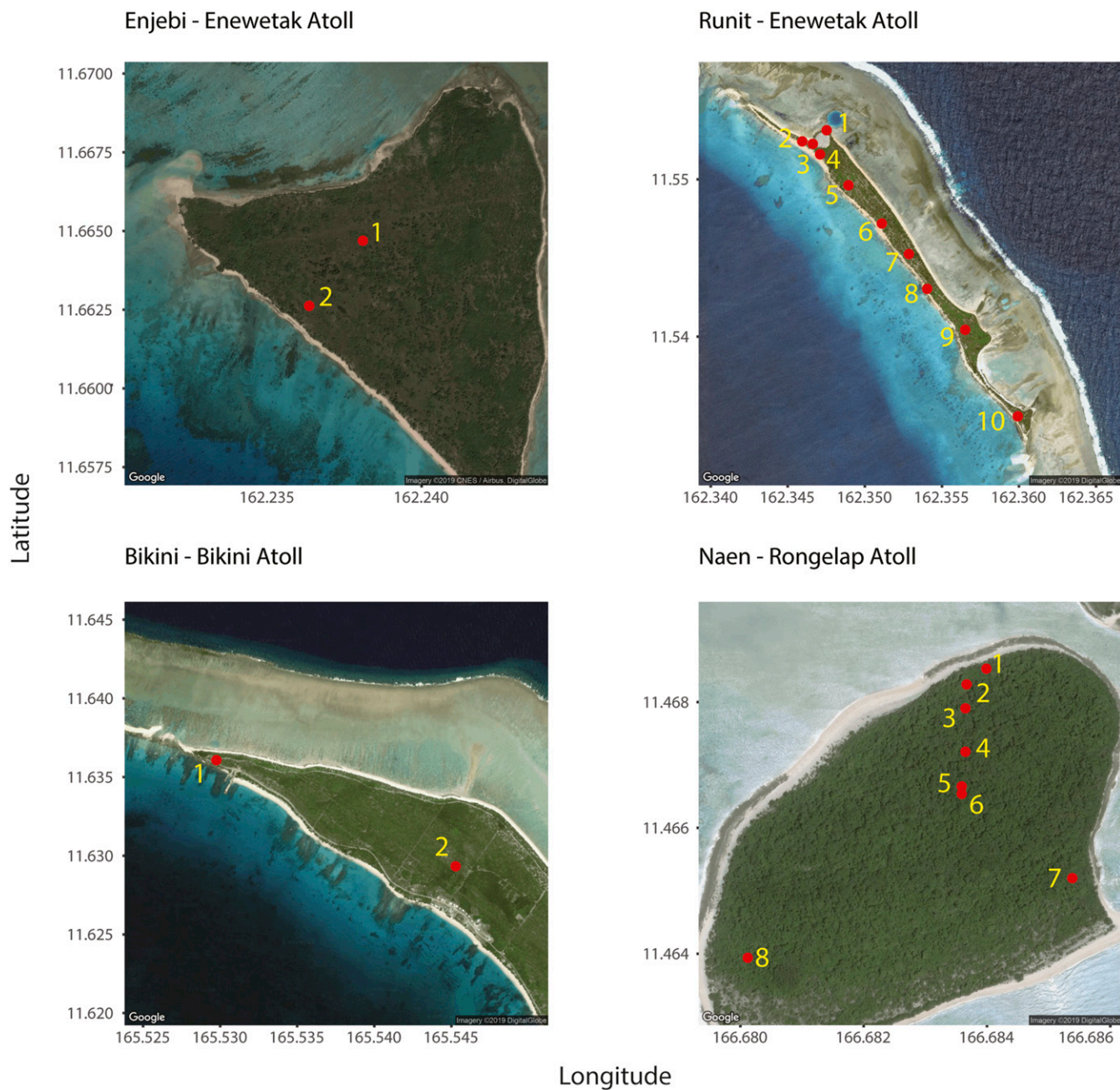


Fig. 6. Locations where soil samples were collected and whose radioisotope concentrations are given in Fig. 5.

Enewetak Atoll should also be warned against spending time on northern islands, including Enjebi Island.

Bikini. Consistent with our previous study, we find highly elevated gamma radiation levels on Bikini Island (21). When including data points from all collection periods, the average value of background gamma radiation is 190 mrem/y, which is nearly double the agreed-on limit set by the Republic of the Marshall Islands/US governments for total nuclear testing-related exposure; namely, 100 mrem/y (Fig. 4). Furthermore, both Bikini samples had highly elevated concentrations of ^{241}Am , ^{137}Cs , and $^{239+240}\text{Pu}$ in soil. In particular, ^{241}Am soil activity concentrations in Bikini were up to 3 times higher than in soil samples from PSRER in Belarus and up to 150 times higher than in soil samples from the Rocky Flats Environmental Technology Site in Colorado (22, 23). $^{239,240}\text{Pu}$ concentrations were

up to 15–1,000 times higher than in samples from areas affected by the Chernobyl and Fukushima disasters (29, 30).

The levels of radiation appear to be significantly lower on Enyu Island compared with Bikini Island, both for the external gamma radiation and for radioactive isotope concentrations in the soil. The difference in their external gamma radiation levels is statistically significant ($P < 0.01$), and radioisotope activity concentrations were 2–12 times higher in Bikini than in Enyu soil samples. These results suggest that perhaps some radiological clean-up may have been done on Enyu Island. Nevertheless, the radiation levels on Bikini Island, which served as the primary island for habitation on the atoll, before and in the aftermath of the testing, are too high for relocation to Bikini. This is true even before radiation in food, which may represent the most significant exposure pathway, is taken into account (32).

Rongelap. Our previous work suggested that external gamma radiation levels on Rongelap Island were slightly elevated compared with those on Majuro Island, and the conclusion regarding suitability for human habitation on this island would need to rest on data pertaining to radioactive levels in food (21). As part of this study, we focused on Naen Island measurements. Naen is located in the north, and was previously reported to have much higher radiation levels than Rongelap Island (25). We found external gamma radiation values on Naen to be the highest of any island we investigated, well above the 100 mrem/y agreement between the Republic of the Marshall Islands and the United States governments (MOU). The Naen mean is higher than Bikini's partially due to the larger number of data points collected in the interior of the island compared with the edges; the highest radiation point that we measured still comes from Bikini Island.

In addition, radioactive isotope activity concentrations in soil were high in 7 soil samples from Naen Island, in particular for ^{241}Am , ^{137}Cs , and $^{239+240}\text{Pu}$, and higher in all cases than concentrations in the Bikini samples. The sample near the ocean side had lower values for these isotopes and a much higher value for ^{238}Pu . This is an interesting discovery, given that Runit soil samples near and far from the Runit dome showed a similar signature. A possible explanation for the unexpectedly high radiation levels in Naen could be because Naen was on the direct path of wind of the nuclear explosions (25). The activity concentration levels and signature are also consistent with an untested hypothesis that some of the waste from the cleanup done on Rongelap may have been transported to Naen Island, similar to what was done on Runit Island. In summary, given the lifestyle of people living on remote atolls, it appears to us that clean-up of Naen, and possibly other northern Rongelap Atoll islands, would be needed before full atoll resettlement can be envisioned.

Utirik. Our studies on Utirik were not comprehensive due to time constraints. As in the case of southern islands in the Enewetak Atoll, Utirik Atoll had a very flat distribution of external gamma radiation values and no hot spots. The overall mean measurement of radiation is 8.7 mrem/y, and compared with the mean external gamma exposure on Majuro, which is 9.5 mrem/y, we can conclude that the measured radiation is no longer elevated from the nuclear testing fallout. In addition, radionuclide activity concentration levels in soil on Utirik were relatively low. Some samples are above the zero or near the zero Majuro background measurements, but still significantly lower than the levels seen on Bikini, Naen, Enjebi, and Runit islands. Although $^{239,240}\text{Pu}$ levels are significantly below the MOU action limit, they are still higher than levels in Fukushima and Chernobyl accident affected areas. Further monitoring of radiation levels in soil on Utirik Atoll should be considered.

Materials and Methods

External Gamma Radiation.

Gamma radiation detectors. We conducted gamma radiation measurements using Ludlum model 44–20 Gamma scintillators and operated using Ludlum model 2241–2 survey meters. The scintillators are 3 inches in diameter and are composed of thick sodium iodide crystals optically coupled to photomultiplier tubes. The detectors are sensitive to gamma radiation over an energy range of 60 keV to 2 MeV. The survey meters were read out visually by a liquid-crystal digital display. The same detectors were used in our previous study (21).

Detector calibration. The Ludlum scintillators and survey meters were each calibrated by Ludlum Measurements, Inc. before their delivery in July and August 2015, and subsequently by us, as was described previously (21).

External gamma radiation data collection. Data were collected by a team of 2 researchers. One member would read out the Ludlum detector results,

while the second member recorded the value and the location of the measurement using a Garmin eTrax GPS, saving it as a waypoint record. The Ludlum scintillator was pointed from waist height. Measurements were typically taken at ~100–200-m separations, although the consistency in distance traveled between data points was not strictly enforced.

On the islands of Ikuren, Japtan, Naen, Elluk, Utirik, Aon, Enyu, and Bikini (2017) and Bikini and Enjebi (2018), the teams walked around the islands. However, over many regions, there were areas of high brush that could not be penetrated and where no measurements were made.

Data analysis. We used the Shapiro–Wilk test to assess the normality of our data by island. On the basis of our findings, we determined nonparametric statistical tests were more appropriately applied to our data than parametric tests. We used the Wilcoxon Rank-Sum test and Kruskal–Wallis tests to compare measured external gamma radiation across islands and to compare our measurements to safety standards for radiation exposure. In addition, we examined our data geostatistically. This involved constructing empirical semivariograms, fitting several variogram models, and selecting the most appropriate model for our data on the basis of model selection criteria including minimized weighted least-squares, minimized loss function, and leave-one-out cross-validation. We used our semivariogram model to perform ordinary kriging interpolation to estimate the radiation levels over the extent of each island.

All statistical and geostatistical analyses were performed using the R Project for Statistical Computing (R). We acquired satellite maps of each of the islands by querying Google Earth's Application Program Interface using RGoogleMaps, an R package. We used the Environmental Systems Research Institute (ESRI) Arc-Map to clip masks of each of the islands, which we then used to define the interpolation extent. Interpolation was performed using ordinary kriging in R.

Radioisotope Concentrations in the Soil.

Soil data collection. Varying amounts of soil samples were taken from different islands in 2017 and 2018 at random locations that were accessible to the researchers. A researcher scooped the top 5–10 cm of soil into a 50-mL conical tube, using a shovel. One sample was taken on Aon, Medren, and Japtan, and 2 samples were taken on Bikini, Enyu, Utirik, Rongelap, and Enewetak. Four, 6, and 10 samples were taken on Majuro, Naen, and Runit, respectively. The samples were shipped back to the United States for analysis.

Radioisotope concentrations in soil. All soil samples were sent to Gel Laboratories LLC in Charleston, South Carolina (<http://www.gel.com>), to conduct analysis of the presence of 4 radioactive isotopes: ^{241}Am , ^{137}Cs , plutonium-238 (^{238}Pu), and $^{239+240}\text{Pu}$. Given the small samples size for most islands, single values are reported, rather than any summary statistics, which are given for Naen and Runit islands.

Gel Laboratories performed the radiochemical analysis of the soil samples. The subsample precipitate was dissolved in acid, and the elements were subsequently separated by ion exchange resins. After radiochemical separation, concentrations were determined by isotopic alpha emission (33). Minimum detection limits for alpha emission are estimated to be 1 pCi/g or 37 Bq/kg. Measurements of ^{137}Cs activity were performed using gamma detection techniques, and minimum detection limits for this process are estimated to be 0.1 pCi/g or 3.7 Bq/kg.

ACKNOWLEDGMENTS. We thank the people of Bikini, Utirik, Rongelap, and Enewetak for approving this project and allowing us access to numerous islands. We also thank the late Bill Graham for guiding us through the approval process in 2017 and the Marshall Islands Nuclear Commission for guiding us through the approval process in 2018. We thank Indies Trader for providing the infrastructure, transportation, and overall support for the research trip. We also thank Autumn Bordner, Danielle Crosswell, Gemma Sahwell, and Aliza Silverstein for making background gamma radiation measurements on several islands. We thank Cassandra Fernandes for her help with shipping and handling of soil samples and Gel Laboratories for their speedy delivery of the soil analysis. We thank Thomas Morgan for his overall guidance on radiation safety issues. We thank Senator and Speaker Kenneth Kedi for suggesting that we extend our studies to include Naen Island in Rongelap Atoll; the radiation levels from Naen Island were much higher than we had expected. This project was funded and supported by Columbia University.

1. J. J. Bellucci *et al.*, Distribution and behavior of some radionuclides associated with the Trinity nuclear test. *J. Radioanal. Nucl. Chem.* **295**, 2049–2057 (2013).
2. W. C. Roesch, "US–Japan joint reassessment of atomic bomb radiation dosimetry in Hiroshima and Nagasaki. DS86 Dosimetry System 1986" (Tech. Rep. 35050238, International Atomic Energy Agency, Vienna, Austria, 1987), vol. 1.
3. Atomic Energy Commission, "Summary of the Atomic Energy Act, 42 U.S.C. §2011 et seq." (1946).

4. J. Wang, S. A. L. Caccamise, L. Wu, L. A. Woodward, Q. X. Li, Spatial distribution of organochlorine contaminants in soil, sediment, and fish in Bikini and Enewetak Atolls of the Marshall Islands, Pacific Ocean. *Chemosphere* **84**, 1002–1008 (2011).
5. S. L. Simon, A. Bouville, C. E. Land, H. L. Beck, Radiation doses and cancer risks in the Marshall Islands associated with exposure to radioactive fallout from Bikini and Enewetak nuclear weapons tests: Summary. *Health Phys.* **99**, 105–123 (2010).

6. United States Department of Energy (USDOE), "United States nuclear tests: July 1945 through September 1992" (Tech. Rep. DOE/NV-209-REV 15, U.S. Department of Energy, Nevada Operations Office, Las Vegas, NV, 2000).
7. S. L. Simon, W. L. Robison, A compilation of nuclear weapons test detonation data for U.S. Pacific ocean tests. *Health Phys.* **73**, 258–264 (1997).
8. J. Niedenthal, A history of the people of Bikini following nuclear weapons testing in the Marshall Islands: With recollections and views of elders of Bikini Atoll. *Health Phys.* **73**, 28–36 (1997).
9. E. T. Lessard, N. A. Greenhouse, R. P. Miltenberger, "Reconstruction of chronic dose equivalents for Rongelap and Utirik residents: 1954 to 1980" (Tech. Rep. BNL 51257, US Department of Energy, Washington, DC, 1980).
10. W. L. Robison, W. A. Phillips, M. E. Mount, B. R. Clegg, C. L. Conrado, "Reassessment of the potential radiological doses for residents resettling Enewetak Atoll" (National Technical Reports Library, 12621946, International Atomic Energy Agency, Vienna, Austria, 1980).
11. T. F. Hamilton, W. L. Robison, "Overview of radiological conditions on Bikini Atoll" (Tech. Rep. UCRL-MI 208228, Lawrence Livermore National Laboratory, Livermore, CA, 2004), pp. 1–18.
12. S. L. Simon *et al.*, A comparison of independently conducted dose assessments to determine compliance and resettlement options for the people of Rongelap Atoll. *Health Phys.* **73**, 133–151 (1997).
13. US National Academy of Sciences, National Research Council (US), Committee on Radiological Safety in the Marshall Islands, *Radiological Assessments of the Resettlement of Rongelap in the Republic of the Marshall Islands* (National Academies Press, Washington, DC, 1994).
14. S. Simon, J. C. Graham, A. W. Borchert, Concentrations and spatial distribution of plutonium in the terrestrial environment of the Marshall Islands. *Sci. Total Environ.* **229**, 21–39 (1999).
15. J. Takada, M. Yamamoto, Radiological status of Rongelap island in 1999. *J. Radioanal. Nucl. Chem.* **252**, 261–266 (2002).
16. W. L. Robison, T. F. Hamilton, Radiation doses for Marshall Islands Atolls affected by U.S. nuclear testing: All exposure pathways, remedial measures, and environmental loss of (¹³⁷Cs). *Health Phys.* **98**, 1–11 (2010).
17. J. C. Graham, S. L. Simon, A study of ¹³⁷Cs in soil profiles from the Marshall Islands. *Sci. Total Environ.* **183**, 255–268 (1996).
18. P. S. Harris, S. L. Simon, S. A. Ibrahim, Urinary excretion of radionuclides from Marshallese exposed to fallout from the 1954 Bravo nuclear test. *Health Phys.* **99**, 217–232 (2010).
19. W. L. Robison, V. E. Noshkin, Radionuclide characterization and associated dose from long-lived radionuclides in close-in fallout delivered to the marine environment at Bikini and Enewetak Atolls. *Sci. Total Environ.* **237-238**, 311–327 (1999).
20. L. C. Sun, A. R. Moorthy, E. Kaplan, J. W. Baum, C. B. Meinhold, Assessment of plutonium exposures in Rongelap and Utirik populations by fission track analysis of urine. *Appl. Radiat. Isot.* **46**, 1259–1269 (1995).
21. A. S. Bordner *et al.*, Measurement of background gamma radiation in the northern Marshall Islands. *Proc. Natl. Acad. Sci. U.S.A.* **113**, 6833–6838 (2016).
22. A. Grubich, V. I. Makarevich, O. M. Zhukova, Description of spatial patterns of radionuclide deposition by lognormal distribution and hot spots. *J. Environ. Radioact.* **126**, 264–272 (2013).
23. S. E. Hulse, S. A. Ibrahim, F. W. Whicker, P. L. Chapman, Comparison of ²⁴¹Am, (^{239,240}Pu and ¹³⁷Cs concentrations in soil around rocky flats. *Health Phys.* **76**, 275–287 (1999).
24. R. E. Cunningham, H. D. Thornburg, "Decontamination Limits for Americium-241. U.S.NRC" (Tech. Rep. HPPOS-183 PDR-9111210288, U.S. Nuclear Regulatory Commission, Washington, DC, 1981).
25. W. L. Robison *et al.*, The northern Marshall Islands radiological survey: Data and dose assessments. *Health Phys.* **73**, 37–48 (1997).
26. T. J. Yasunari *et al.*, Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. *Proc. Natl. Acad. Sci. U.S.A.* **108**, 19530–19534 (2011).
27. M. Hoshi *et al.*, Fallout radioactivity in soil and food samples in the Ukraine: Measurements of iodine, plutonium, cesium, and strontium isotopes. *Health Phys.* **67**, 187–191 (1994). Correction in: *Health Phys.* **67**, 127 (1994).
28. Y. Muramatsu *et al.*, Measurement of ²⁴⁰Pu/²³⁹Pu isotopic ratios in soils from the Marshall Islands using ICP-MS. *Sci. Total Environ.* **278**, 151–159 (2001).
29. G. Yang, J. Zheng, K. Tagami, S. Uchida, Plutonium concentration and isotopic ratio in soil samples from central-eastern Japan collected around the 1970s. *Sci. Rep.* **5**, 9636 (2015).
30. S. Schneider *et al.*, Plutonium release from Fukushima Daiichi fosters the need for more detailed investigations. *Sci. Rep.* **3**, 2988 (2013).
31. M. J. Widlansky, A. Timmermann, W. Cai, Future extreme sea level seesaws in the tropical Pacific. *Sci. Adv.* **1**, e1500560 (2015).
32. C. E. W. Topping *et al.*, In situ measurement of cesium-137 contamination in fruits from the northern Marshall Islands. *Proc. Natl. Acad. Sci. U.S.A.* **116**, 15414–15419.
33. S. C. Goheen *et al.*, Eds., "Sequential Separation of Americium and Plutonium by Extraction Chromatography" in *DOE Methods for Evaluating Environmental and Waste Management Samples* (Pacific Northwest Laboratory, Richland, WA, 1997), RP800.