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BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: November 21, 1978

TO: Distribution

FROM: F. Cua F. T. Cua

SUBJECT: Determination of Pu Bone Burden Through Teeth Analysis

Question: Is it relevant and weighty enough for a 189 from DBER?

Outline of F. T. Cua's Research Proposal:

I. Objective 1: To develop an alternative technique to urine sampling for the calculation of low level plutonium bone burden--namely, the determination of plutonium activity concentration in teeth samples.

Rationale 1: Current state-of-the-art technique for the analysis of Pu in urine samples is such that the minimum detectable limit is in the region of the range of activity concentration of Pu in Marshall Islands urine samples. In addition, bioassay sampling suffers from several deficiencies:

a) assumption has to be made on the model of Pu distribution and excretion;
b) counting statistics, chemical recovery and uncertainty of a 24 hour urine sample results in errors that average 25%.

Objective 2: To develop a technique whereby the fraction of Pu inhaled versus Pu ingested can be obtained and thereby serve as a cross-check to the current method of vegetation-diet study and air-sampling resuspension program--namely, the autoradiography of teeth samples.

Rationale 2: Dose assessment through environmental monitoring in the Marshall Islands suffers from the following deficiencies:

a) use of models for Pu distribution and retention extrapolated from animal data to reference man;
b) assumption of constant continuous uptake which certainly is not the case;
c) difficulties in continuous air sampling due to lack of power in the islands.

Note: The proposed study will make use of animal data and will need certain unavoidable assumptions, but they are by far more controllable and less uncertain.

II. Materials: Teeth samples from animals under study at the following laboratories:

Definite:

a) Battelle Northwest Laboratory (c/o J. F. Park, FTS 8-444-3375)
Beagle Dogs - $^{238}PuO_2$, $^{239}PuO_2$, $^{239}Pu(NO_3)_4$ inhalation studies and injection studies (to be verified)

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b) Lovelace Biomedical and Environmental Research Laboratory (c/o J. A. Mewhinney, FTS 8-475-8770)
Beagle Dogs - ^{238}Pu, ^{239}Pu inhalation studies

Possibilities:

c) University of Utah (c/o Webster Jee, FTS 8-588-5500/581-6728)

d) Los Alamos Scientific Laboratory (c/o James McInroy, FTS 8-843-4709)

e) Argonne National Laboratory (c/o Marika Bhattacharyya, FTS-8-972-4160

Bone data (published or unpublished) that correspond to these teeth samples.

Teeth samples from the Marshall Islands. There are currently 62 samples which can be broken down as follows:

	Island		
a)	Bikini	1	
	Rongelap	26	
	Utirik	35	
	Sex/Age		
b)	Male adult >15 yrs. old		14
	Female adult >15 yrs. old		22
	Male children ≤15 yrs. old		5
	Female children ≤15 yrs. old		11
	Unidentifiable		5
	Same person extracted at different dates		5
	State		
c)	Permanent < Good / Bad (Decayed)		
	Deciduous < Good / Bad (Decayed)		
	Location		
d)	Lower Jaw Teeth		
	Upper Jaw Teeth		
	Kind of Teeth		
e)	Incisor		
	Canine		
	Premolar		
	Molar		
	Wisdom		
	Time and Date of Extraction		
d)	Jan. 1977	9	(All from Rongelap)
	June 1977	11	(All from Rongelap)
	Sept. 1977	1	(From Bikini)
	Jan. 1978	36	(32 from Utirik)
	Unidentifiable	5	

[redacted]

Additional Teeth Samples Necessary:

a) Control samples for different categories, (b)+(c)+(e)+(f)

b) Pending the results from the currently available samples, additional samples for activity concentration determination and autoradiography.

It will be obtained based on a flexible but carefully thought out experimental design.

III. Methods: Activity Concentration

Each Marshall Island sample selected would be analyzed for ^{238}Pu, $^{239,240}Pu$ via the ion-exchange fixation-elution-electrodeposition technique and alpha counting, for ^{90}Sr via the HEDHP extraction of ^{90}Y—separation as the oxalate and beta counting, and for ^{241}Am (if detectable) via the gamma counting of the eluent. Only the plutonium data will be used for F. T. Cua's Ph.D. research. Each animal tooth sample selected will be analyzed for ^{238}Pu or $^{239,240}Pu$. A quality assurance program will be set up between BNL and the laboratory from which the samples are obtained since the animal bone data are generated at the other laboratories.

All equipment is currently available at the Safety and Environmental Protection Division.

Autoradiography

Numerous autoradiographic techniques for preparing histologic section of undecalcified teeth are under evaluation. Each human and animal teeth samples selected will be subjected to the technique developed ultimately. Each section will be kept for future ^{90}Sr studies.

Equipment Necessary:

a) reagents used in preparing the samples
b) bioplastic or paraffin wax
c) embedding apparatus
d) mechanical grinder or microtome or microsawing apparatus
e) slides and emulsions and plates (α & β tracks)
f) photographic processing equipment
g) staining reagents
h) microscope (video camera???)

Question: Would it be difficult to arrange time for the use of the equipment currently available in SEPD and other departments such as Biology and Medical?

IV. Results:

1. ^{238}Pu or $^{239,240}Pu$ activity concentration in the teeth samples of different kinds of animals exposed through different routes of entry
2. Bone data
3. Teeth (1) to Bone (2) ratios for different animals for different routes of entry
4. ^{238}Pu and $^{239,240}Pu$ activity concentration of MI teeth samples as a function of age, sex, extraction date, kind of tooth, island, etc.

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5) Autoradiography of animal teeth samples selected according to certain criteria
6) Bone autoradiograph from other sources (published or unpublished)
7) Comparison of (5) and (6)
8) Autoradiography of M.I. teeth samples selected according to certain criteria
9) Comparison of (5) and (8)
10) Determination from (9) fraction ingested versus inhaled from different pattern (if any) in the distribution of Pu impacted on the teeth (from chewing) and circulated (from inhalation and from the transfer from gut to blood of ingested material)
11) Use of appropriately weighted ratios (3) to get bone burden by multiplying (3) by (4)

V. Conclusions:

a) Pu bone burden from teeth data
b) Comparison to bioassay results
c) Fraction of Pu ingested versus Pu inhaled
d) Comparison to vegetation--diet study and air sampling results
e) Comparison with other human teeth--human bone data (if available)

VI. Cost Itemization:

a) Salary of research collaborator
b) Travel expenses of research collaborator
-possible trip to Marshall Islands during teeth extraction or to other laboratories
c) Expenses to be incurred for the autoradiography
d) Use of equipment and personnel assistance from SEPD/Medical

Side Gains of F. T. Cua's Research Proposal:

a) The effect of the dental program on the retention of Sr and Pu in these teeth samples can be studied.
b) ^{90}Sr, ^{241}Am (if present) activity concentration and ^{90}Sr autoradiograph can be obtained from the M.I. samples analyzed for Pu.
c) ^{90}Sr teeth results can be compared to corresponding ^{90}Sr bioassay result (if urine samples are obtained from people whose teeth are extracted). Unlike ^{239}Pu, ^{90}Sr are easily detected in the M.I. urine samples (see report by F. T. Cua to Tommy McCraw).
d) If animal teeth sample and bone data can be obtained, the whole proposal can be applied to ^{90}Sr.

FTC/slg

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ADDENDUM

Quantitation and Clarification:

I.1) Minimum Detectable Level for Urine (LASL procedure):

10 fCi/liter at 95% confidence limits

Range of Pu in urine from Bikini Islands (EML procedure):

1 - 10 fCi/liter

*EML has ceased its Pu analysis operations since

Plutonium concentration in Rongelap/Utirik Islands: teeth samples (initial data):

Rongelap	185±167 fCi/gm
Utirik	207±198 fCi/gm

tooth weight between 1 and 2 gms

Definitely detectable using the BNL procedure.

Note: Bikini/Rongelap or Utirik ratio for Pu in urine is comparable (EML data) however, Bikini/Rongelap Pu in soil is at least 4X and Bikini/Utirik Pu in soil is 20X.

2) Assumptions that have to be made in solving for the Pu body burden from excretion data

a. ICRP 10A excretion model holds for Marshallese
b. Distribution of ^{239}Pu in man model (see attached) is applicable

3) The average error of 25% expressed in I. Rationale 1(b) needs to be corrected.

LASL accuracy for ^{239}Pu determination in urine is 100%

precision is ±10%

chemical recovery ranges 41 - 46%

24 hour urine sample for reference adult man - 1.4 l/day

Some samples are as low as 340 ml/day which amounts to 76% error. An average of 890 ml/day results in 36% error. The error then averages 46% and can be as high as 86% as long as the chemical recovery factor is known accurately and no other systemic error come to play. A control urine program to screen non-24 hour samples should decrease the error considerably.

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4) The assumption that has to be made for the teeth experiment is needed in the extrapolation from animal data, e.g., distribution of Pu in teeth samples of animals ÷ distribution of Pu in teeth samples of humans.

5) Calculations to show plausibility of autoradiography on the MI samples:

Tooth

specific gravity: (ICRP 23 Reference Man): 2.09 g/cm^3

weight: 1 gm

average activity concentration: 200 fCi/gm

$$200\ fCi/gm \times 2.09\ g/cm^3 \times 1\ gm = 418\ fCi/cm^3$$

let 1 slice be 10 μm

$$418\ fCi/cm^3 \times 10\ \mu m \times 10^{-4}\ cm/\mu m = 0.418\ fCi/cm^2$$

$$= 0.000928\ dpm/cm^2$$

Since $A = \lambda N$

$$A/\lambda = N$$

$$\frac{.000928\ dpm/cm^2}{\frac{0.693}{24,360 \times 5.25 \times 10^3\ min}} = N$$

$$1.71 \times 10^7\ atoms = N\ in\ 1\ cm^2$$

Note: 1 in a million Pu emits 1α/wk
One week: 17.1 α tracks/cm^2 (detectable)

Possible interference from other α emitters: Uranium to Plutonium ratio 1 to 100; Thorium to Plutonium 1 to 30, however, the Th α's are much lower in energy.

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TABLE I

A SUMMARY OF THE NUMBERS OF MARSHALLESE AND U. S. SERVICEMEN ON THE ATOLLS EAST OF BIKINI WHO WERE EXPOSED TO FALLOUT FROM THE TEST "BRAVO" ON MARCH 1, 1954 (2)

Number of People in Group	Time of Commencement of Fallout Hrs.	Time of Evacuation Hrs.	Est. Total r Air Dose Roentgens
1. Rongelap - 64	H +4-6	H +50 (16 people) H +51 (48 people)	175
2. Ailinginae*- 18	H +4-6	H +58	69
3. Rongerik - 28 U. S. Servicemen	H +6.8	H +28.5 (8 men) H +34 (20 men)	78
4. Utirik - 157	H +22	Started H +55 Completed H +78	14

* The people in the Ailinginae group were Rongelapese who happened to be fishing on Ailinginae at the time of exposure.

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