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# Concentration of Beryllium (Be) and Depleted Uranium (DU) in Marine Fauna and Sediment Samples from Illeginni and Boggerik Islands at Kwajalein Atoll

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# Concentration of Beryllium (Be) and Depleted Uranium (DU) in Marine Fauna and Sediment Samples from Illeginni and Boggerik Islands at Kwajalein Atoll

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## Abstract

Lawrence Livermore National Laboratory (LLNL) personnel have supported United States Air Force (USAF) ballistic missile flight tests for about 15 years for Peacekeeper and Minuteman missiles launched at Vandenberg Air Force Base (VAFB). Associated re-entry vehicles (RV's) re-enter at Regan Test Site (RTS) at the U.S. Army base at Kwajalein Atoll (USAKA) where LLNL has supported scoring, recovery operations for RV materials, and environmental assessments.

As part of ongoing USAF ballistic missile flight test programs, LLNL is participating in an updated EA being written for flights originating at VFAB. Marine fauna and sediments (beach-sand samples) were collected by United States Fish and Wildlife Service (USFWS), National Marine Fisheries Service (NMFS), and LLNL at Illeginni Island and Boggerik Island (serving as a control site) at Kwajalein Atoll. Data on the concentration of DU (hereafter, U) and Be in collected samples was requested by USFWS and NMFS to determine whether or not U and Be in RV's entering the Illeginni area are increasing U and Be concentrations in marine fauna and sediments. LLNL agreed to do the analyses for U and Be in support of the EA process and provide a report of the results.

There is no statistically significant difference in the concentration of U and Be in six species of marine fauna from Illeginni and Boggerik Islands ( $p = 0.14$  for U and  $p = 0.34$  for Be). Thus, there is no evidence that there has been any increase in U and Be concentrations in marine fauna as a result of the missile flight test program.

Concentration of U in beach sand at Illeginni is the same as soil and beach sand in the rest of the Marshall Islands and again reflects an insignificant impact from the flight test program.

Beach sand from Illeginni has a mean concentration of Be higher than that from the control site, Boggerik Island. Seven of 21 samples from Illeginni had detectable Be. Four samples had a concentration of Be ranging from 4 to 7 ng g<sup>-1</sup> [4 to 7 parts per billion (ppb)], one was 17 ppb, one was 0.14 parts per million (ppm), and one was 0.48 ppm. These extremely low concentrations of an insoluble form of Be again indicate no impact on marine life or human health at Illeginni as a result of the missile flight test program.

Concentration of Fe in marine fauna muscle tissue is much higher at Illeginni Island than at Boggerik Island (control site) as a result of legacy iron piers, dump sites for iron metal along the island, and scrap iron randomly distributed along extensive portions of the reef line as part of programs conducted in the 1960's through 1980's that were not part of the recent flight test program.

## Introduction

Lawrence Livermore National Laboratory (LLNL) personnel have supported United States Air Force (USAF) ballistic missile flight tests for more than 14 years for Peacekeeper and Minuteman missiles launched at Vandenberg Air Force Base (VAFB). Associated re-entry vehicles (RV's) re-enter at Regan Test Site (RTS) at the U.S. Army base at Kwajalein Atoll (USAKA) where LLNL has supported scoring, recovery operations for RV materials, and environmental assessments. An Environmental Assessment (EA) was written by LLNL for the USAF program in 1988 (Robison and Shinn, 1988), a more extensive EA was written in

1992 (Robison and Cederwall, 1992), and periodic reports have been submitted to USAKA and RTS commanders in the intervening years (Robison and Shinn, 1990; Robison, 1992). Also, letters have been submitted to the commanding officer of USAKA after each supported mission describing whether or not Illeginni Island was affected by the mission and if so, what the recovery operation would entail. Soil samples have been collected for U and Be analysis after every mission where U and Be were deposited on the island. In each case, hi-volume air samplers were run for a 4 to 5 week period to determine the

concentration of U and Be in air to compare with federal guidelines for the general public.

As part of ongoing USAF ballistic missile flight test programs, LLNL is participating in an updated EA being written for flights originating at VFAB. Marine fauna and sediments (beach-sand samples) were collected by United States Fish and Wildlife Service (USFWS), National Marine Fisheries Service (NMFS), and LLNL at Illeginni Island and Boggerik Island (serving as a control site) at Kwajalein Atoll. Data for concentration of U and Be in collected samples was requested by USFWS and NMFS to determine whether or not U and Be in RV's entering the Illeginni area are increasing U and Be concentrations in marine fauna and sediments. LLNL agreed to do the analyses for U and Be to

support the EA process and to provide a report of the results.

## Methods

### Collection and initial preparation of marine fauna

Different species of fish, eels and crabs were collected from reef flats at Illeginni Island and a control site at Boggerik Islet in August 2004 by staff from the NMF and USFWS. The dates of collection, type of sample, and number of specimens are listed in Table 1. Fauna samples were collected using traps, throw-nets and spears, and packed frozen ready for shipment to LLNL.

Table 1. Marine fauna collected at Illeginni and Boggerik Islands

Date	Location	Species	No.
Illeginni Island			
06/08/04	Ocean side	Snowflake eel ( <i>Siderea picta</i> )	3
06/09/04	Ocean side	Snowflake eel ( <i>Siderea picta</i> )	6
06/09/04	Ocean side	Rock crab ( <i>Graspsus tenuicrustatus</i> )	15
06/09/04	Ocean side	White-tip shark liver ( <i>Triaenodon obesus</i> )	1 <sup>a</sup>
06/09/04	Ocean side	Black-tip shark liver ( <i>Carcharhinus melanopterus</i> )	1 <sup>a</sup>
06/09/04	Lagoon side	Snowflake eel ( <i>Siderea picta</i> )	6
06/09/04	Lagoon side	Rock crab ( <i>Graspsus tenuicrustatus</i> )	12
06/10/04	Ocean side	Snowflake eel ( <i>Siderea picta</i> )	1
06/10/04	Lagoon side	Snowflake eel ( <i>Siderea picta</i> )	4
06/10/04	Lagoon side	Rock crab ( <i>Graspsus tenuicrustatus</i> )	2
06/10/04	Lagoon side	Farmer fish ( <i>Stegastus</i> sp)	38
a = whole liver			
Boggerik Island, Control Site			
06/6,7/04	Boggerik	Moray eel ( <i>Gymnothorax jaranicus</i> )	1
06/6,7/04	Boggerik	Snowflake eels ( <i>Siderea picta</i> )	6
06/6,7/04	Boggerik	Rock crab ( <i>Graspsus</i> sp)	16
06/6,7/04	Boggerik	Farmer fish ( <i>Stegastus</i> sp)	6
06/6,7/04	Roi Namur	Farmer fish ( <i>Stegastus</i> sp)	5

At LLNL, the marine fauna samples were thawed and rinsed to remove any adhering sand or other debris before being placed into clean photo trays for dissection. Muscle tissue was dissected from all species along with liver tissue from the larger fish and eels. Care was exercised to prevent

cross-contamination between tissue types. The dissected material from each composite sample was diced, placed in plastic freezer containers and dried by lyophilization. The dried material was then re-weighed to estimate the wet/dry ratio of tissue, and then homogenized in a laboratory blender

### Collection and initial preparation of sediment (beach sand)

Beach sands at Illeginni Island and Boggerik Islet were collected using plastic cups, the material placed in plastic bags, and shipped frozen to LLNL for analysis. NMF and NFWS personnel initially selected a series of sites along Illeginni for collecting beach-sand samples (red triangles in Fig. 1). Additional sites along the ocean-side reef (black triangles in Fig. 1) were selected by LLNL personnel to provide additional coverage of the down-wind, down-trajectory side of the island. The sample site coordinates for Illeginni Island are listed in Table 2. NMF and NFWS personnel were responsible for collecting beach sand at Boggerik Islet control site on both ocean and lagoon sides of the islet.

To prepare samples for chemical analysis, the bulk sediments were placed in metal cans and dried by lyophilization. Lids were placed on the cans and the dried material homogenized for 24-h by agitation on a rolling mill. Large shell fragments and pebbles were removed prior to sample analysis by sieving samples through a 16-mesh (1.6 mm) NBS screen.

### Chemical Processing

Chemical analyses of marine fauna were performed on 10 g aliquots of dried material or as much material as available. The samples were initially weighted into 500 ml glass beaker and dry ashed in an oven for 64 hours. The ashed residues was then transferred to acid leached polypropylene

digestion vessels and dissolved by heating on a hot-block with addition of 40 g of a 1:1 mixture of concentrated nitric acid and 30% hydrogen peroxide. Sample dissolution was continued with periodic additions of a few milliliters of hydrogen peroxide until the digest was clear and colorless.

Sediment samples were treated in a similar fashion using 1 g aliquots of sample matrix and addition of 20 g of the concentrated nitric acid and hydrogen peroxide mixture. Total dissolution of sediments was insured by dissolution of any residue in nitric-hydrofluoric acid. All samples were then evaporated to near-dryness, re-hydrated in 2% ultra-pure nitric acid, and filtered through a 0.45  $\mu\text{m}$  syringe filter to constant volume. Appropriate gravimetric dilutions were then prepared for chemical analysis by serial dilution of the stock digest.

Be) and U were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) with addition of 1 ppm of lithium-6 ( $^6\text{Li}$ ) (for Be analysis) and 1 ppb of uranium-233 ( $^{233}\text{U}$ ) (for U analysis) as internal standards. Iron and calcium were analyzed separately using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). Control samples included blanks and approximately 10% of blind replicates prepared from the stock digest. A series of MAPEP inter-comparison soils were also prepared and analyzed as quality control samples for the Be and U analyses (reported separately).

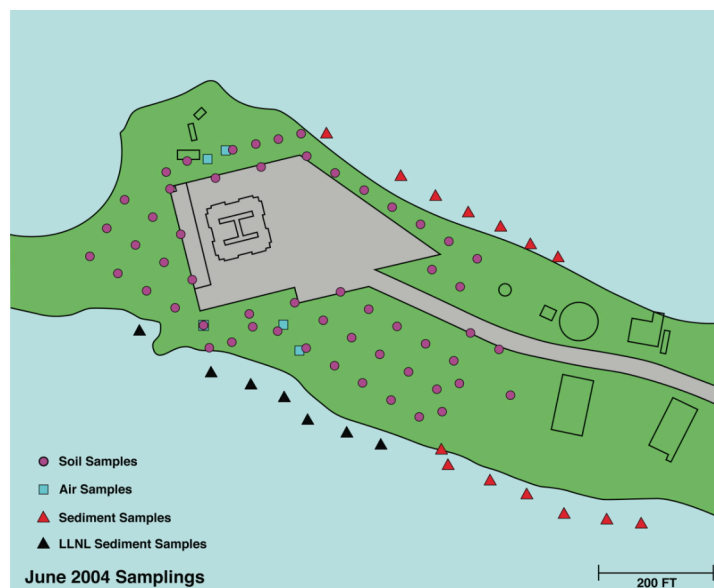


Fig.1 Marine sediment (beach sand) sample locations are identified by color triangles. Also on the figure are terrestrial soil sample sites and hi-volume air sampling locations.

Table 2. Location and coordinates of marine sediment (beach sand) collected at Illeginni Island

Date	Location	Latitude	Longitude
06/09/04	Ocean side	9° 5' 10.32"	167° 28' 17.44"
06/09/04	Ocean side	9° 5' 10.40"	167° 28' 16.84"
06/09/04	Ocean side	9° 5' 10.50"	167° 28' 16.10"
06/09/04	Ocean side	9° 5' 10.84"	167° 28' 15.44"
06/09/04	Ocean side	9° 5' 11.08"	167° 28' 14.80"
06/09/04	Ocean side	9° 5' 11.35"	167° 28' 14.05"
06/09/04	Ocean side	9° 5' 11.61"	167° 28' 13.94"
06/09/04	Ocean side	9° 5' 13.69"	167° 28' 8.67"
07/18/04	Ocean side	9° 5' 11.77"	167° 28' 12.84"
07/18/04	Ocean side	9° 5' 12.00"	167° 28' 12.25"
07/18/04	Ocean side	9° 5' 12.28"	167° 28' 11.64"
07/18/04	Ocean side	9° 5' 12.57"	167° 28' 11.21"
07/18/04	Ocean side	9° 5' 12.75"	167° 28' 10.59"
07/18/04	Ocean side	9° 5' 13.02"	167° 28' 9.92"
06/09/04	Lagoon side	9° 5' 15.00"	167° 28' 16.00"
06/09/04	Lagoon side	9° 5' 15.22"	167° 28' 15.49"
06/09/04	Lagoon side	9° 5' 15.53"	167° 28' 14.98"
06/09/04	Lagoon side	9° 5' 15.78"	167° 28' 14.40"
06/09/04	Lagoon side	9° 5' 16.07"	167° 28' 13.84"
06/09/04	Lagoon side	9° 5' 16.42"	167° 28' 13.23"
06/09/04	Lagoon side	9° 5' 17.17"	167° 28' 11.94"

### Statistical analysis of the data

The Mann-Whitney U test (Siegel, 1956), a powerful non-parametric statistical test to determine if two independent sample sets were drawn from the same population, was used to evaluate marine fauna and sediment data sets. Non-parametric statistical tests are more powerful for cases where the number of samples in the set is small and the distribution is unknown. The Mann-Whitney U test is far more powerful than parametric tests such as the Student T-test under such conditions. When the number of samples in one of the two sets of data exceeded 20, the Mann-Whitney U statistic was used to calculate the Z statistic (a parametric test) to determine the probability under the null hypothesis.

### Uranium Concentration in Coral soils

Concentration of U in coral soil in the Marshall Islands is discussed in detail in Robison et al., 2001. A brief summary is given here with tables

abstracted from Robison et al., 2001. The mean concentration of U in several closed system corals (live functioning coral reefs) from around the world is  $2.9 \mu\text{g g}^{-1}$  (Table 3). Uranium concentration decreases in open-system coral soils and beach sand (Thurber et al., 1965) as is evident in the results shown in Tables 4 through 7. Concentration of U ranges between  $1.6$  to  $2.0 \mu\text{g g}^{-1}$  for 3220 coral sands and soils measured in the northern Marshall Islands including results from the Republic of the Marshall Islands Nationwide Radiological Survey (RMI-NRS). Other results from the RMI-NRS show a mean value for U of  $2.1 \mu\text{g g}^{-1}$  for atolls arbitrarily classed as mid-atolls (Table 7) although the mean value for Kwajalein was  $1.7 \mu\text{g g}^{-1}$ . The mean value for the southern atolls was a little higher again with a mean of  $2.6 \mu\text{g g}^{-1}$ . So, a mean U concentration of  $1.6$  to  $2.0 \mu\text{g g}^{-1}$  in coral soil and beach sand is well established in the northern Marshall Islands and some of the more northern of the mid-atolls.

Table 3. Concentration of U ( $\pm 1$  standard error) in corals from global locations

Location	Number of $^{238}\text{U}$ samples	Mean $\mu\text{g g}^{-1}$	Source
Enewetak	36	$2.9 \pm 0.1$	Thurber et al., 1965
Florida Keys	15	$2.7 \pm 0.1$	Broecker & Thurber, 1965
Hawaii <sup>a</sup>	6	$2.5 \pm 0.2$	Veeh & Turekian, 1968
Samoa <sup>b</sup>	5	$2.3 \pm 0.2$	Veeh & Turekian, 1968
Tahiti <sup>b</sup>	5	$2.1 \pm 0.1$	Veeh & Turekian, 1968
Tuamotub	8	$1.8 \pm 0.1$	Veeh & Turekian, 1968
Enewetak	18	$3.9 \pm 0.1$	Barnes et al., 1956
Florida Keys <sup>c</sup>	2	$2.9 \pm 0.4$	Tatsumoto & Goldberg, 1959

<sup>a</sup> Three samples each of two genus of corals.

<sup>b</sup> Three samples of one genus of coral and two samples of another genus of coral

<sup>c</sup> Two genus of corals

Table 4. The concentration  $\pm 1$  standard error of  $^{238}\text{U}$  in coral soil from Majuro, Kwajalein, Wotje, Ponape, and Truk Atolls

Location	Number of samples	Mean $\mu\text{g g}^{-1}$	Source
Majuro <sup>a</sup>	8	$2.2 \pm 0.3^e$	University of Washington (UW), Nelson <sup>f</sup> , 1979a
Majuro <sup>a</sup>	5	$1.7 \pm 0.2$	LLNL <sup>g</sup> , Robison et al., 2001
Majuro <sup>b</sup>	40	$1.7 \pm 0.1$	LLNL <sup>f</sup> , Robison et al., 2001
Wotje <sup>c</sup>	17	$2.1 \pm 0.1$	UW, Nelson <sup>f</sup> , 1979b
Kwajalein <sup>d</sup>	47	$1.9 \pm 0.1$	LLNL <sup>g</sup> , Robison et al., 2001
Ponape	12	$2.8 \pm 0.3$	UW, Nelson <sup>f</sup> , 1979a
Truk	13	$3.6 \pm 0.3$	UW, Nelson <sup>f</sup> , 1979a
All samples	149	$2.1 \pm 0.1$	

<sup>a</sup> Majuro to Laura.

<sup>b</sup> Majuro, Enemanet, and Eneko Islands

<sup>c</sup> Wormej and Wotje Islands.

<sup>d</sup> Gagan, Gellinam, Roi Namur, and Illeginni Islands.

<sup>e</sup> Standard error of the mean.

<sup>f</sup> Data generated by gamma spectroscopy.

<sup>g</sup> Data generated by ICP mass spectrometry.



Table 5. The concentration  $\pm 1$  standard error of  $^{238}\text{U}$  in soil and lagoon sediments at the northern Marshall Islands

Location	Number of samples	Mean $\mu\text{g g}^{-1}$	Source	Method
Bikini Islands <sup>a</sup>	100	2.1 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
Rongelap Island	15	1.7 $\pm$ 0.1	LLNL, Robison et al., 2001	mass spec.
Rongelap northern islands	47	1.4 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
Rongelap southern islands	177	1.7 $\pm$ 0.05	LLNL, Robison et al., 2001	gamma spec.
Mejit Island	43	1.6 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
Ailuk Islands <sup>b</sup>	16	2.2 $\pm$ 0.3	UW, Nelson, 1979b	gamma spec.
Ailuk Islands <sup>c</sup>	99	1.9 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
Utirik Islands <sup>d</sup>	6	2.9 $\pm$ 0.5	UW, Nelson, 1979b	gamma spec.
Utirik Islands <sup>d</sup>	547	1.6 $\pm$ 0.05	LLNL, Robison et al., 2001	gamma spec.
Likiep Island <sup>e</sup>	68	2.3 $\pm$ 0.2	LLNL, Robison et al., 2001	gamma spec.
Enewetak Atoll <sup>f</sup>	183	1.9 $\pm$ 0.05	LLNL, Robison et al., 2001	gamma spec.
Rongerik Atoll <sup>g</sup>	25	1.5 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
All samples	1328	1.8 $\pm$ 0.05		
Bikini lagoon <sup>h</sup>		2.8 $\pm$ 0.1	Marshall and Schell, 1974 LLNL, Robison et al., 2001	alpha spec. mass spec. and gamma spec.
Rongelap lagoon	21	2.7 $\pm$ 0.1	LLNL, Robison et al., 2001	gamma spec.
Enewetak lagoon	76	4.0 $\pm$ 0.2	LLNL, Robison et al., 2001	gamma spec.
		Reference Value	Range	
Eneu Island <sup>i</sup>	1	1.6 $\mu\text{g g}^{-1}$	0.9 to 3 $\mu\text{g g}^{-1}$	IAEA AQCS, 1998/99

<sup>a</sup> Weighted mean of Nam(13), Iroij(4), Odrik(1), Lomilik(2), Bikini(7), Aerokojlo(36), Lele(5), Eneman(3), Enedrik(16), Lukoj(9), and Jelele(4).

<sup>b</sup> Ailuk and Bigen Islands.

<sup>c</sup> Kapen, Enijabro, Enejelar, Bigen, and Aliet Islands.

<sup>d</sup> Aon and Eerukku Islands, for Nelson; Utirik, Aon, Bigrak, Eerukku Islands for LLNL.

<sup>e</sup> Likiep, Agohy, and Etoile Islands.

<sup>f</sup> Enjebi Island.

<sup>g</sup> Jedibberbid, Latoback, Rongerik, Eniwetak, and Bock Islands

<sup>h</sup> No of samples; Marshall & Shell-11; Noshkin-20; Noshkin-58

<sup>i</sup> Homogenized soil sample sent to International Atomic Energy Agency (IAEA); 69 laboratories around the world analyzed the sample

Table 6. The concentration  $\pm 1$  standard error of  $^{238}\text{U}$  in soil at Enewetak Atolla

Location	Number of samples	Mean $\mu\text{g g}^{-1}$
Southern Islands		
Anniji	29	1.4 $\pm$ 0.1
Japtan	131	2.0 $\pm$ 0.1
Medren	304	1.7 $\pm$ 0.1
Enewetak	422	1.7 $\pm$ 0.1
All samples	886	1.8 $\pm$ 0.05
Northern and Eastern Islands		
Aje	15	1.8 $\pm$ 0.2
Lujor	45	1.4 $\pm$ 0.1
Aomen	25	1.8 $\pm$ 0.2
Bijire	46	1.5 $\pm$ 0.1
Lojwa	55	1.6 $\pm$ 0.1
Alembel	50	1.7 $\pm$ 0.1
Runit	70	1.5 $\pm$ 0.1
Enjebi	183	1.9 $\pm$ 0.1
All samples	489	1.7 $\pm$ 0.05

A from Robison et al., 2001

Table 7. Concentration ( $\pm 1$  standard error) of  $^{238}\text{U}$  in atoll soils in the Marshall Islands<sup>a</sup>

Southern Atolls	Mean $\mu\text{g g}^{-1}$	Mid-Atolls	Mean $\mu\text{g g}^{-1}$	Northern Atolls	Mean $\mu\text{g g}^{-1}$
Ailinglaplap (24) <sup>b</sup>	2.6 $\pm$ 0.20	Aur (24)	2.2 $\pm$ 0.1	Ailinginae (54)	1.6 $\pm$ 0.1
Ebon	2.2 $\pm$ 0.16	Kwajalein (54)	1.7 $\pm$ 0.1	Bikinu (340)	1.8 $\pm$ 0.1
Arno (24)	3.3 $\pm$ 0.16	Erikub (12)	1.6 $\pm$ 0.1	Ailuk (18)	1.9 $\pm$ 0.1
Jaluit (24)	2.2 $\pm$ 0.26	Lae (18)	2.5 $\pm$ 0.2	Enewetak (44)	1.6 $\pm$ 0.1
Kili (6)	3.0 $\pm$ 0.31	Lib (12)	2.4 $\pm$ 0.1	Mejit (11)	1.8 $\pm$ 0.1
Knox (6)	2.5 $\pm$ 0.31	Likiep (36)	2.2 $\pm$ 0.1	Rongelap (106)	1.7 $\pm$ 0.1
Majuro (72)	2.5 $\pm$ 0.16	Maleolap (24)	1.5 $\pm$ 0.1	Rongerik (28)	1.2 $\pm$ 0.1
Namorik (12)	2.2 $\pm$ 0.41	Namu (24)	2.7 $\pm$ 0.2	Taongi	1.8 $\pm$ 0.1
Mili (24)	2.5 $\pm$ 0.16	Ujae (18)	2.3 $\pm$ 0.2	Taka (24)	2.0 $\pm$ 0.1
Jabat (6)	2.8 $\pm$ 0.32	Ujelang (42)	2.4 $\pm$ 0.2	Utirik (25)	1.8 $\pm$ 0.1
.		Wotho (18)	1.9 $\pm$ 0.1	Jemo (6)	2.2 $\pm$ 0.3
		Wotje (24)	1.9 $\pm$ 0.1		
Total (216)	2.6 $\pm$ 0.08	Total (306)	2.1 $\pm$ 0.05	Total (368)	1.7 $\pm$ 0.05

<sup>a</sup> Data from the RMI Nationwide Radiological Survey conducted by Dr. Steve Simon<sup>1</sup> (see Robison et al., 2001)<sup>b</sup> Number of samples are in parentheses.<sup>1</sup> Dr. Steven L. Simon, National Cancer Institute, 6120 Executive Blvd., MSC7238, Executive Plaza South, Bethesda, MD 20892-7238

## Results

Results of analysis of U and Be are listed in Table 8 for marine fauna collected at Illeginni and Boggerik Islands. Concentrations are listed as  $\text{ng g}^{-1}$  and include means, medians, and standard deviations. Mean U concentration and associated standard deviation for all samples at Illeginni Island is  $9.8 \pm 5.3 \text{ ng g}^{-1}$  and is  $9.0 \pm 8.0 \text{ ng g}^{-1}$  for Boggerik

Island. Mean Be concentration and standard deviation for Illeginni and Boggerik is  $1.5 \pm 2.7 \text{ ng g}^{-1}$  and  $1.7 \pm 2.2 \text{ ng g}^{-1}$ , respectively. There is no statistically significant difference in the concentration of Be ( $p \geq 0.34$ ) and U ( $p \geq 0.14$ ) between Illeginni Island fauna samples and Boggerik Island control site fauna samples.

Table 8. Be and U concentration in all marine fauna at Illeginni and Boggerik Islands

Date	Location	Species	U $\text{ng g}^{-1}$	Be $\text{ng g}^{-1}$	Date	Location	Species	U $\text{ng g}^{-1}$	Be $\text{ng g}^{-1}$
Illeginni Island					Boggerik Island, Control Site				
06/08/04	Ocean	Snowflake eel liver	12	-0.34	06/6,7/04	Boggerik	Moray eel muscle	0.9	0.77
06/08/04	Ocean	Snowflake eel muscle	4.2	-0.43	06/6,7/04	Boggerik	Moray eel liver	8.3	1.3
06/09/04	Lagoon	Rock crab	6.8	7.2	06/6,7/04	Boggerik	Snowflake eels muscle	17	4.7
06/09/04	Lagoon	Snowflake eel liver	15	-0.085	06/6,7/04	Boggerik	Snowflake eels liver	21	0.024
06/09/04	Lagoon	Snowflake eel muscle	11	-0.70	06/6,7/04	Boggerik	Snowflake eels muscle	4.1	-0.17
06/09/04	Ocean	Black-tip shark liver	11	0.33	06/6,7/04	Boggerik	Snowflake eels liver	24	-0.37
06/09/04	Ocean	Rock crab	4.2	7.7	06/6,7/04	Boggerik	Rock crab	3.1	4.8
06/09/04	Ocean	Rock crab	5.4	5.6	06/6,7/04	Boggerik	Rock crab	6.0	4.3
06/09/04	Ocean	Snowflake eel liver	9.9	0.039	06/6,7/04	Boggerik	Rock crab	4.0	3.8
06/09/04	Ocean	Snowflake eel muscle	9.7	5.6	06/6,7/04	Boggerik	Farmer fish	8.1	-0.23
06/09/04	Ocean	White-tip shark liver	5.7	-0.036	06/6,7/04	Roi Namur	Farmer fish	2.3	-0.24
06/10/04	Lagoon	Farmer fish	3.4	1.2			<b>Mean</b>	<b>9.0</b>	<b>1.7</b>
06/10/04	Lagoon	Rock crab	11	1.2			<b>Stdev</b>	<b>8.0</b>	<b>2.2</b>
06/10/04	Lagoon	Snowflake eel muscle	14	-6.9			<b>Median</b>	<b>6.0</b>	<b>0.77</b>
06/10/04	Lagoon	Snowflake eel liver	18	-0.28			<b>No.</b>	<b>11</b>	<b>11</b>
06/10/04	Lagoon	Snowflake eel muscle	4.0	0.77					
06/10/04	Lagoon	Snowflake eel liver	14	0.40					
06/10/04	Lagoon	Snowflake eel liver	24	-0.19					
06/10/04	Ocean	Snowflake eel liver	4.0	-0.34					
06/10/04	Ocean	Snowflake eel muscle	10	3.2					
06/10/04	Ocean	Snowflake eel muscle	7.9	2.2					
		<b>Mean</b>	<b>9.8</b>	<b>1.5</b>					
		<b>Stdev</b>	<b>5.3</b>	<b>2.7</b>					
		<b>Median</b>	<b>10</b>	<b>0.33</b>					
		<b>No.</b>	<b>21</b>	<b>21</b>					

Results of U and Be analysis for samples of marine sediment (beach sand) are listed in Table 9. Mean concentration at Illeginni Island for U is  $1.6 \pm 0.41 \mu\text{g g}^{-1}$  which is the same as the concentration observed throughout the Marshall Islands. Mean U concentration for Boggerik Island is  $0.63 \pm 0.15 \mu\text{g g}^{-1}$ . This low concentration of U in beach-sand samples collected at Boggerik is due to the origin of the samples. They were distinctly different in color and size than coral sands from Illeginni because they contained a high

percentage of mollusc shells rather than sand of coral origin. More detail is given in the Discussion section.

Mean concentration of Be at Illeginni is  $0.027 \pm 0.11 \mu\text{g g}^{-1}$  while it is  $-0.00056 \mu\text{g g}^{-1} \pm 0.0042 \mu\text{g g}^{-1}$ , (essentially undetectable at the detection limit) at Boggerik. Mean concentration of  $27 \text{ ng g}^{-1}$  (27 parts per billion) for Be in marine sediment is slightly higher at Illeginni where Be was detected at very low concentrations in only 6 out of 21 samples while being undetectable in all Boggerik Island samples. They test significantly different at  $p = 0.018$ .

Table 9. Be and U in marine sediment (beach sand) at Illeginni and Boggerik Islands

Date	Side	Sample no.	U $\mu\text{g g}^{-1}$	Be $\mu\text{g g}^{-1}$	Date	Side	U $\mu\text{g g}^{-1}$	Be $\mu\text{g g}^{-1}$
Illeginni Island				Boggerik Island, Control Site				
06/09/04	Ocean	1	1.7	-0.0086	06/6,7/04	Ocean	0.48	-0.0043
06/09/04	Ocean	2	1.3	-0.0075	06/6,7/04	Ocean	0.57	-0.0060
06/09/04	Ocean	3	1.1	-0.0073	06/6,7/04	Ocean	0.38	-0.0073
06/09/04	Ocean	4	1.4	-0.0070	06/6,7/04	Ocean	0.61	-0.0057
06/09/04	Ocean	5	1.3	-0.0089	06/6,7/04	Ocean	0.71	-0.0070
06/09/04	Ocean	6	1.4	-0.0086	06/6,7/04	Ocean	0.40	-0.0070
06/09/04	Ocean	7	1.5	0.0044	06/6,7/04	Lagoon	0.66	-0.0072
07/18/04	Ocean	8	3.1	0.48	06/6,7/04	Lagoon	0.76	-0.0070
07/18/04	Ocean	A	1.6	-0.0044	06/6,7/04	Lagoon	0.60	-0.0066
07/18/04	Ocean	B	1.5	0.0030	06/6,7/04	Lagoon	0.75	-0.0071
07/18/04	Ocean	C	1.6	0.0073	06/6,7/04	Lagoon	0.91	-0.0074
07/18/04	Ocean	D	1.6	0.0072	06/6,7/04	Lagoon	0.68	-0.0090
07/18/04	Ocean	E	1.4	0.017		<b>Mean</b>	<b>0.63</b>	<b>-0.0056</b>
06/09/04	Ocean	F	2.0	0.14		<b>Stdev</b>	<b>0.15</b>	<b>0.0042</b>
06/09/04	Lagoon	1	1.7	-0.0083		<b>Median</b>	<b>0.64</b>	<b>-0.0070</b>
06/09/04	Lagoon	2	1.4	0		<b>No.</b>	<b>12</b>	<b>12</b>
06/09/04	Lagoon	3	1.4	-0.0043				
06/09/04	Lagoon	4	1.5	-0.0042				
06/09/04	Lagoon	5	1.3	-0.0056				
06/09/04	Lagoon	6	1.4	-0.0030				
06/09/04	Lagoon	7	1.4	-0.0038				
		<b>Mean</b>	<b>1.6</b>	<b>0.027</b>				
		<b>Stdev</b>	<b>0.41</b>	<b>0.11</b>				
		<b>Median</b>	<b>1.4</b>	<b>-0.0042</b>				
		<b>No.</b>	<b>21</b>	<b>21</b>				

Concentration of Fe and Ca in marine sediments is listed in Table 10. The mean concentration of Fe in marine sediment from Boggerik Island is  $0.0031 \pm 0.00075 \text{ mg g}^{-1}$  and from Illeginni Island  $1.2 \pm 1.6 \text{ mg g}^{-1}$ . Iron was found in sediment samples from both the lagoon and ocean sides of Illeginni Island and

the concentrations are greater by about a factor of 400 than those at Boggerik Island. Calcium mean concentration in all sediment samples was about  $340 \text{ mg g}^{-1}$ , or 34%, indicating  $\text{CaCO}_3$  sediment of marine origin.

Table 10. Fe and Ca concentration in marine sediment at Illeginni and Boggerik Islands

Date	Side	Sample no.	Fe $\text{mg g}^{-1}$	Ca $\text{mg g}^{-1}$	Date	Side	Fe $\text{mg g}^{-1}$	Ca $\text{mg g}^{-1}$
Illeginni Island					Boggerik Island, Control Site			
Beach sand					Beach Sand			
06/09/04	Ocean	1	0.016	338	06/6,7/04	Ocean	0.0030	333
06/09/04	Ocean	2	0.0083	334	06/6,7/04	Ocean	0.0025	333
06/09/04	Ocean	3	0.009	325	06/6,7/04	Ocean	0.0035	328
06/09/04	Ocean	4	0.067	323	06/6,7/04	Ocean	0.0029	336
06/09/04	Ocean	5	0.15	358	06/6,7/04	Ocean	0.0022	362
06/09/04	Ocean	6	0.054	373	06/6,7/04	Ocean	0.0042	351
06/09/04	Ocean	7	0.72	336	06/6,7/04	Lagoon	0.0036	318
06/09/04	Ocean	8	4.6	331	06/6,7/04	Lagoon	0.0027	360
07/18/04	Ocean	A	0.44	357	06/6,7/04	Lagoon	0.0023	317
07/18/04	Ocean	B	1.2	332	06/6,7/04	Lagoon	0.0029	321
07/18/04	Ocean	C	0.17	327	06/6,7/04	Lagoon	0.0031	331
07/18/04	Ocean	D	0.38	331	06/6,7/04	Lagoon	0.0047	336
07/18/04	Ocean	E	0.70	353		<b>Mean</b>	<b>0.0031</b>	<b>335</b>
07/18/04	Ocean	F	3.0	363		<b>Stdev</b>	<b>0.00075</b>	<b>15</b>
06/09/04	Lagoon	1	0.73	315		<b>Median</b>	<b>0.0030</b>	<b>333</b>
06/09/04	Lagoon	2	5.9	338		<b>No.</b>	<b>12</b>	<b>12</b>
06/09/04	Lagoon	3	1.6	332				
06/09/04	Lagoon	4	2.3	331				
06/09/04	Lagoon	5	0.58	369				
06/09/04	Lagoon	6	0.87	344				
06/09/04	Lagoon	7	0.37	382				
		<b>Mean</b>	<b>1.2</b>	<b>340</b>				
		<b>Stdev</b>	<b>1.6</b>	<b>18</b>				
		<b>Median</b>	<b>0.57</b>	<b>336</b>				
		<b>No.</b>	<b>21</b>	<b>21</b>				

Table 11 contains results of Fe analysis of marine fauna liver and muscle at Illeginni and Boggerik Islands. Mean concentration of Fe in liver samples is  $3.4 \pm 3.5 \text{ mg g}^{-1}$  for Illeginni and  $2.1 \pm 2.4 \text{ mg g}^{-1}$ . There is no statistically significant difference ( $p \geq 0.10$ ) between the two sites. The concentration in muscle tissue is a different story. For Illeginni and Boggerik Islands the mean concentration of Fe is  $0.040 \pm 0.052 \text{ mg g}^{-1}$  and  $0.011 \pm 0.0045 \text{ mg g}^{-1}$ , respectively. The means are statistically different at  $p = 0.001$ .

There are two very high concentrations of Fe in muscle tissue of rock crabs at Illeginni (0.14 and 0.16, bold in the table). These are the result of crabs living and crawling around in reef areas that have legacy scrap iron distributed over extensive areas. These two unusually high concentrations could perhaps drive the statistical difference between Illeginni and Boggerik. This possibility is tested in the data in Table 12

Table 11. Fe concentration in muscle and liver of marine fauna

Date	Side	Species	Fe mg g <sup>-1</sup>	Date	Side	Species	Fe mg g <sup>-1</sup>	Fe µg g <sup>-1</sup>
Illeginni Island liver data				Illeginni Island muscle data				
06/08/04	Ocean	Snowflake eel liver	1.6	06/08/04	Ocean	Snowflake eel muscle	0.023	23
06/09/04	Lagoon	Snowflake eel liver	2.0	06/09/04	Lagoon	Rock crab	0.016	16
06/09/04	Ocean	Black-tip shark liver	1.3	06/09/04	Lagoon	Snowflake eel muscle	0.022	22
06/09/04	Ocean	Snowflake eel liver	2.8	06/09/04	Ocean	Rock crab	0.14	140
06/09/04	Ocean	White-tip shark liver	0.35	06/09/04	Ocean	Rock crab	0.16	160
06/10/04	Lagoon	Snowflake eel liver	9.0	06/09/04	Ocean	Snowflake eel muscle	0.017	17
06/10/04	Lagoon	Snowflake eel liver	9.9	06/10/04	Lagoon	Farmer fish	0.024	24
06/10/04	Lagoon	Snowflake eel liver	2.6	06/10/04	Lagoon	Rock crab	0.012	12
06/10/04	Ocean	Snowflake eel liver	1.1	06/10/04	Lagoon	Snowflake eel muscle	0.012	12
		Mean	3.4	06/10/04	Lagoon	Snowflake eel muscle	0.019	19
		Stdev 1	3.5	06/10/04	Lagoon	Snowflake eel muscle	0.023	23
		Median	2.0	06/10/04	Lagoon	Snowflake eel muscle	0.016	16
		No.	9			Mean	0.040	40
						Stdev.	0.052	52
						Median	0.021	21
						No.	12	12
Boggerik Island, control site liver data				Boggerik Island, control site muscle data				
06/6,7/04	Boggerik	Moray eel liver	4.8	06/6,7/04	Boggerik	Moray eel muscle	0.0037	3.7
06/6,7/04	Boggerik	Snowflake eels liver	1.1	06/6,7/04	Boggerik	Snowflake eels muscle	0.015	15
06/6,7/04	Boggerik	Snowflake eels liver	0.41	06/6,7/04	Boggerik	Snowflake eels muscle	0.009	9
		Mean	2.1	06/6,7/04	Boggerik	Rock crab	0.010	10
		Stdev.	2.4	06/6,7/04	Boggerik	Rock crab	0.013	13
		Median	1.1	06/6,7/04	Boggerik	Rock crab	0.0071	7.1
		No.	3	06/6,7/04	Boggerik	Farmer fish	0.011	11
				06/6,7/04	Namur	Farmer fish	0.018	18
						Mean	0.011	11
						Stdev.	0.0045	4.5
						Median	0.011	11
						No.	8	8

Table 12 contains results of Fe analysis of marine fauna muscle at Illeginni and Boggerik Islands (control site) with the two very high values for rock crabs deleted from the Illeginni data set. In this case, the

mean Fe concentration in muscle samples from Illeginni and Boggerik is  $0.018 \pm 0.0045 \text{ mg g}^{-1}$  and  $0.011 \pm 0.0045 \text{ mg g}^{-1}$ , respectively. The results still test significantly different ( $p = 0.001$ ).

Table 12. Concentration of Fe in muscle of marine fauna (rock crabs deleted)

Date	Side	Species	Fe $\text{mg g}^{-1}$	Fe $\mu\text{g g}^{-1}$
Illeginni Island muscle data Side				
06/08/04	Ocean	Snowflake eel muscle	0.023	23
06/09/04	Lagoon	Rock crab	0.016	16
06/09/04	Lagoon	Snowflake eel muscle	0.022	22
06/09/04	Ocean	Snowflake eel muscle	0.017	17
06/10/04	Lagoon	Farmer fish	0.024	24
06/10/04	Lagoon	Rock crab	0.012	12
06/10/04	Lagoon	Snowflake eel muscle	0.012	12
06/10/04	Lagoon	Snowflake eel muscle	0.019	19
06/10/04	Lagoon	Snowflake eel muscle	0.023	23
06/10/04	Ocean	Snowflake eel muscle	0.016	16
		<b>Mean</b>	<b>0.018</b>	<b>18</b>
		<b>Stdev</b>	<b>0.0045</b>	<b>4.5</b>
		<b>Median</b>	<b>0.018</b>	<b>18</b>
		<b>No.</b>	<b>10</b>	<b>10</b>
Boggerik Island, muscle data				
06/6,7/04	Boggerik	Moray eel muscle	0.0037	3.7
06/6,7/04	Boggerik	Snowflake eel muscle	0.015	15
06/6,7/04	Boggerik	Snowflake eel muscle	0.009	9
06/6,7/04	Boggerik	Rock crab	0.010	10
06/6,7/04	Boggerik	Rock crab	0.013	13
06/6,7/04	Boggerik	Rock crab	0.0071	7.1
06/6,7/04	Boggerik	Farmer fish	0.011	11
06/6,7/04	Roi Namur	Farmer fish	0.018	18
		<b>Mean</b>	<b>0.011</b>	<b>11</b>
		<b>Stdev.</b>	<b>0.0045</b>	<b>4.5</b>
		<b>Median</b>	<b>0.011</b>	<b>11</b>
		<b>No.</b>	<b>8</b>	<b>8</b>

## Discussion

The fact that there is no increase in concentration of either U or Be in marine fauna is not unexpected because both of these elements are insoluble in the chemical form in which they exist in the re-entry vehicle. Even if they were soluble the mixing with the ocean as a result of ocean current flow (even as low as a centimeter per second) would lead to concentrations orders of magnitude below the natural concentration of these elements in sea water (McCright, 1996). Consequently, one would not expect to find any increase in U and Be concentration in reef or pelagic fish, eels, turtles, filter feeders or other marine fauna as was indeed the case.

Concentration of U in beach sand collected from Illeginni Island is the same as that found in soil and beach sand throughout the Marshall Islands. Thus, there is no indication of increased U in beach sand at Illeginni Island. Uranium concentration in beach sand from Boggerik Island (control site) was about half (mean =  $0.63 \mu\text{g g}^{-1}$ ) of that in the Kwajalein beach sand (mean =  $1.6 \mu\text{g g}^{-1}$ ). The lower concentration of U at Boggerik results from a difference in origin of its sediment (sands) from those of Illeginni. Beach sand samples from Illeginni are typical of sand of coral origin. They have fine texture, are relatively uniform in size, and are light tan to white in color. The Boggerik sand samples, on the other hand, have a coarse texture, a large range of particle size, contain lots of mollusc shells, and are pink in color. It is well known by marine geochemists that mollusc shells are low in U concentration and rarely exceed  $0.5 \mu\text{g g}^{-1}$  (Veeh and Burnett, 1982) in the first 20 to 50 y after deposition. The lower concentration observed in the Boggerik beach sand is a direct result of the fact they consist mostly of mollusc shells rather than sands of coral origin. There are many instances in the Marshall Islands where ocean circulation and deposition patterns are such that there is a high deposition of mollusc shells on beaches relative to coral sand. In these cases it would be expected to find a lower concentration of U in the sample relative to a beach sand totally of coral origin.

Beach sands at Illeginni Island have a concentration of Fe that is about a factor of 400 greater than those from Boggerik Island. Concentration of Fe in sea water in areas where marine fauna were collected must also be much higher at Illeginni because mean Fe concentration in muscle tissue from marine fauna was higher by a factor of 4 at Illeginni relative to Boggerik

( $0.004 \text{ mg g}^{-1}$  versus  $0.011 \text{ mgmg}^{-1}$ , respectively). This difference was not observed in liver samples because liver tissue naturally concentrates large quantities of Fe that effectively mask the smaller increase due to increased concentrations of Fe in the water.

## Conclusions

Based on analysis of marine fauna described in this paper there is no difference between U and Be concentrations at Illeginni Island and the control site, Boggerik Island. Thus, there is no evidence that there has been any increase in U and Be concentrations in marine fauna as a result of the missile flight test program.

Concentration of U in coral beach sand at Illeginni is the same as soil and coral beach sand in the rest of the Marshall Islands and again reflects an insignificant impact from the flight test program.

Beach sand from Illeginni has a mean concentration of Be higher than that from Boggerik Island. Seven of 21 samples from Illeginni had detectable Be. Four samples had a concentration of Be ranging from 4 to  $7 \text{ ng g}^{-1}$  [4 to 7 parts per billion (ppb)], one was 17 ppb, one was 0.14 parts per million (ppm) and one was 0.48 ppm. These extremely low concentrations of an insoluble form of Be again indicate no impact on marine life or human health at Illeginni as a result of the missile flight test program.

Concentration of Fe in marine fauna muscle tissue is much higher at Illeginni Island than at Boggerik Island (control site) as a result of legacy iron piers, dump sites for iron metal along the island, and scrap iron randomly distributed along extensive portions of the reef line as part of programs conducted in the 1960's through 1980's that were not part of the recent flight test program.

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## Units used

1 part per thousand (ppt) =  $1 \text{ mg g}^{-1} = 0.001 \text{ g g}^{-1}$   
 1 part per million (ppm) =  $1 \mu\text{g g}^{-1} = 0.000001 \text{ g g}^{-1}$   
 1 part per billion (ppb) =  $1 \text{ ng g}^{-1} = 0.000000001 \text{ g g}^{-1}$



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## Appendix A – Analytical Quality Control Data

### 1. Blind Duplicates

#### Blind duplicate analyses of marine sediments and fauna

		Sediments	Sediments
		U	Be
<b>Illeginni Island</b>		$\mu\text{g g}^{-1}$	$\mu\text{g g}^{-1}$
	ocean 3	1.06	-0.0073
duplicate	ocean 3	1.05	-0.0045
	ocean 6	1.4	-0.0086
duplicate	ocean 6	1.4	-0.0057
	laggon 4	1.52	-0.0042
duplicate	laggon 4	1.51	-0.0028

#### Boggerik Island

	ocean 3	0.376	-0.0073
duplicate	ocean 3	0.373	-0.0059
	lagoon 1	0.655	-0.0083
duplicate	lagoon 1	0.657	-0.0058

		Fauna	Fauna
		U	Be
<b>Illeginni island</b>		$\mu\text{g g}^{-1}$	$\mu\text{g g}^{-1}$
	ocean	0.0042	0.0077
duplicate	ocean	0.0042	0.0073
	ocean	0.0054	0.0056
duplicate	ocean	0.0050	0.0053
	ocean	0.0097	0.0056
duplicate	ocean	0.0096	0.0062
<b>Boggerik Island</b>			
	lagoon	0.0060	0.0043
duplicate	lagoon	0.0059	0.0043
	ocean	0.0031	0.0048
duplicate	ocean	0.0033	0.0051

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