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Research Progress Report 1

The objective of my research is to determine the plutonium distribution in human teeth samples and compare it with that found in animal samples. Figure 1 and Table 1 are pictures and captions on alpha and beta autoradiographs of teeth samples from beagles injected with different radionuclides. Only if the distribution in human teeth samples correspond to that in animal samples can one use teeth to bone ratios from animal experiments to obtain bone burden from human teeth activity concentration for human beings.

The experimental procedure involves fission track autoradiography on thin sections of embedded teeth. Fission track autoradiography has to be used instead of alpha autoradiography because the level of activity concencentration in the teeth are of such low quantities, around 200 fCi/gm, that it will take several months to obtain an alpha autoradiograph. In view of the fact that Marshall Islands soil samples have shown trace quantities of uranium and thorium, it was deem necessary that Pu/U ratios be obtained to allay doubt on the origin of the fission tracks in the autoradiographs. Attempts will be made to obtain Pu/Th and Pu/Sr as well as Pu/gamma emitting radionuclides on each entire tooth.

Briefly, I have done a trial run for fission track autoradiography. Details of the procedure on the two teeth samples used are described in Table 2. Table 3 lists the modifications to be tried on the next batch of samples. Figure 2 shows the flow chart for obtaining the different ratios on the whole teeth samples that will be subjected to the radiochemistry procedures. At the moment, I have done qualitative identification of gamma emitting radionuclides and have found for possible radionuclides the daughter products of Th-232 and U-238.

That is all for now.

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Figure 1

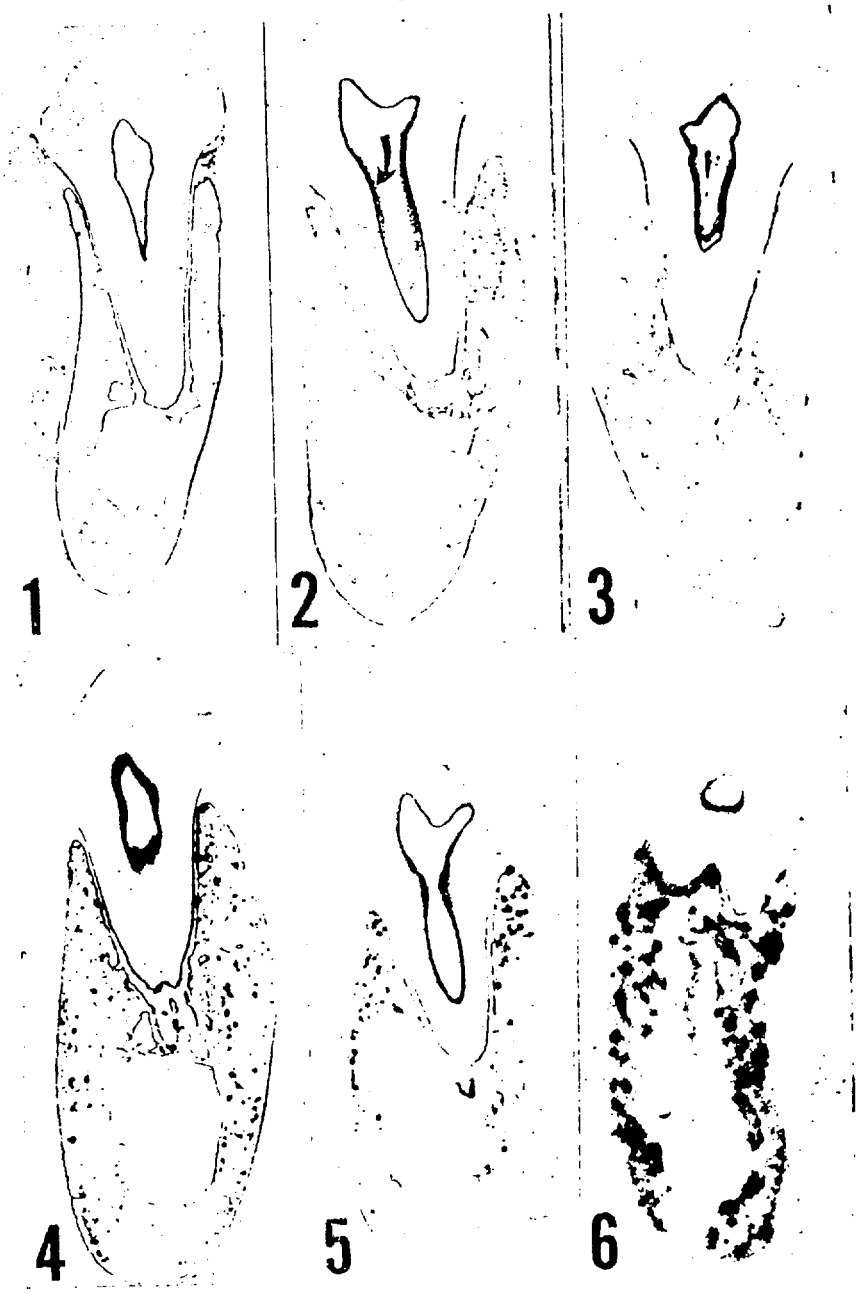

PLATE I

Radioisotopes in the Teeth of Dogs by WSS Jee and JS Arnold
Arch. Oral Biol. Vol. 2, pp. 215-238, 1960.

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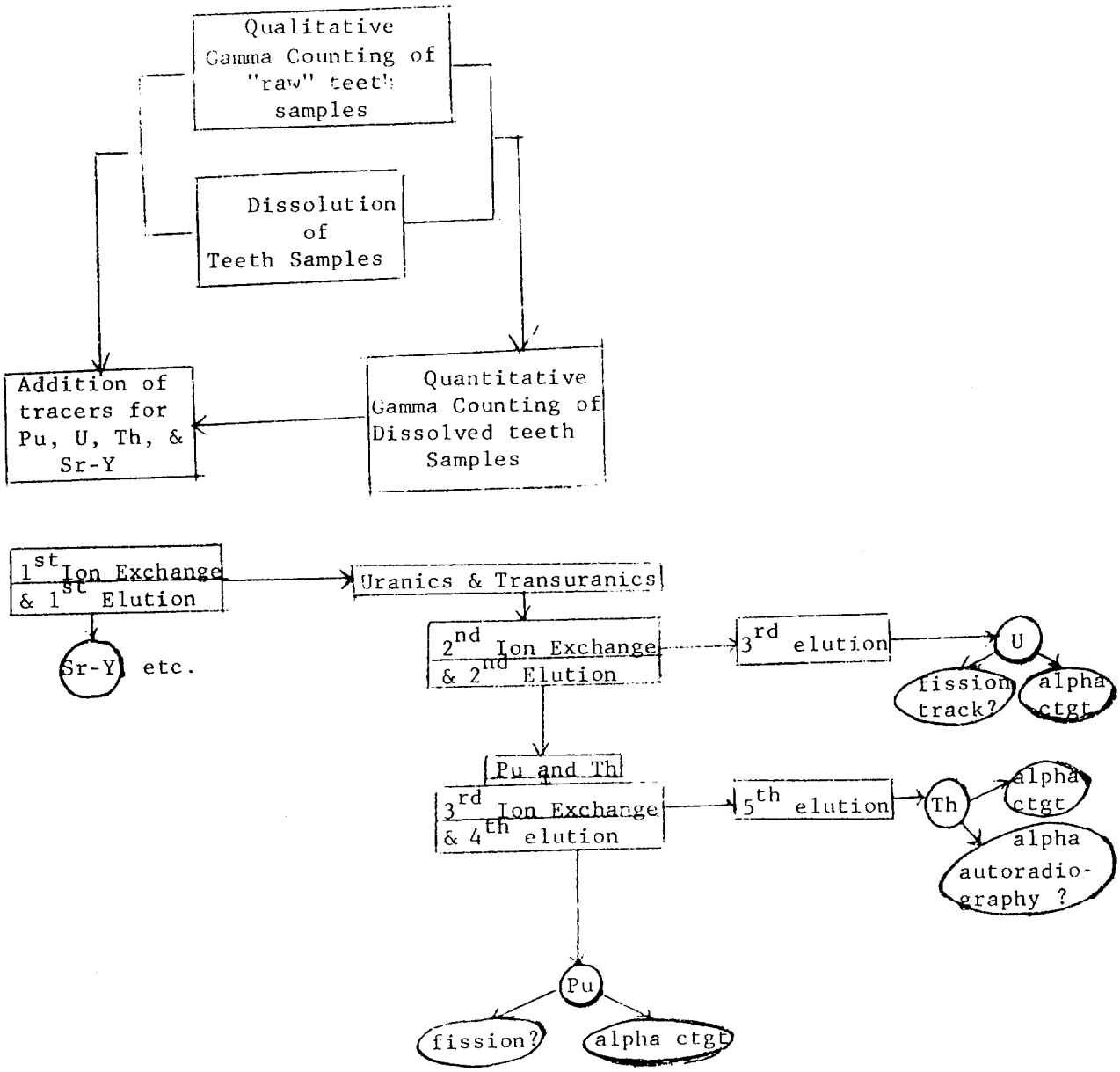
Figure 2



Results: Pu/gammas, Pu/Sr-Y, Pu/U, Pu/Th

if fission and alpha autoradiography have to be done, the kind of tracers which is used for obtaining recovery yields has to be well-chosen to insure non-addition of extraneous fission tracks or alpha tracks.

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Table 1

PLATE I

FIG. 1. Deposition pattern of plutonium in tooth and mandible 1 day after injection. ×3·6.

FIG. 2. Deposition pattern of plutonium in tooth and mandible 777 days after injection. Note non-uniform deposits of activity in post-injection dentine (arrow). ×3·6.

FIG. 3. Deposition pattern of plutonium in tooth and mandible 1576 days after injection. Note altered pattern. ×3·6.

Fig. 4. Deposition pattern of radium in tooth and mandible 12 days after injection. ×3·6.

FIG. 5. Deposition pattern of mesothorium in tooth and mandible 780 days after injection. ×3·6.

Fig. 6. Deposition pattern of strontium-90 in tooth and mandible 960 days after injection. ×3·6.

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Table 2

Fission Track Autoradiography Procedure used on two teeth samples from the Marshall Islands

Description of teeth: no ID
Tooth A was an intact incisor
Tooth B was a molar with very little of the roots attached

Step	Description	Comments
Dehydration	2 to 3 changes in absolute ethanol	To minimize outgassing when teeth are being irradiated and to permit the polymer to infiltrate the teeth
Infiltration	placed in unpolymerize methyl methacrylate + 1% (by weight) of catalyst, benzoyl peroxide for 2 days	full infiltration of teeth is more difficult than of bone
Embedding	replacement of above by methyl methacrylate + accelerator (25:1 by volume)	The accelerator causes the polymeri-zation of the methyl methacrylate through an exothermic reaction
Curing	allowed the sample to solidi-fy in a vacuum dessicator overnight	Better curing can be done by using a pressure chamber (e.g. 250 lb/in^2 and 50^oC)
Sectioning	use of carborundum saw to remove parts of glass mold	The cured material stuck to the glass mold. Such should not have occured if a mold releaser is used.
	Ten sections ranging from 356 μm to 1816 μm in thick-ness were obtained from the two teeth samples with a macrotome 2 precision saw(100 mm diameter diamond blade, water cooled)	Thinnest section obtained of encapsulated teeth had been 250 μm, according to the instruction manual
Preparation for irradiation	Each section was sandwiched between lexan foils(250 μm thick, 2.85 cm diameter) & 5 pairs from each teeth stacked one after the other and wrapped in pure Al foil	Wrapping in Al foil is supposed to provide good contact between teeth and Lexan
Irradiation	10^{13} nt/cm^2sec neutron flux for 2 hrs at the Medical Research Reactor which has a thermal to fast neutron ratio of about 33. Ambient temperature in the capsule is greater than 50^0C. The	A low n_f/n_{th} ratio is desired to prevent U-238 fast fission and $^{27}Al(n_f,\alpha)^{24}Na$ reaction Control teeth were inaccessible at the time. Control lexan was not included in the irra-diated batch this time(plain

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cont.

Step	Description	Comments
	neutrons were supposed to come from all directions	forgetfulness)
Etching	20 minutes etching at 5.94N KOH ± 0.495 at 60.5°C, 40 minutes etching at 5.72N KOH ± 0.305 at 60.3°C, 80 minutes etching at 5.14N ± 0.428 at 60.3°C	The foils were totally immersed in 3.5 l KOH after GM reading on the foils were less than 1 mR/hr
Rinse	15-20 minutes in circulating distilled water	
Drying	86°C oven till dry	
Tracks viewing	a) tracks from 20 min. etching are needle like, for 40 and 80 min. are small and big oval-shaped holes respectively	a) length and width of the track and the track density have not been measured
	b) tracks that have to be U or Pu fission tracks abound on foils in direct contact with the Al foil and on the edges of the foil.	b) though the Al foil has a high chemical purity, U contamination can easily come about through contact with the atmosphere, etc.
	c) Images of the tooth and the embedding material is present	c) due to inadequate contact, images of the dentine, pulp, enamel are not distinguishable
	d) Star aggregates are present though sparse and can be found mainly where the tooth is but is also visible where the tooth isn't	d) aggregates could be clusters of Pu in the teeth or resulting from background U-238 conversion to Pu-239
	e) Needle like tracks from 40 min and 80 min etchings seem to be buried in sections where the dentine of the tooth was	
	f) photomicrographs and polaroid shots of tracks on the viewing screen were taken	

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Table 3

Modification of the Procedure used for Fission Track Autoradiography of Marshall Islands Teeth Samples

Step	Modification
Dehydration	none
Infiltration	none
Embedding and Curing	instead of Solution B from Polyscience Inc. as accelerator, use 2,2' Azobisiso butyrontrile ALBN (0.5% by weight). The methyl methacrylate plus ALBN will be exposed to uv light,overnight-1 day. The new procedure should result in a harder polymer. To provide easy release of the specimen from the mold, a release agent such as silicon grease (20% by wt.) dissolved in methyl methacrylate can be used to coat the inside of the glass mold.
Sectioning	Sections as thin as 250 μm will be attempted.
Preparation for Irradiation	a) If Al foil were to be used, several pieces of lexan or polyethylene discs should be placed between the foil and the "teeth sandwich" b) A new set-up without the use of Al foil is being planned
Irradiation	Instead of the "rabbit" pneumatic tube irradiation facility, the "aluminum can" radial set-up will be used; the former has a n_{th}/n_{fast} ratio of 33 and temperature is greater than or equal to 50°C whereas the latter has a ratio of 300 and temp. is greater than or equal to 30°C. Irradiation time has to be increased considerably since the former has a neutron flux of 10^{13} nt/cm^{2}sec whereas the latter of $3x10^{12}$ nt/cm^{2}sec, smaller by a factor of 3. The irradiation time used previously is too short, as observed from the Pu tracks, and may have to be increased 2-3x. Thus the new irradiation time may be 8-10 hrs.
Etching	for long irradiation time, greater than or equal to 10 hrs at the $3x10^{12}$ nt/cm^{2}sec flux, Billie Oltman of ANL suggests lower temperature and longer etching time, 44°C, 90 minutes. If 60°C is to be retained, 35 minutes etching time was recommended. He also recommends partial immersion instead of total immersion- etch the side against the teeth specimen only. The set-up used at ANL is 6.25M KOH in a 1500 mL pyrex tray on top of a hot plate with temperature measured by a thermometer. This should improve signal to noise ratio and also make track counting less confusing.

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Step	Modification
Rinse	none
Drying	air dry or oven dry
Tracks viewing	with the use of a micrometer slide, the track length can be measured. Hopefully, the signal to noise ratio would be better and good contact between teeth and lexan obtained so that a good fission track autoradiograph detailing Pu distribution in teeth can be acquired.

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