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Marshall Islands Program Field Operations Report

Individual Radiological Protection Monitoring of Utrōk Atoll Residents Based on Whole-Body Counting of Cesium-137 (^{137}Cs) and Plutonium Bioassay

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Executive Summary

This report contains individual radiological protection surveillance data developed during 2006 for adult members of a select group of families living on Utrōk Atoll. These Group I volunteers all underwent a whole-body count to determine levels of internally deposited cesium-137 (^{137}Cs) and supplied a bioassay sample for analysis of plutonium isotopes. Measurement data were obtained and the results compared with an equivalent set of measurement data for ^{137}Cs and plutonium isotopes from a second group of adult volunteers (Group II) who were long-term residents of Utrōk Atoll. For the purposes of this comparison, Group II volunteers were considered representative of the general population on Utrōk Atoll. The general aim of the study was to determine residual systemic burdens of fallout radionuclides in each volunteer group, develop data in response to addressing some specific concerns about the preferential uptake and potential health consequences of residual fallout radionuclides in Group I volunteers, and generally provide some perspective on the significance of radiation doses delivered to volunteers (and the general Utrōk Atoll resident population) in terms of radiological protection standards and health risks.

Based on dose estimates from measurements of internally deposited ^{137}Cs and plutonium isotopes, the data and information developed in this report clearly show that neither volunteer group has acquired levels of internally deposited fallout radionuclides specific to nuclear weapons testing in the Marshall Islands that are likely to have any consequence on human health. Moreover, the dose estimates are well below radiological protection standards as prescribed by U.S. regulators and international agencies, and are very small when compared to doses from natural sources of radiation in the Marshall Islands and the threshold where radiation health effects could be either medically diagnosed in an individual or epidemiologically discerned in a group of people.

In general, the results from the whole-body counting measurements of ^{137}Cs are consistent with our knowledge that a key pathway for exposure to residual fallout contamination on Utrōk

Atoll is low-level chronic uptake of ^{137}Cs from the consumption of locally grown produce (Robison *et al.*, 1999). The error-weighted, average body burden of ^{137}Cs measured in Group I and Group II volunteers was 0.31 kBq and 0.62 kBq, respectively. The associated average, annual committed effective dose equivalent (CEDE) delivered to Group I and Group II volunteers from ^{137}Cs during the year of measurement was 2.1 and 4.0 mrem. For comparative purposes, the annual dose limit for members of the public as recommended by the National Council on Radiation Protection and Measurements (NCRP) and the International Commission on Radiological Protection (ICRP) is 100 mrem. Consequently, specific concerns about elevated levels of ^{137}Cs uptake and higher risks from radiation exposure to Group I volunteers would be considered unfounded. Moreover, the urinary excretion of plutonium-239 (^{239}Pu) from Group I and Group II volunteers is statistically indistinguishable. In this case, the error-weighted, average urinary excretion of ^{239}Pu from Group I volunteers of 0.10 μBq per 24-h void with a range between -0.01 and 0.23 μBq per 24-h void compares with an error-weighted average from Group II volunteers of 0.11 μBq per 24-h void with a range between -0.20 and 0.47 μBq per 24-h void. The range in urinary excretion of ^{239}Pu from Utrōk Atoll residents is very similar to that observed for other population groups in the Marshall Islands (Bogen *et al.*, 2006; Hamilton *et al.*, 2006a; 2006b; 2006c, 2007a; 2007b; 2007c) and is generally considered representative of worldwide background.

The corresponding average CEDE delivered to Group I volunteers from internally deposited ^{137}Cs and ^{239}Pu is estimated to be around 2.1 mrem and 1.2 mrem, respectively. The total CEDE (i.e., 3.3 mrem for the current year of measurement) is therefore very small when compared to the threshold where radiation health effects could be either medically diagnosed in an individual or epidemiologically discerned in a group of people. Of significance and perhaps most importantly, Group I volunteers from the *special interest group*¹ of families identified above can be assured that the measurement data contained in this report and the continuing radiological protection monitoring program clearly show that they have not acquired levels of internally deposited fallout radionuclides that are likely to have any consequence on human health. It is also of interest to note that the total dose based on estimates of internally deposited ^{137}Cs and ^{239}Pu in Group I volunteers (i.e., 3.3 mrem CEDE for the current year of measurement) is well below the annual safety standard of 15 mrem as imposed by the Marshall Islands Nuclear Claims Tribunal (NCT). The same would be true even if we were to consider the

¹ A group of family members identified by community leaders as potential program participants and constituted all the Group I volunteers.

uncertainty and variability in dose estimates. For example, previous studies show that the upper and lower 95% confidence limits on inter-individual variability in dose estimates based on environmental data typically lie within a ~threefold factor of the population average value (Bogen *et al.*, 1997). We have demonstrated that this is also largely true of the variability seen in dose estimates based on direct measurements of internally deposited radionuclides. This would imply that the CEDE delivered to Group I volunteers could range from 1 to 9 mrem where the maximum estimated CEDE of 9 mrem is still less than the 15 mrem annual safety standard adopted by the NCT. Moreover, while this comparative study is limited to adult volunteers, dose estimates for the general population on Utrōk Atoll that include children, teenagers and adults from both sexes will likely fall within a similar range. Any further conjecture about current and potential future radiation risks to the resident population from radiation exposure can be most rigorously assessed by continuation of the radiological protection monitoring program.

Background

This preliminary report contains comparative individual radiological protection surveillance data for Utrōk Atoll residents obtained from whole body counting and plutonium bioassay. Measurements of internally deposited ^{137}Cs and plutonium isotopes are compared between two groups of adult volunteers. Local atoll government representatives identified an initial group of potential candidates ($N=8$) for participation in the radiological surveillance monitoring program based on expressions of concern that these families had been exposed to elevated levels of fallout contamination in the environment. The second group of volunteers ($N=12$) from Utrōk Atoll was used

as a comparison group and simply represented the next subgroup of volunteers from Utrōk Atoll who expressed interest in participating in the program. The primary selection criterion for Group II program participation was a requirement that all volunteers should be long-term residents of Utrōk Atoll so that they could be considered as representative of the general population. A total of 12 adult volunteers participated in this second phase of the program but we were unable to obtain a whole-body count from one individual. All volunteers had to be willing to travel to Majuro Atoll to receive a whole-body count and provide a 24-h (urine) void bioassay sample.

Methodology

A full description of the radiological surveillance monitoring program on Utrōk Atoll can be found elsewhere (Hamilton *et al.*, 2007d). Whole-body counts and plutonium bioassay measurements were performed on Group I and Group II volunteers at different times but using identical, standardized procedures. The Utrōk whole-body counting facility and plutonium bioassay collection program are based on Majuro Atoll because of the lack of infrastructure support on Utrōk Atoll. As a consequence, program volunteers were all required to travel to Majuro Atoll and stay at a local hotel until the necessary arrangements could be made for each volunteer to receive a whole-body count and provide a 24-h (urine) void sample for bioassay. Travel and accommodation were provided at no expense to the volunteers. The hotel also served as the main collection point where bioassay samples could be collected under a relatively clean and controlled environment. The whole-body counting operation and plutonium bioassay collections were conducted by trained Marshallese technicians based on documented procedures developed by researchers from the Lawrence Livermore National Laboratory (LLNL). The bioassay samples were then taken to the Majuro whole-body counting facility, stabilized by

addition of acid, and shipped to the Lawrence Livermore National Laboratory for analysis by accelerator mass spectrometry (AMS) (Hamilton *et al.*, 2004; 2007d)

The most abundant isotopes of plutonium in radioactive debris generated from nuclear weapons testing are ^{239}Pu and plutonium-240 (^{240}Pu). The relative sensitivity of mass spectrometry for low-level determination of activity concentrations of ^{239}Pu and ^{240}Pu in bioassay samples will depend on several factors including the specific activity and relative abundance of the isotopes concerned. Based on the mass spectrometric measurements developed for this report, the estimated Minimal Detectable Amount (MDA) for ^{239}Pu and ^{240}Pu was about 0.4 and 1.0 μBq , respectively. We have therefore largely limited our discussion of results to higher precision measurements of ^{239}Pu but include both isotopes in data tables. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio measured in surface soil samples from Utrōk Atoll average around 0.27 (Hamilton, 2007e) and compare with an average reported $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in integrated worldwide-fallout deposition of 0.18 (Krey *et al.*, 1976). The corresponding $^{240}\text{Pu}/^{239}\text{Pu}$ ratios expressed on an activity basis are 0.66 and 1.0, respectively. This implies that dose estimates for Group I and II volunteers

may be as much as twice the values based exclusively on urinary excretion of ^{239}Pu .

The Marshall Islands Radiological Surveillance Monitoring Program (MIRSMP) is approved by the Human Subjects Committee of the Institutional Review Board at the Lawrence Livermore National Laboratory. Moreover, the quality assurance requirements for the operation of whole-body counting facilities in the Marshall Islands, and for the collection and measurement plutonium in bioassay samples, have been

designed to conform with standard requirements of the United States (U.S.) Department of Energy Laboratory Accreditation Program (DOELAP) for occupational monitoring of DOE workers (Hamilton *et al.*, 2007c; Kehl *et al.*, 2007). The use of AMS for low-level plutonium bioassay measurements has also been independently validated by the U.S. National Institute of Standards and Technology (NIST) (McCurdy *et al.*, 2004).

Results and Discussion

The results of whole-body counting measurements of internally deposited ^{137}Cs in the Group I and Group II volunteers are shown in Table 1. With the exception of one individual from Group I, a small but measurable quantity of internally deposited ^{137}Cs was observed in all program volunteers. A number of other whole-body counting measurements of internally ^{137}Cs are at or below the MDA above which a determination can be made with reasonable confidence that ^{137}Cs would otherwise actually be present. Generally, Marshall Islanders are potentially exposed to elevated levels of residual fallout contamination in the environment and so, for program volunteers living on Utröök Atoll, it was not unusual to find measurable quantities of ^{137}Cs in the vast majority of

program participants (Hamilton *et al.*, 2007d).

The error-weighted, average internally deposited ^{137}Cs in Group I volunteers of 0.31 kBq (N=8) compares with an error-weighted average of 0.62 kBq (N=11) in Group II volunteers. Based on the results presented in this report, Group I program volunteers have actually acquired, on average, slightly lower levels of ^{137}Cs compared with the general population living on Utröök Atoll. Consequently, specific concerns about elevated levels of ^{137}Cs uptake and higher risks from radiation exposure to Group I volunteers would be considered unfounded. For further comparative purposes, between 2005 and 2006 over 100 whole-body counts were

Table 1. Internally deposited ^{137}Cs activity (kBq \pm 1 SD) in Group I and Group II volunteers from Utrōk Atoll in the Marshall Islands.

Personal ID	Age Type	Sex	Collection Date	^{137}Cs (kBq)		
				value	MDA	
Group I Volunteers						
UT00160	Adult	Male	2006-03-21	0.56 \pm 0.07	0.32	
UT00212	Adult	Female	2006-03-21	0.00 \pm 0.05 [#]	0.11	
UT00213	Adult	Female	2006-03-21	0.10 \pm 0.04	0.20	
UT00214	Adult	Female	2006-03-21	0.21 \pm 0.07	0.33	
UT00215	Adult	Female	2006-03-21	0.41 \pm 0.07	0.32	
UT00216	Adult	Male	2006-03-21	0.67 \pm 0.07	0.31	
UT00217	Adult	Male	2006-03-21	0.63 \pm 0.08	0.33	
UT00218	Adult	Male	2006-03-21	0.56 \pm 0.07	0.30	
error-weighted average =				0.32 kBq		
Group II Volunteers						
UT00012	Adult	Male	2006-10-25	0.48 \pm 0.07	0.30	
UT00029	Adult	Male	2006-10-25	1.41 \pm 0.09	0.34	
UT00034	Adult	Male	2006-10-25	1.06 \pm 0.09	0.38	
UT00058	Adult	Male	2006-10-25	0.71 \pm 0.07	0.32	
UT00061	Adult	Male	2006-07-12	0.52 \pm 0.06	0.28	
UT00186	Adult	Male	2006-10-25	1.07 \pm 0.09	0.36	
UT00189	Adult	Male	2006-10-26	0.83 \pm 0.09	0.37	
UT00226	Adult	Female	2006-07-10	0.10 \pm 0.04	0.20	
UT00248	Adult	Male	2006-10-25	0.68 \pm 0.08	0.35	
UT00249	Adult	Male	2006-10-25	0.83 \pm 0.08	0.34	
UT00250	Adult	Male	2006-10-25	0.93 \pm 0.09	0.38	
error-weighted average =				0.62 kBq		

[#] the uncertainty in the measurement was given a value equating to half the Minimum Detectable Amount (MDA), i.e., MDA/2

performed on Utrōk Atoll residents (including non-members of the Utrōk Atoll population group) (Hamilton *et al.*, 2007d). The error-weighted, population-average ^{137}Cs body burden based on these measurements was 0.46 ± 0.04 kBq. The associated population-average, annual

CEDE contribution from ^{137}Cs is estimated to be around 3.5 mrem (Hamilton *et al.*, 2007d). The estimated CEDE from ^{137}Cs for Group I volunteers was slightly less, and averaged around 2.1 mrem with a maximum individual dose of 3.4 mrem. For comparative purposes, the CEDE

contribution from internally deposited ^{137}Cs in the Group II volunteers during the year of measurement was 4.0 mrem.

Residents of the Northern Hemisphere are all expected to acquire a small systemic burden of plutonium from general exposure to worldwide-fallout contamination to produce daily urinary excretion rates of around 2-4 μBq (Boecker *et al.*, 1991). In the northern Marshall Islands, long-term chronic exposure to local or regional fallout contamination and significant incremental intakes of plutonium associated with specific activities may increase the urinary excretion of plutonium over that expected from exposure to worldwide-fallout contamination.

The results of the plutonium bioassay measurements on Group I and Group II volunteers are shown in Table 2. The methodologies employed at LLNL for plutonium bioassay are extremely sensitive. While we were able to confirm the presence of trace amounts of ^{239}Pu in bioassay samples collected from Utrōk Atoll volunteers, the levels were barely detectable above our field-blank control samples (Table 2). None of the bioassay samples contained reported ^{240}Pu concentrations above the measurement MDA. In general, the results clearly demonstrate that none of the volunteers have acquired residual systemic burdens of plutonium in excess of historical estimates of urinary excretion of plutonium (Boecker *et al.*, 1991) based systemic

burdens of plutonium acquired from general exposure to worldwide fallout contamination in the Northern Hemisphere.

The error-weighted, average urinary excretion of ^{239}Pu from Group I volunteers was 0.10 μBq per 24-h void with a range between -0.01 and 0.23 μBq per 24-h. This compares with an error-weighted, average urinary excretion of ^{239}Pu from Group II volunteers of 0.11 μBq per 24-h void with a range between -0.20 and 0.47 μBq per 24-h void. The average urinary excretion of plutonium from these two groups is statistically indistinguishable and provide further evidence that Group I volunteers have not acquired residual systemic burdens of fallout radionuclides that would be considered in excess of Group II volunteers. Moreover, plutonium has a relatively long biological half-life of 20 to 50 years. Systemic plutonium acquired from previous exposures will be very slowly removed from the body, which occurs primarily in urine and with a long-term excretion coefficient of $\sim 2 \times 10^{-5}$ of the systemic burden per day. Therefore, the low-level urinary excretion of plutonium from Utrōk Atoll residents also implies that the program volunteers have never acquired a significant systemic burden of plutonium.

Under steady-state conditions (Daniels *et al.*, 2007), the urinary excretion of ^{239}Pu from Group I and Group II volunteers yield

systemic burdens of ^{239}Pu that equate to annual doses of around 24 μrem and 26 μrem (where 1 μrem = 0.001 mrem), respectively, or an annual CEDE of 1.2 mrem and 1.3 mrem, respectively. Even taking into account a doubling of the

estimated dose from undetectable systemic burdens of ^{240}Pu , we conclude that the dose contribution from plutonium is very low and represents a very small fraction of the dose received from natural background radiation in the Marshall Islands.

Conclusion

During March of 2006, a small number of adults from a select group of families living on Utrōk Atoll participated in the MIRSMP using facilities located on Majuro Atoll. These Group I volunteers all underwent whole-body counting to determine levels of internally deposited ^{137}Cs and supplied a urine bioassay sample for analysis of plutonium isotopes. Measurement data were obtained and the results compared with an equivalent set of measurement data for ^{137}Cs and plutonium isotopes from a second group of adult volunteers (Group II) who were long-term residents of Utrōk Atoll. Based on measurements of internally deposited ^{137}Cs and plutonium isotopes, the dosimetric data and information developed in this report clearly show that neither volunteer group acquired levels of internally deposited fallout radionuclides specific to nuclear weapons testing in the Marshall Islands that are likely to have any consequence on human health.

The total annual average CEDE delivered to Group I volunteers from internally deposited ^{137}Cs and ^{239}Pu was

estimated to be 3.3 mrem. The dose limit for members of the public currently recommended by the NCRP and the ICRP, and adopted in the Basic Safety Standards (FAO *et al.*, 1996), is 100 mrem per year. The same numeric dose limit for protection of the public is specified by the U.S. Nuclear Regulatory Commission (NRC) under 10 CFR Part 20 (NRC, 1994). In order to ensure that the dose limit will be met, dose constraints on individual sources and/or practices are often established at a fraction of the dose limit (ICRP, 1991; NCRP, 1993) such as those applied to remediation of radioactive contamination at licensed facilities under the License Termination Rule (LTR) 10 CFR Part 20, Subpart E (NRC, 2004). For example, a site may be considered acceptable for unrestricted use if the total annual effective dose equivalent (TEDE; a form of CEDE) from all exposures does not exceed 25 mrem above normal background and concentrations of residual radioactive material have been reduced to levels as low as reasonably achievable (ALARA). Annual dose criterion for cleanup

of radioactively contaminated sites or, as in this case, the TEDE, usually refer to the sum of the deep-dose equivalent from external exposures during the year of measurement and the 50-y CEDE from intakes of radionuclides during that year.

An alternative approach to gaining some perspective on the radiation dose estimates is to present the results in terms of individual risk. The U.S. Environmental Protection Agency (EPA) uses a risk-based approach to remediation of radioactively contaminated sites based on excess lifetime risks of fatal cancer and cancer incidence. The EPA uses a lifetime cancer risk criterion with an upper bound of 10^{-4} and provides guidance that an annual effective dose equivalent of 15 mrem or less would normally comply with the risk goal. The doses delivered to volunteers from Utrōk Atoll equate to an excess lifetime risk of *fatal* cancer of around 1×10^{-4} or 0.01% (1 in 10,000 exposed individuals). This may be compared to the excess *fatal* cancer risk from background radiation in the Marshall Islands of around 5×10^{-3} or 0.5% (5 per 1000 exposed individuals) and is a very small fraction of the underlying lifetime risk of death from cancer in the United States of around 25%. It should also be noted that under the current system for radiological protection, practices giving rise to annual effective doses of less than 1 mrem are commonly exempted from regulatory requirements on the basis that the doses are

so small that they can be considered negligible (IAEA, 1988) and the excess lifetime risk of fatal cancer *de minimis*.

Based on measurements of internally deposited ^{137}Cs and ^{239}Pu , the average dose delivered to Group I and Group II volunteers from Utrōk Atoll easily satisfy the 100 mrem annual dose criterion (above background and described above) for protection of members of the public and more closely approximates doses that are so small they are considered trivial and exempt from further consideration. This assertion is made on the basis that ^{137}Cs is known to be a major contributor to the total manmade dose in the Marshall Islands with a less significant contribution from plutonium isotopes. However, no consideration has been given to the external dose contribution or the CEDE from potential intakes of other fallout radionuclides such as strontium-90 (^{90}Sr).

In general, radiological protection standards provide a very conservative approach to protecting public health and the environment. The dose estimates presented in this report are already well below radiation safety standards as recommended by U.S. regulators and the community of international agencies, and are very small when compared to the threshold where radiation health effects could be either medically diagnosed in an individual or epidemiologically discerned in a group of

people. Nonetheless, the observed incidence of cancer and other diseases is changing throughout the world including the Pacific Island nations. These changes may simply reflect improved screening and registration procedures but could also result from changes in lifestyle including diet or other personal habits (e.g., cigarette smoking), population migration, and increased environmental exposure to harmful chemicals including radioactive and non-radioactive substances. However, based on albeit limited radiological surveillance

monitoring data developed to date under the MIRSMP, we can provide assurances to the local resident population and the global community that levels of exposure to radioactive fallout contamination on Utrōk Atoll are very low and are likely to have no discernible impact on human health, and that it is most unlikely that any additional cancer fatalities above those normally expected will arise which can be directly attributable to current radiological exposure conditions on the atoll.

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Table 2. Internally deposited plutonium activity ($\mu\text{Bq} \pm 1 \text{ SD}$) in Group I and Group II volunteers from Utrök Atoll in the Marshall Islands.

ID#	Age Type	Gender	Collection Date	$\mu\text{Bq per 24 h void}$			
				^{239}Pu	^{240}Pu		
Group I Volunteers							
UT00160	Adult	Male	22-Mar-06	0.16	± 0.24	-0.17	± 0.57
UT00212	Adult	Female	24-Mar-06	-0.01	± 0.17	-0.17	± 0.50
UT00213	Adult	Female	22-Mar-06	0.21	± 0.21	-0.17	± 0.36
UT00214	Adult	Female	22-Mar-06	0.19	± 0.20	0.12	± 0.35
UT00215	Adult	Female	22-Mar-06	0.06	± 0.18	0.18	± 0.40
UT00216	Adult	Male	22-Mar-06	0.13	± 0.19	-0.17	± 0.37
UT00217	Adult	Male	22-Mar-06	0.23	± 0.24	-0.17	± 0.50
UT00218	Adult	Male	22-Mar-06	0.00	± 0.15	-0.17	± 0.31
error-weighted average =				0.10		-0.08	
Group II Volunteers							
UT00012	Adult	Male	23-Oct-06	0.30	± 0.23	-0.16	± 0.37
UT00029	Adult	Male	23-Oct-06	0.47	± 0.31	0.33	± 0.50
UT00034	Adult	Male	23-Oct-06	0.34	± 0.25	0.17	± 0.40
UT00058	Adult	Male	23-Oct-06	0.35	± 0.25	0.16	± 0.38
UT00061	Adult	Male	23-Oct-06	0.05	± 0.18	0.44	± 0.48
UT00062	Adult	Male	23-Oct-06	0.05	± 0.19	0.13	± 0.36
UT00186	Adult	Male	23-Oct-06	0.39	± 0.25	-0.16	± 0.37
UT00189	Adult	Male	23-Oct-06	0.20	± 0.26	-0.16	± 0.52
UT00226	Adult	Female	23-Oct-06	-0.20	± 0.16	-0.16	± 0.45
UT00248	Adult	Male	23-Oct-06	0.03	± 0.17	0.10	± 0.34
UT00249	Adult	Male	23-Oct-06	-0.04	± 0.16	-0.16	± 0.36
UT00250	Adult	Male	23-Oct-06	0.46	± 0.32	0.76	± 0.69
error-weighted average =				0.11		0.05	
Process Field Blanks[#]							
Field Blank	not applicable		24-Mar-06	-0.06	± 0.14	-0.17	± 0.33
Field Blank	not applicable		24-Mar-06	0.00	± 0.15	0.09	± 0.32
Field Blank	not applicable		28-Mar-06	0.00	± 0.15	-0.17	± 0.31
Field Blank	not applicable		28-Mar-06	-0.06	± 0.14	-0.17	± 0.34
Field Blank	not applicable		11-Aug-06	0.13	± 0.20	-0.16	± 0.36
Field Blank	not applicable		11-Aug-06	0.06	± 0.17	0.15	± 0.35
Field Blank	not applicable		11-Aug-06	-0.05	± 0.16	-0.16	± 0.34
error-weighted average =				-0.01		-0.09	

[#] Process field blanks were collected in the Marshall Islands and handled in exactly the same manner as bioassay samples. The results provide a measure of the background concentration of plutonium introduced as part of sample handling and analysis procedures.

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