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RECENT EXPERIENCE IN THE ANALYSIS OF POSTMORTEM SAMPLES FOR PLUTONIUM[a]

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ABSTRACT

Procedures are described for isotopic analysis of large autopsy tissue samples for $^{239,240}Pu$ and ^{238}Pu at Hanford. Internal tracers (^{242}Pu, ^{236}Pu) are presently used to measure radiochemical recovery to an accuracy of $\pm$ 5%. Sample sizes may be up to at least 400g for soft tissues and up to 50g for bone with recoveries of 70 $\pm$ 30%. Some critical points of the procedure, problems and their solutions are discussed.

INTRODUCTION

A program of analysis of tissue samples collected postmortem from former plant workers and the general population in the vicinity of the Hanford complex has been carried on since 1949. This program has emphasized measurement of plutonium in tissues although some samples have been examined for uranium and certain gamma emitters. In addition to the ongoing program, in recent years autopsy tissue samples from exposed workers have been obtained by the United States Transuranium Registry and have been analyzed in this laboratory.

Until mid-1974, radiochemical analyses were performed by the method of Schwendiman, et al. (1951) or modifications thereof. In that procedure total plutonium alpha activity was determined using lanthanum fluoride coprecipitation followed by organic solvent extraction separation, electrodeposition on SS discs, autoradiography and alpha track microscopy.

Since 1974 analyses have employed alpha energy analysis as the final measurement step, primarily as a result of the availability of relatively pure tracers and the yield uncertainty associated with the autoradiography procedure. In part, the presently described procedure grew out of the need for thinner, cleaner deposits on the electrodeposited discs required for resolution of the plutonium alpha peaks in the energy spectrum. Plutonium isotopes measured are $^{239,240}Pu$ and ^{238}Pu. Because ^{239}Pu and ^{240}Pu alpha particles have nearly identical energies, they are not resolved in the spectrum analysis and are referred to collectively as ^{239}Pu.

The separation techniques have been optimized for our purposes from published chemical techniques (Campbell, Moss, 1964; Magno, et al. 1969 and others). The electrodeposition procedure is patterned largely after Talvitie (1972). Complete details of the various techniques which have been combined in the total analytical procedure are presented in the Appendix.

DISCUSSION

A. Use of Tracers

Since it is possible to lose portions of a sample during any step of the analysis, it is desirable to introduce an internal tracer at the earliest possible step in the procedure. This technique assures that all losses of the isotope of interest will likewise be reflected in a proportional loss of tracer. As a consequence, ^{242}Pu tracer is added even before the sample is dried.

Under certain circumstances, addition of tracer to the entire sample before aliquoting can complicate the analysis. For example, if the amount of ^{239}Pu in the sample is much greater than expected, the ^{239}Pu peak tails into the ^{242}Pu region as shown in Figure 1 and interferes with the determination of the tracer yield. Since tailing of one peak into another in an alpha spectrum occurs from a peak of higher energy into a peak of lower energy, a similar tailing problem is encountered when ^{236}Pu is introduced as tracer. In this instance, the tail of the higher energy ^{236}Pu alpha peak interferes with the lower energy ^{238}Pu region as shown in Figure 2. Hence either ^{242}Pu or ^{236}Pu used as internal tracer presents similar difficulties albeit in different parts of the alpha energy spectrum. Because ^{238}Pu levels are generally expected to be quite low, ^{242}Pu has been chosen over ^{236}Pu as the principal tracer to be used initially for most samples. If the ^{239}Pu activity in the sample is large enough to adversely affect the ^{242}Pu spectrum, a second smaller aliquot of the sample is rerun using ^{236}Pu tracer to allow calculation of ^{239}Pu in the second aliquot. Using this measure of concentration of ^{239}Pu in the sample, the amount of ^{239}Pu present in the first aliquot can be calculated and a radiochemical recovery of ^{239}Pu can be determined. This yield in turn can be used to calculate ^{238}Pu in the first aliquot. Although this technique does not correct for losses during dry- and wet-ashing, in those instances when the ^{239}Pu activity is high enough to interfere with the ^{242}Pu tracer, it does allow calculation of an estimate for both ^{239}Pu and ^{238}Pu.

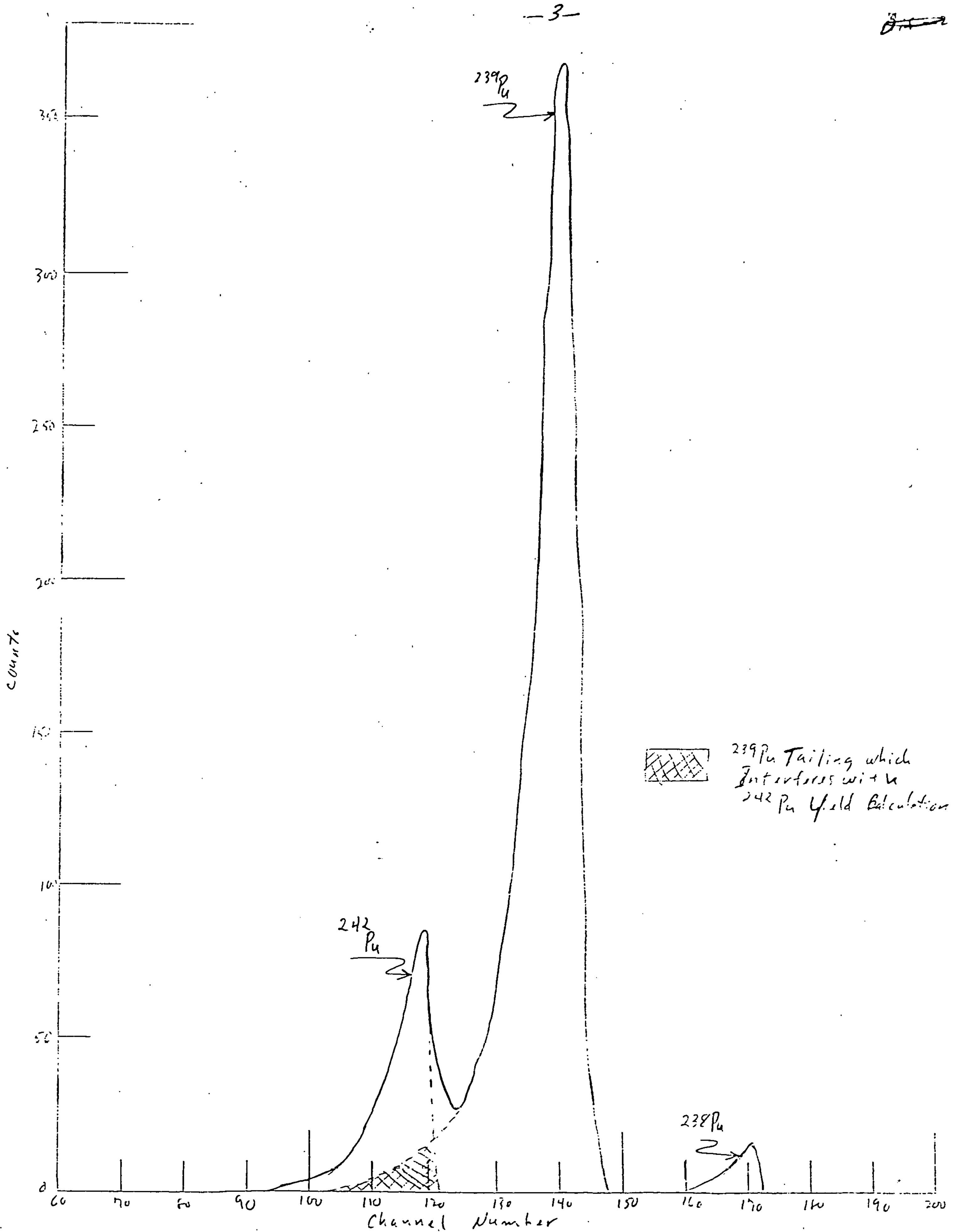

Figure 1. Alpha Energy Spectrum of Plutonium in Human Liver Containing ^{242}Pu Added as Tracer.

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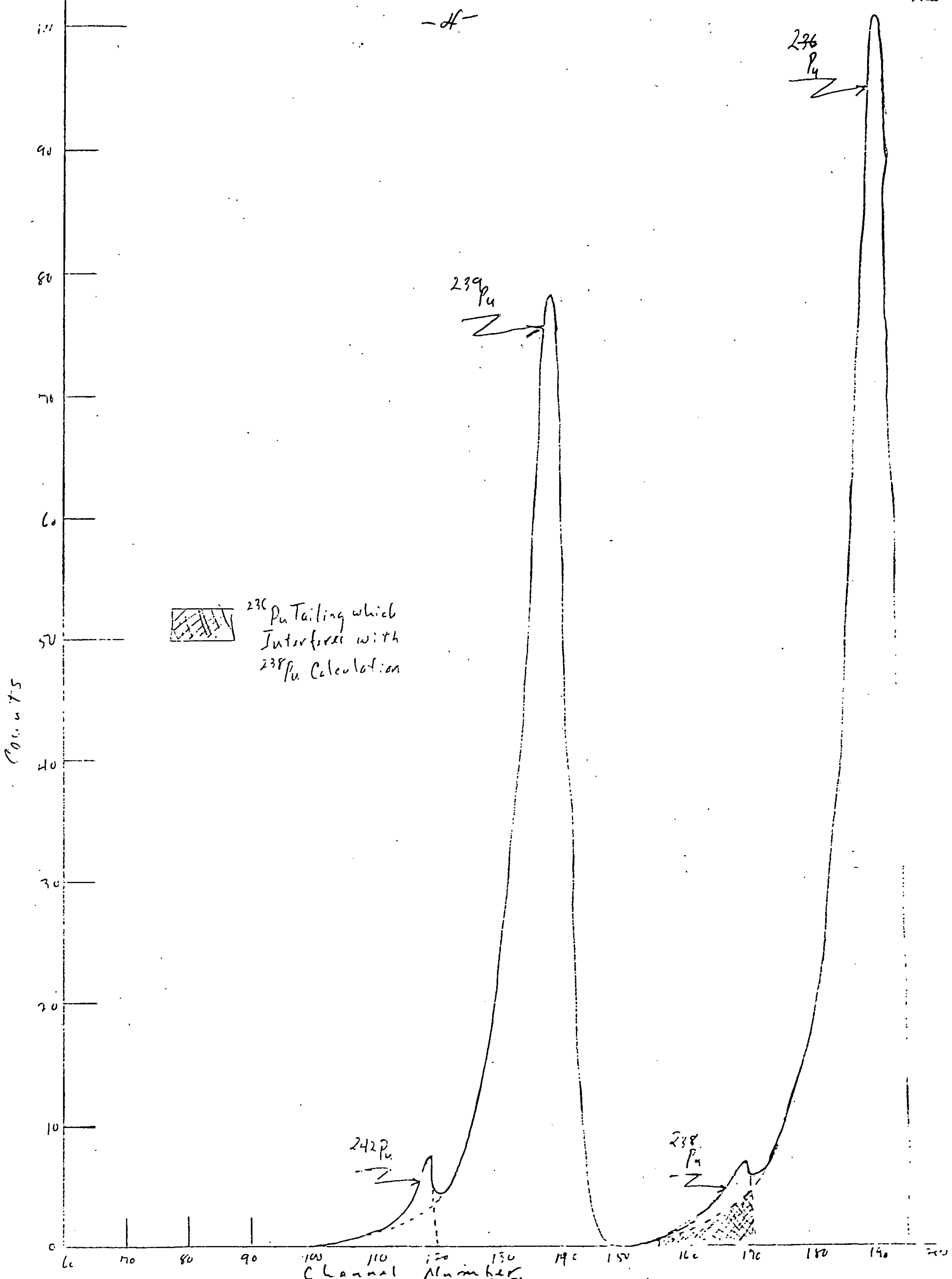

Figure 2. Alpha Energy Spectrum of Plutonium in Liver Containing ^{242}Pu and ^{236}Pu Added as Tracers

B. Ashing the Sample

Liver samples are particularly difficult to dry ash because at about 300°C the samples tend to swell and overflow the beaker. Whenever possible, beakers are selected so that the volumes of liver samples are restricted to no more than one-fifth the total beaker capacity to allow for this swelling. Other samples may fill the beaker to any desired level. As the temperature nears 300°C, liver samples require close attention. When slight puffing occurs, the temperature is held at that point for about 24 hours, which allows the slow escape of volatile compounds with a minimum of swelling. The temperature can then again be progressively raised as experience dictates.

All samples which may contain insoluble refractory PuO_2 particles are treated specially to insure complete dissolution. A mixture of strong HNO_3 with .01N HF has been used successfully as a solvent for refractory PuO_2. According to Cleveland (1967) the dilute HF has a catalytic effect on the dissolution rate of PuO_2.

One particularly difficult type of sample to ash without losses is a mixture of formalin with large amounts of fat. Several of the usual ashing techniques have been tried. Direct heating on a hot plate causes the formalin to superheat and bump with resultant losses from the sudden release of vapors. Drying under a heat lamp distills off low boiling fractions of the fat but eventually ceases before the sample is dry. Ashing in an oven by slowly elevating the temperature eventually ignites the gasses evolving from the liquid which burn rapidly in an uncontrolled manner, once again resulting in losses.

Samples of this nature have been successfully ashed in a well-ventilated hood or cool oven by inserting a braided-cheesecloth wick through the organic phase into the formalin. A heat lamp is used to evaporate the formalin drawn up by the wick. The wick is then ignited and the organic compounds are burned. A typical sample with a half-inch diameter wick will burn about 25 mls of fat per hour. The formalin evaporates at a somewhat slower rate.

C. Isolation of Plutonium

To measure plutonium at the lowest levels, it is desirable to use the largest sample aliquot possible while retaining an adequate tracer recovery. Experience before adoption of the present tracer procedure had shown that yield was inversely proportional to the weight of sample analyzed. This relationship was especially pronounced for bone but less so for liver, lung or other tissues. The new procedure allows a much larger bone sample to be analyzed with about the same radiochemical recovery because the dependence on sample weight is markedly less. For other types of samples, the dependence of yield upon sample weight is not clearly demonstrated. Aliquots are therefore taken which should allow tracer yields to be greater than 50%, and still leave half the sample for possible future measurements.

In the procedure, a number of factors have been found to significantly reduce the yield from the resin column separation. Once loading of the sample has begun, careful attention is required to keep the flow rate at 1 ml/min or slightly less. The flow is not interrupted until the plutonium is eluted, resulting in higher, more consistent yields. Residual chloride from the column, and residual fluoride in the sample can also cause lower yields. Testing to assure the complete absence of chloride in the wash solution, and adding boric acid to the samples to complex any residual fluoride from the earlier dissolution steps have made significant improvements in radiochemical yields.

D. Electrodeposition

Electrodeposition is carried out in a cell similar to that reported by Schwendiman, et al. (1951) but modified to hinder capture of gas bubbles at the surface of the disc. Such bubbles can isolate the disc from the electrolyte and interfere with the electrodeposition. The electrolyte used in the cell consists of 1.28*M* $(NH_4)_2SO_4$ - .007*M* HF.

Adjustment of pH in the electrolyte is critical to obtain clean, thin deposits with high recovery. If the pH is slightly higher than pH 2.3, a dark deposit builds up on the disc which degrades the resolution of the alpha spectrum. A slight shift to

the acidic side of pH 2.3 yields a clean, almost invisible deposit, but the yields tend to be low and erratic. The pH is best adjusted using NH_3 gas with thymol blue as indicator. The yellowish pink end-point is optimum. If this point is overshot into the green or blue color, lowering the pH with acid will often result in a low yield.

The addition of dilute HF to the electrolyte, although slightly reducing the yield, appears to improve the consistencies of recoveries. Discs are kept covered to limit exposure to air for at least 6-8 hours prior to counting to eliminate radon daughter contamination.

E. Alpha Energy Analysis

Each 256 channel quadrant of the 1024 multichannel analyzer is set to store electrical pulses from alpha particles detected with energies from 3.5 - 6.5 MeV. A second spectrometer uses a 4096 channel analyzer and each 1024 channel quadrant is set to detect alpha energies from 3 to 9 MeV. These ranges cover most nuclides of interest. Typically alpha resolution is about 60 keV FWHM (full width at half maximum) and is detected with an average efficiency of 36%. The average background in the 20 channels used from the 1024 channel analyzer to calculate ^{239}Pu is 0.0052 ± 0.0033 dpm, and in the ^{238}Pu channels is 0.0104 ± 0.0072 dpm, where the indicated errors are one standard deviation about the mean.

Four of the eight detectors are of the gold-surfaced variety which are extremely fragile to physical abrasion of the gold surface. Diodes are replaced when their surfaces have become contaminated because according to the manufacturer they cannot be cleaned. The other four detectors are aluminized so that the active surface can with care be cleaned of loose contamination. Although the resolution capabilities of the gold-surfaced diodes are superior to those of the aluminumized diodes, in practice, the electrodeposited source determines the overall resolution which does not normally approach the capabilities of the diode. Hence the possibility of cleaning the aluminized diode makes it a more economical choice without apparent loss of sensitivity or resolution. To minimize diode replacement due to contamination, samples are screened by ZnS scintillation counting, and only samples with the lowest activity are counted

with the gold-surfaced diodes.

A significant advantage of using alpha energy analysis over autoradiography, other than the ability to determine the amounts of specific radioisotopes, is the ability to calculate accurately the radiochemical recovery of each aliquot analyzed through the use of tracer isotopes of the same element. This is particularly useful in the analysis of large autopsy samples which have inherent problems of erratic and often low recoveries. When autoradiography is used, the correction of samples for radiochemical recovery is based on analyses of similar samples (usually bovine) to which known amounts of the desired radionuclide have been added. The average recovery of many such proxy samples is then used to correct the true sample for procedural losses. The uncertainty in yield factors derived in such a manner for large autopsy samples which are subject to erratic yields is considerable. Actually, the uncertainty in the yield directly controls the accuracy to which an analytical result may be reported, regardless of the level of activity in the sample. A typical plot of radiochemical recovery versus weight of bovine liver analyzed by this substitution method is shown in Figure 3. These data indicate that no result can be reported to an accuracy of better than $\pm$ 25% (1σ) because of the uncertainty in the yield. In contrast, a plot of similar data for human lung obtained by the modified procedure of this paper is shown in Figure 4. The error bars on selected data points indicate typical propagated errors about measurements of radiochemical yield. Contribution to the total error on the final plutonium results calculated from these recoveries is less than $\pm$ 10% (1σ).

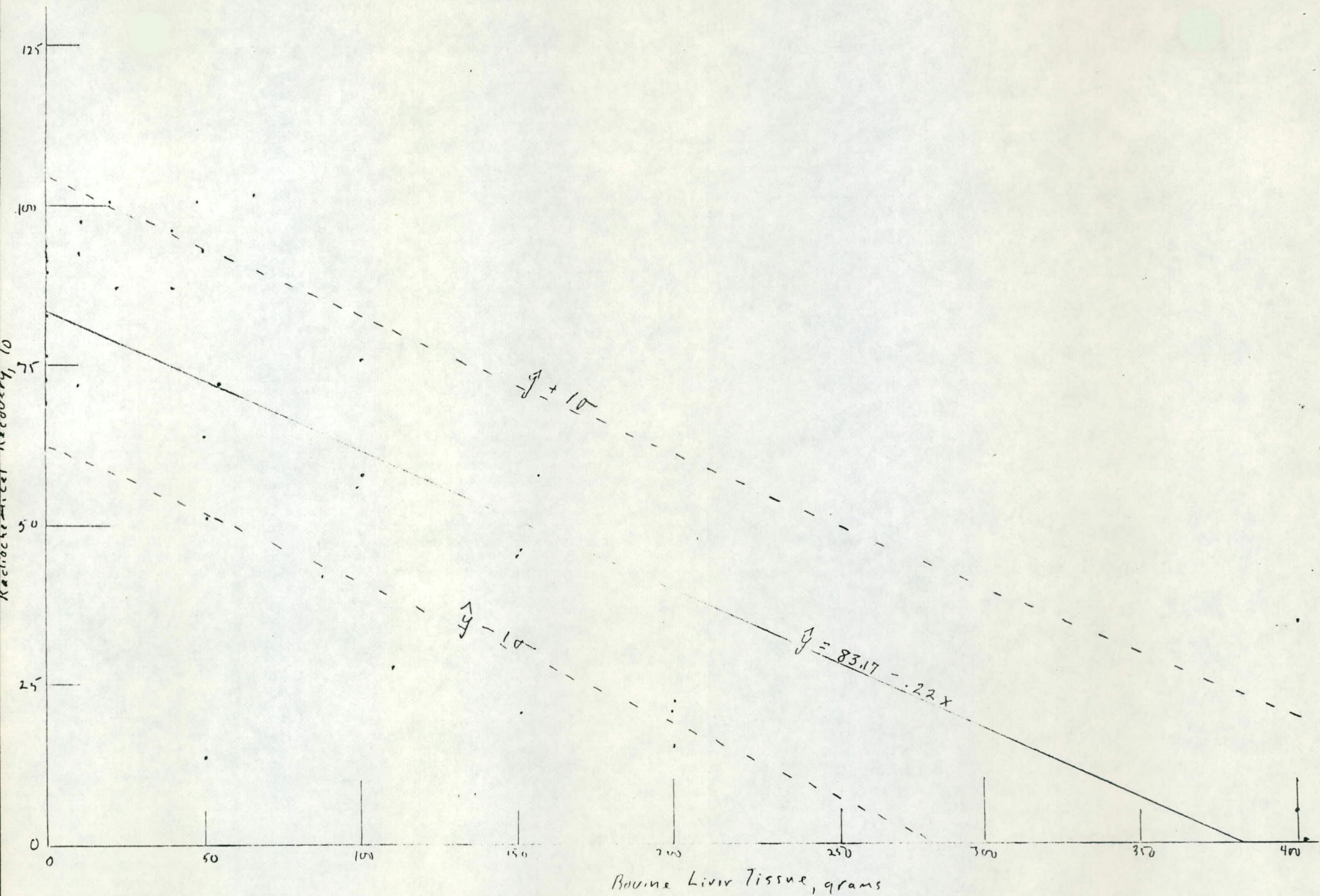

Figure 3. Radiochemical Recovery versus Weight of Bovine Liver Tissue Analyzed Showing Upper and Lower 1σ Limits from Total Plutonium Alpha Measurements

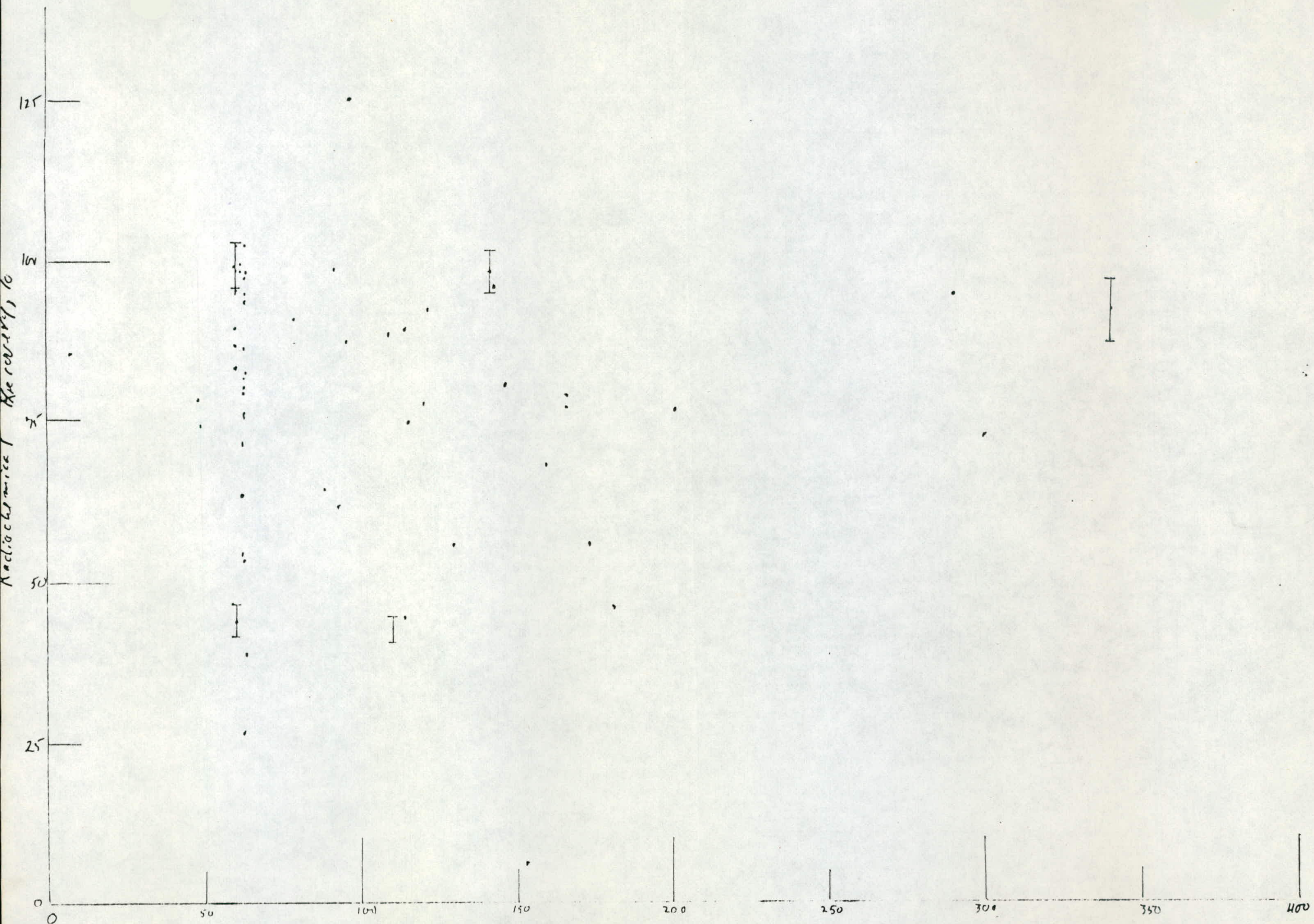

Figure 4. Radiochemical Recovery Versus Weight of Human Lung Tissue Analyzed by Alpha Energy Analysis. Error Bars Shown Are Upper and Lower 1σ Limits.

Autopsy tissue and bone samples have been analyzed for $^{239,240}Pu$ and ^{238}Pu at very low levels in our laboratory using adaptations of various published procedures. While radiochemical recoveries are sometimes erratic for all types of samples, the use of internal plutonium isotope tracers in each sample analyzed allows the radiochemical yield for that analysis to be determined very precisely and therefore confidence can be put in analytical results based on even low recoveries.

Since using either ^{242}Pu or ^{236}Pu exclusively as internal tracers in samples having unknown activity levels results in loss of data, both tracers are used. Plutonium-242 is used as the initial tracer added to the total sample and ^{236}Pu need only be used in those instances in which sufficient ^{239}Pu activity is present in the sample to interfere with the accurate determination of the ^{242}Pu recovery.

Sample sizes varying from 400 grams for soft tissues to about 50 grams for bone have been analyzed with tracer recoveries normally falling within the 70 $\pm$ 30% range. With 90% tracer recovery, detection levels of ^{239}Pu and ^{238}Pu are presently 0.02 and 0.03 dpm (9 and 14 fCi), respectively.

ACKNOWLEDGMENTS

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APPENDIX

PROCEDURE

I. Sample Ashing and Dissolution

Autopsy samples are received frozen or formalin-fixed and are stored frozen or refrigerated until analyzed.

A. Dry-Ashing - After thawing the sample, enough ^{242}Pu tracer is added to insure that about 2 dpm will be in the aliquot to be analyzed. The sample is then thoroughly charred either under an infrared heat lamp or in a stainless steel lined furnace at temperatures less than 150°C. The charred sample is dry ashed in the oven by slowly raising the temperature to 500°C over several days' time. The samples are kept at 500°C for at least 24 hours.

B. Wet Ashing - Dissolution of the sample ash begins by the addition of 16N HNO_3 and 30% H_2O_2. The mixture is heated on a hot plate to near dryness and 30% H_2O_2 is carefully added. Sequential additions of 16N HNO_3 and 30% H_2O_2 to the nearly dry sample are continued until the residue is white. If tars and oils resist this treatment, fuming HNO_3 is substituted for 16N HNO_3. If this treatment fails to solubilize the organic material, 20-30 ml of 16N HNO_3 are added to the sample, followed by 10 ml of 70% $HClO_4$. The mixture is boiled down and fumed until nearly dry, HNO_3 is added, and the sample is taken to near dryness. Additional portions of HNO_3 are added and boiled down until no fuming is evident and the sample is again nearly dry. This HNO_3-$HClO_4$ treatment may have to be repeated

for stubborn samples. Bone samples are set aside at this point while other types of tissue are treated further in preparation for anion exchange column separation. The residue is transferred to a teflon beaker with 8N HNO_3. Hydrofluoric acid is added, and the sample is taken to near dryness under an infrared lamp. The HNO_3 - HF treatment is repeated until complete sample dissolution is achieved. The solution is once more taken to dryness with HNO_3 without baking.

The solution is transferred back to the original glass beaker with 8N HNO_3 - .01N HF. This acid mixture is also added to bone samples and the samples are boiled to near dryness, and the acid treatment is repeated at least three times for samples which may contain refractory plutonium oxide particles. The residue is dissolved in 7.7N HNO_3 and all samples are analyzed by gamma ray spectrometry for major gamma emitters present using large NaI(Tl) well crystals described elsewhere (Wogman et al. 1967; Wogman, 1970). The solution is then diluted to a known volume in a volumetric flask with 7.7N HNO_3 for aliquoting and storage.

II. Isolation of Plutonium

A. Phosphate Coprecipitation

Aliquots are taken for analysis such that one-half of the total sample is analyzed up to a maximum equivalent of about 400g for soft tissues and up to about 50g for bone samples. Soft tissue aliquots are taken to dryness on a hot plate and then dissolved in 4N HNO_3. Forty milligrams of calcium carrier and 6 mg of iron carrier (Fe^{+3}) are added, followed

by 1 ml of 1M phosphoric acid. Ammonium hydroxide is added until a permanent precipitate of calcium ammonium phosphate forms, and then 5 ml in excess are added. The precipitate is centrifuged and 2 - 3 drops of NH_4OH are added to the clear supernate to test for complete precipitation. Two milligrams of Fe^{+3} carrier are added, the sample swirled, and the $Fe(Oh)_3$ precipitate is centrifuged on top of the phosphate precipitate. The supernate is discarded to waste. The precipitate is washed with 10 ml of distilled water, once again centrifuged, and the supernate discarded. The precipitate is dissolved in 150-200 ml of 7.7N HNO_3. Approximately 2 ml of saturated boric acid solution are added to all samples (including bone samples) which are then heated in boiling water for 15 minutes, cooled for 10 minutes and finally 2 ml of freshly prepared 3M $NaNO_2$ are added. The solution is allowed to stand overnight to insure that the plutonium adjusts to the +4 oxidation state.

B. Preparation of Anion Exchange Column

A distilled water slurry of BioRad AG-1-X2 (50-100 mesh) anion exchange resin (Cl^- form) is allowed to settle in a 7 mm diameter column to a depth of 10 cm on a glass wool support. The column has a 100 ml reservoir. A second glass wool plug is inserted at the top of the resin.

The resin is washed with the following solutions, allowing time for the column to drain to within several millimeters above the top glass wool plug between additions: 2 column volumes of 12N HCl, 2 column volumes of 6N HCl, 2 column volumes of distilled water, and then enough 7.7N HNO_3 to remove all chlorides. Turbidity of the eluate is tested with

$AgNO_3$ to detect residual chloride. The flow rate is adjusted to 1 ml/min during the final washing.

C. Column Separation

Tissue sample solutions are diluted with 7.7N HNO_3 to 200 ml and bone sample solutions are diluted to 500 ml. The sample solution is passed through the resin column at a flow rate of 1 ml/min followed by a 7.7N HNO_3 wash equal in volume to the sample load. The eluate is saved for future americium analysis. When the liquid is about 1/2 cm from the top glass plug, 20 ml of 10N HCl are added in three increments. When the 10N HCl is again within 1/2 cm of the glass plug, plutonium is eluted with 40 ml of freshly prepared 6N HCl - 0.024% HI solution. The eluate is collected in a 100 ml beaker. The elution is repeated with 10 ml HCl-HI solution. Ten ml of 16N HNO_3 are added to the combined eluates and the plutonium-bearing solution is dried under a heat lamp without baking.

The residue is dissolved in 100 ml warm 7.7N HNO_3 and then put through a second prepared column as above.

III. Electrodeposition

The residue from the second column is dissolved from the bottom of the 100 ml beaker with 250 µl 36N H_2SO_4. 1500 µl of distilled water and one drop of 0.02% thymol blue indicator are added. The pH is adjusted to the yellow endpoint with NH_3 gas. The solution is transferred to an electrodeposition cell described elsewhere (Schwendiman et al. 1951) with two 1000 µl rinses of .36N H_2SO_4 - .014N HF. The pH is adjusted to 2.3 with NH_3 and H_2SO_4-HF acid

mixture by visual comparison to a reagent blank electrodeposition cell which has been adjusted to pH 2.3 as measured by a pH meter. The two hour electrodeposition employs a constant current power supply providing 240 mA (0.6 A/cm^2) through a stationary platinum anode to the electrolyte. The deposit is plated on 1/2 in diameter pre-polished stainless steel discs. Thirty seconds before termination of the electrodeposition, 5 ml of 1:10 NH_4OH are added. At termination, the deposition is stopped by raising the platinum anode out of the electrolyte and immediately pouring the electrolyte from the cell. The cell is flushed 3 times with 1% NH_4NO_3 - 1:99 NH_4OH, followed by a distilled water rinse. The cell is quickly disassembled and the disc is rinsed with a stream of ethyl alcohol which has been adjusted to pH 8 with NH_4OH. The excess alcohol is absorbed from the edge of the disc on a paper towel, and the disc is then dried on a porcelain-topped hot plate at no more than 250°C.

IV. Alpha Energy Analysis

A. Instrumentation

The electrodeposited plutonium is counted in a vacuum chamber for 1000 to 1333 minutes using a 450 mm^2 silicon surface-barrier detector. The pulse generated by the detector is fed through preamplification, amplification and pulse shaping stages to a true multiplexing routing system which enters the information from each of four detectors into the four quadrants of either a 1024 or 4096 channel analyzer with no loss of information due to coincidence and with no introduction of extraneous information due to cross talk between quadrants.

B. Calculations

Data output from the analyzer is by means of teletype or a parallel printer. The analyzer can integrate selected peak areas for printout, but all other calculations are done by hand with the assistance of a programmable calculator. Percent tracer yield and fCi plutonium are calculated by the following formulae:

$$\% \text{ Tracer Yield} \pm 1\sigma = G \pm g = \frac{[(A-B) \pm b]\ (C \pm c)(100)}{D \pm d} \qquad \text{Eq. 1}$$

where A = gross counts per minute (c/min) in the tracer peak region chosen

B = background c/min in the tracer peak region chosen

b = one sigma counting error about (A-B) calculated by Eq. 3 or 4.

C = reciprocal of the counting efficiency

c = one sigma error about C

D = dis/min of tracer added

d = one sigma error about D

$$\text{fCi } ^{239}\text{Pu or } ^{238}\text{Pu} \pm 1\sigma = \frac{[(E-F) \pm f]\ (C \pm c)\ (450.45 \text{ fCi/dpm})}{G \pm g} \qquad \text{Eq. 2}$$

where E = gross c/min in the chosen peak region of the plutonium isotope of interest

F = background c/min in the plutonium peak region

f = one sigma counting error about E-F

C,c = as defined above

G±g = Tracer Yield ± 1σ from Eq. 1.

One sigma counting errors are calculated by one of the following methods:

1) If the gross sample counts are $\geq$ 15 counts, then,

$$(E-F) \pm f \text{ of Eq. 2} = E-F \pm \sqrt{\frac{E}{t_s} + \frac{F}{t_B}} \qquad \text{Eq. 3.}$$

where, t_s = sample counting time

t_B = background counting time

2) If the gross sample counts are < 15 counts, then the upper and lower end points of the 95% confidence interval about the observed net counts are obtained from table D.2 as reported by Nicholson (1963) and the net observed counts $\pm$ 1σ are estimated from the following:

$$(E-F) \pm f \text{ of Eq. 2} = (E-F) \pm \frac{(UL-LL)}{(4)\ (t_s)} \qquad \text{Eq. 4}$$

where, UL = upper end point of the 95% confidence interval

LL = lower end point of the 95% confidence interval

Statistical errors about the count rate, the counting efficiency, the tracer yield, and the tracer dis/min are carried through to to the final results by means of Eq. 5

$$\frac{(A \pm a)\ (B \pm b)}{(C \pm c)} = \frac{AB}{C} \pm \frac{AB}{C}\sqrt{\prod_i (1 + V_i) - 1} \qquad \text{Eq. 5}$$

where i = a, b, c, ...

$$V_i = \left(\frac{a}{A}\right)^2, \left(\frac{b}{B}\right)^2, \left(\frac{c}{C}\right)^2, \ldots$$

which is the formula for the relative variance of a product or quotient in terms of the relative variance of each of the factors.