[redacted]

and subject to further refinement.

The Bikini urine was collected at Kili Island. Since this location was relatively unaffected by the testing program, contamination by dust was unlikely.

The photon-electron rejecting alpha liquid scintillation spectroscopy (PERALS) was developed by Jack McDowell at Oak Ridge National Laboratory (ORNL). We used this system at Brookhaven National Laboratory (BNL) to estimate Pu. The following is a brief outline of the procedure we used to analyze the Bikini urine samples obtained two years after the Bikinians evacuated Bikini Atoll. The samples were urine from males (see Table One) and were composited into a total of eleven liters. Ten liters were divided into ten one-liter samples and five of these were analyzed by the PERALS method at BNL. Another five were analyzed by the PERALS method and by elecro-deposition techniques at ORNL. One liter was analyzed for ^{90}Sr. The result was virtually the same for all samples which were measured for ^{239}Pu.

The BNL samples were usually analyzed in groups of four. Each group consisted of one chemical blank, one known standard sample and two urine samples. A chemical blank was used to measure background radioactivity in all of the chemicals added. A standard sample consisted of the chemical blank to which we added 20 to 50 fCi of ^{238}Pu. We used a standard sample to estimate recovery and to reassure ourselves that the full-energy peak of the standard spectra corresponded to channels assigned to ^{238}Pu.

Usually, 1000 ml of the urine sample was placed in a beaker and wet ashed by the addition of nitric acid. Evaporation occurred and a few drops of H_2O_2 were added by us until all the organics were driven off. At the end of the wet ash the solution was placed in 2N HNO_3 and treated with $FeSO_4$ and $NaNO_2$ in succession to convert all the Pu to the 4+ oxidation state. The solution was then contacted with Adogen - 364 to remove the Pu. Traces of U, Th and other interferring nuclides were removed with a few washings of the Adogen - 364 organic phase with 0.7N HNO_3. We anticipated only Pu in the sample to be counted. The estimated recovery of interferring elements was tabulated in Table Two.

The Pu was back extracted into an inorganic phase with a mixture of 0.5M $LiClO_4$ and 0.5 M $HClO_4$. The combined extract was carefully evaporated to remove all HNO_3. The resultant residue was dissolved in a mixture of H_2O, a solution of sodium peroxydisulfate and a pinch of silver perchlorate. The solution was then contacted with an extractive scintillator mixture containing Naphthalene, HDEHP and PBBO in Toluene. The scintillator, which held the Pu, was transferred to a glass ampule and bubbled with oxygen-free argon. The ampule was sealed while bubbling with argon to keep out air.

The sample was counted in our laboratory using one of four liquid scintillation counters built in 1983.

At the beginning and end of each scintillation count a 10,000 dpm ^{239}Pu standard was examined and the peak channels were noted. The urine samples were counted for 200,000 seconds. Thus far, we have been able to analyze 6 individual plus the composite Marshallese urine samples. Over the last year we tested hundreds of quality control or chemical recovery samples in order to perfect the technique.

Presently, we have only a few results for comparison to Bikini urine. All results are tabulated in Table Three. I will have four or five more comparison samples when I visit on August 16th. One Majuro adult male exhibited activity in the ^{210}Po region of the spectra. We are examining our ^{210}Po recovery which should have been negligible and I will report on this later. The level of ^{210}Po in an average cigarette smoker's urine is about 65 fCi l^{-1}. In addition, even nonsmokers urine on the shelf for a few years will contain about 1000 fCi l^{-1} of ^{210}Po due to the decay of ^{210}Pb.

An example of one of the Bikini spectral results is given as Figure One. I have included the comparison ^{238}Pu spectra and overlayed it on the ^{239}Pu spectra in this figure.

The PERALS system was recently subjected to a synthetic urine quality control test. We detected different levels of ^{238}Pu. The results were told to us by Al Robinson of your laboratory. He stated we were within a standard deviation of 5 to 10% for the high range samples and within a standard deviation of 20% for the 100 fCi range samples. The greater deviation for the 100 fCi range samples was due to the fact that we analyzed only one-tenth of the sample volume. This would appear to be excellent.

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model. Moss indicated the Langham model underestimated urinary excretion of Pu especially at longer times past intake.

I plan to present the calculations in detail on August 16th and review our PERALS results with you. I will re-examine Pu results obtained in previous years and examine two other intake scenarios, continuous declining intake and continuous increasing intake.

We feel the committed dose equivalent estimates should be judged with some caution because they are based on a few recent Pu results. We ourselves have questions regarding ^{210}Po activity found in one comparison sample. We recognize there are different opinions regarding systemic excretion of Pu, different opinions regarding committed versus annual dose equivalent and different views regarding the importance of total bone volume dose versus bone surface dose. I have plotted bone surface annual dose equivalent, as estimated using the four above models, on Figure Two. The assumption was constant continuous intake at a level in agreement with the 41 fCi l^{-1} excretion of Pu measured in Bikinian's urine.

I expect many questions to be generated and will supply more information at the meeting. Best regards.

Sincerely,

Edward T. Lessard

Edward T. Lessard
Program Manager
Marshall Islands Radiological
Safety Program

ETL/lg

cc: R. Ray
J. Baum

Table One

Bikini Urine Sample

ID#	Sample Date	Age	Years on Bikini	Sample Volume, ml
966	7-31-80	56	4	890
2060	8-1-80	50	8	660
2102	8-4-80	54	6	1070
6001	7-31-80	76	7	880
6068	7-30-80	56	6	890
6166	7-31-80	55	9	1040
6017	7-31-80	49	8	910
6033	7-30-80	27	6	1030
6086	7-30-80	44	8	900
6118	8-1-80	22	6	1070
6125	8-4-80	35	9	970
6128	8-4-80	31	9	980

Table Two

Estimated Recovery of Elements Using PERALS

Chemistry Methods

Element	% Recovery
Uranium	$<10^{-3}$
Thorium	$<10^{-3}$
Americium	$<10^{-3}$
Plutonium	88±6
Bismuth	$<10^{-3}$
Radium	$<10^{-3}$
Neptunium	88±6
Polonium	* Available August 16th
Lead	$<10^{-3}$

Table Three

Recent Results for Plutonium in Urine

Subject	Location	^{239}Pu (fCi)	^{210}Po (fCi)
Adult Male	New York	<10	<10
Adult Male	New York	<10	<10
Adult Male	Majuro	<10	120
Child	Rongelap	<10	<10
Child	Rongelap	<10	<10
Child	Bikini	14	<10
Child	Bikini	12	<10
Child	Rongelap	<10	<10
12 Adult Males	Bikini	41±9.5	<10

Table Four

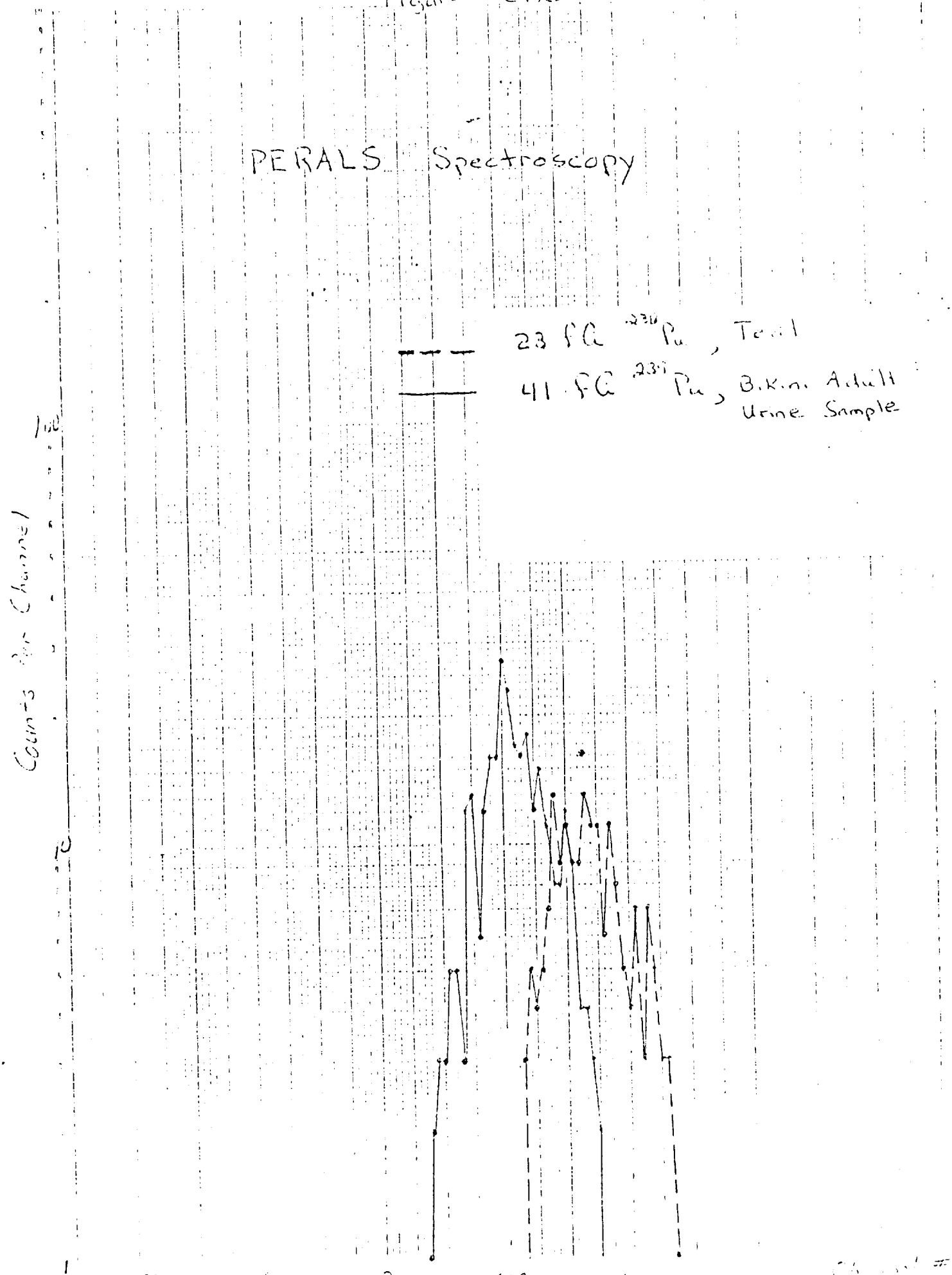
^{239}Pu Committed Dose Equivalent Estimate[1], rem

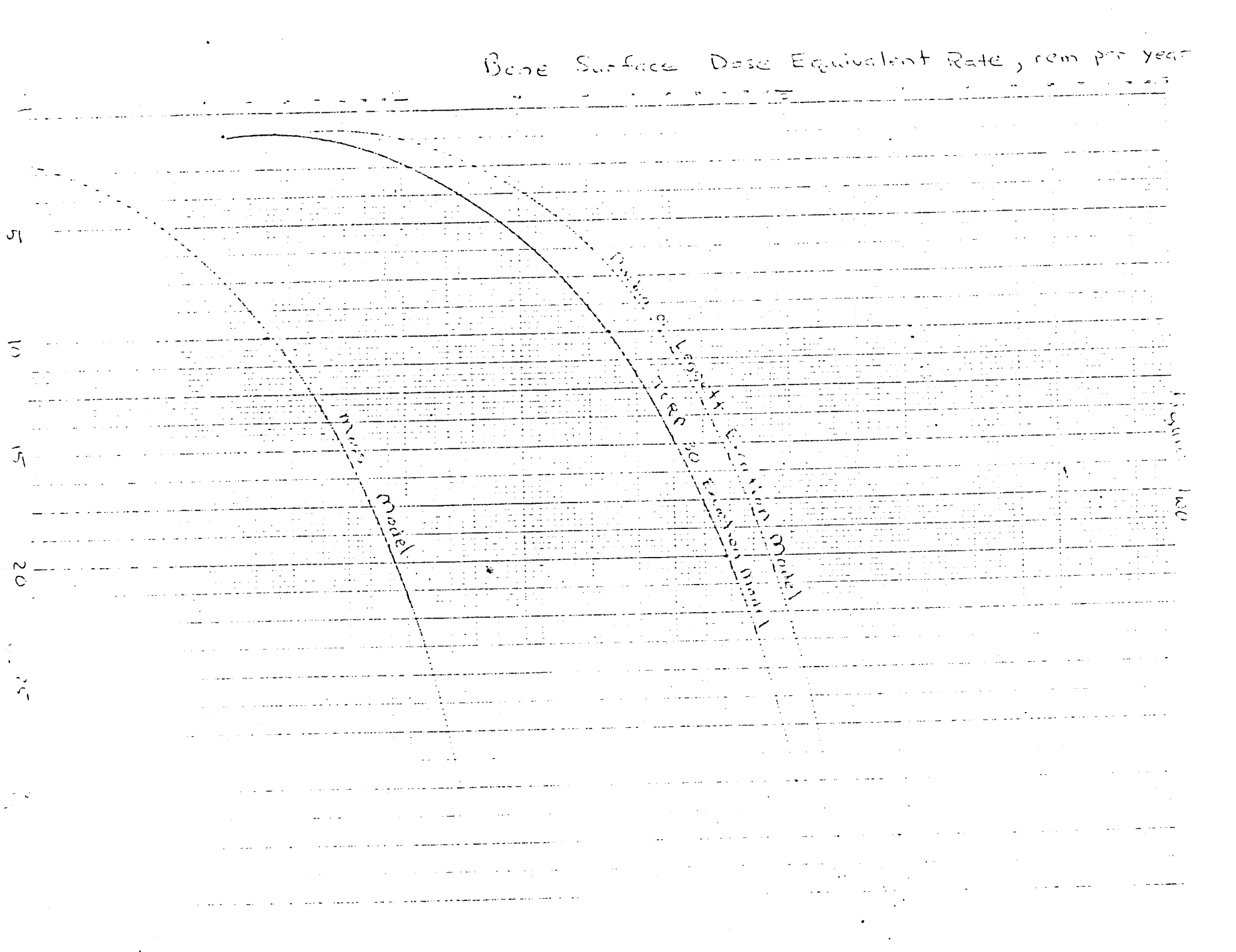
Excretion Model	Bone Surface	Liver	Red Marrow	Gonads	Effective	Comments
ICRP30	121	25.3	9.19	0.0161	6.66	Model Published
Leggett	169	35.3	12.8	0.0224	9.28	Model Published
Durbin	172	35.9	13.0	0.0229	9.46	Model Published
Moss	15.9	3.32	1.21	0.00211	0.874	Oral Report

[1]Former Bikini adults, constant continuous intake model and urine collected in August 1980.

Figure One

PERALS Spectroscopy




Bone Surface Dose Equivalent Rate, rem per year
5
10
15
20
Model
Model

REPOSITORY PNNL

COLLECTION Marshall Islands

BOX No. 5690

FOLDER Enewetak

DOCUMENT DOES NOT CONTAIN ECI

Reviewed by DJ Crosier Date 5/1/97