

PROCEDURES FOR DETERMINATION OF PLUTONIUM-239, 240 IN  
SOIL SAMPLES

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la Determinacion de Plutonio-239,240 en Muestras de Suelos".  
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Translated by

SCITRAN COMPANY  
1482 East Valley Road  
Santa Barbara, California 93108  
(805)969-2413  
FAX (805)969-3439

JUNTA DE ENERGIA NUCLEAR ["NUCLEAR ENERGY COUNCIL"]  
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DIVISION: RADIOLOGICAL ENVIRONMENT OPERATIONS UNIT

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TITLE:        PROCEDURES FOR DETERMINATION OF PLUTONIUM-239, 240 IN  
              SOIL SAMPLES.

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Revision No.	0	1	2	3	4
Prepared by	Emma Iranzo				
	/signature/				
Date	9-2-1986				
Reviewed by	Emilio Iranzo				
	/signature/				
Date	10-3-1986				
Supervised by	[illegible]				
Quality	/signature/				
Assurance					
Date	12-5-86				
Approved by	F. Mingot				
	/signature/				
Date	12-6-86				

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TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Principle of the Method
  - 3.2 Instrumentation and Materials
    - 3.2.1 Instrumentation
    - 3.2.2 Special reagents
  - 3.3 Treatment of the Sample
    - 3.3.1 Pretreatment
    - 3.3.2 Wet incineration
    - 3.3.3 Addition of marker
  - 3.4 Radiochemical Separation
    - 3.4.1 Preparation of the Ionic Resin Column
    - 3.4.2 Ion Exchange and Elution
  - 3.5 Purification and Electrodeposit
  - 3.6 Measurement
    - 3.6.1 Equipment
    - 3.6.2 Preparation of Standard Solution
    - 3.6.3 Sample Measurement
  - 3.7 Calculations
    - 3.7.1 Calculation of Lower Limit of Detection according to the Currie Criterion
    - 3.7.2 Calculation of Minimum Detectable Radiation
    - 3.7.3 Calculation of Errors
    - 3.7.4 Calculation of recovery
  - 3.8 Expression of Results
4. RESPONSIBILITIES
5. REFERENCES
6. ENCLOSURES
  - 6.1 Additional Treatment of Soil
  - 6.2 Decontamination

## 1. PURPOSE

To describe the method used in the Radiological Environment Operations Unit for determination of plutonium in soil samples.

## 2. SCOPE

Method is applicable to both surface and deep soil samples.

## 3. COMPOSITION

3.1 Principle of the Method

Levels of Pu 239 and Pu 240 in soil samples are determined through extraction of the plutonium contained in the samples with acid mixtures, separation of the specified radionuclides by ion exchange, purification and separation by electrodeposit and final measurement by alpha spectrometry.

3.2 Instrumentation and Materials

## 3.2.1 Instrumentation

- Ion exchange columns of 8 ml volume.
- Stainless steel planchets of 14 mm diameter.
- Electrodeposit cells.
- Electrodeposit device with platinum anode.
- Alpha spectrometer with silicon barrier semiconductor detectors.

- Aluminum blocks
- Centrifuge
- Crucible
- Centrifuge tubes and precipitate receptacles

### 3.2.2 Special Reagents

- Pu 236 marker solution
- DOWEX AG 1 x 2 (50-100 mesh) anionic resin  
in chloride form
- Nitric acid
- Hydrochloric acid
- Hydrofluoric acid
- Hydroxylamine hydrochlorate
- Ammonium oxalate
- Ammonium hydroxide

## 3.3 Treatment of the Sample

### 3.3.1 Pretreatment

- a) Homogenize the sample
- b) Dry 8 to 12 grams of sample in a crucible  
at 120°C, to eliminate moisture.
- c) Calcine at 600°C for 6 hours, to destroy  
organic material.

### 3.3.2 Wet Incineration

- a) Place 10 grams of each calcined sample in  
100 ml centrifuge tubes.
- b) Add 50 ml of 1M HF-12M HNO<sub>3</sub> solution to  
each sample.
- c) Cover the centrifuge tubes with watch  
crystals and place them on an aluminum block. Maintain at a  
temperature of 160-180°C for 15 minutes to extract the plutonium  
contained in the sample.
- d) Remove the centrifuge tubes from the block  
and, while hot, collect the particles adhering to the tube walls  
during the boiling process; allow to return to room temperature.

- e) Centrifuge the tubes for 5 minutes at 2000 rpm.
- f) Remove the tubes from the centrifuge and decant into a precipitate receptacle.
- g) Repeat steps b) to f) a minimum of three times.
- h) Wash out the solid portion remaining in the tubes after decantation, with 15-20 ml of HNO<sub>3</sub>, 7, 8N.
- i) Centrifuge this washed portion for 5 minutes at 2000 rpm and decant as before.
- j) Pour the decantation liquid into a gauged glass vessel, washing it several times with HNO<sub>3</sub>, 7, 8N.
- k) Place a portion corresponding to 0.5 g of soil into a centrifuge tube and dry it on an aluminum block at a temperature above 90°C, to eliminate projections.
- l) In those cases where the radioactivity is very low, and the entire sample must be taken, the procedure indicated in Enclosure 6.1 will be followed.

### 3.3.3 Addition of Marker

- a) Add 100 ml of HNO<sub>3</sub>, 7, 8N and 1 ml of Pu 236 solution containing about 74 mBq (2pCi) of this radionuclide, in order to calculate the analytic recovery to use in the corresponding calculations.
- b) Cover the tubes with watch crystals and heat at 90°C for 90 minutes.
- c) Leave overnight at room temperature.

## 3.4 Radiochemical Separation

### 3.4.1 Preparation of the ionic resin column

- a) Place spun glass in the column tube and moisten it with distilled water.
- b) Fill the column with resin suspended in distilled water.

- c) Permit the resin (DOWERX AG-1 X 2, 50-100 mesh) to settle.
- d) Wash the resin with HNO<sub>3</sub>, 7, 8N solution until it does not give a chloride reaction with silver nitrate.
- e) Allow the nitric acid to pass completely before going on to the next step.

#### 3.4.2 Ion exchange and elution

- a) Pass the nitric solution containing the sample through the anionic resin AG 1 X 2 (50-100 mesh) in chloride form with a flow rate not to exceed 2 ml/min.
- b) Wash three times with 50 ml portions of HNO<sub>3</sub>, 7, 8N.
- c) Discard the washing liquids.
- d) Add 15 ml of HCl concentrate. Discard the effluent.
- e) Add about 0.25 g of hydroxylamine hydrochlorate to the column.
- f) Elute the plutonium held in the column with three 5 ml portions of HCl 0, 5N.

#### 3.5 Purification and electrodeposit

- a) Evaporate the eluted matter under an infrared lamp until dry.
- b) Dissolve the residue in 1 ml of HCl 12N.
- c) Evaporate under an infrared lamp until dry.
- d) Add 3 ml of 4% ammonium oxalate and 1 ml of hydrochloric acid 1N.
- e) Lightly heat under infrared lamp; approximately one minute.
- f) Transfer the solution to the electrolytic cell, washing the vessel several times with distilled water which is added to the cell until it totals 10 ml.
- g) Electrodeposit at a current intensity of 200 mA for three hours.

h) After three hours, without disconnecting the current, add 2 ml of ammonium hydroxide (50% by volume) to bring the electrolyte's pH to 7.5, and verify the same with litmus paper.

i) Separate the electrolytic cell from the stand, previously disconnecting the current.

j) Wash the cell with distilled water.

k) Separate the planchet from the cell and wash it with distilled water.

l) Allow the planchet to dry and flame it.

### 3.6 Measurement

#### 3.6.1 Equipment

The radioactivity of the electrodeposited Pu 239 + Pu 240 is measured by alpha spectrometry with silicon barrier semiconductor detectors Model TR-21-307-100 (ORTEC), with a nominal active area of 300 mm<sup>2</sup>, a resolution of 21 Kev and a dead time of 0.5  $\mu$ sec. The operating power supply is 100 volts.

These detectors are connected to an ORTEC multichannel analyzer and to a DIGITAL PDP-11.23 microcomputer with the RT-11 operating system.

#### 3.6.2 Preparation of the Standard Solution

The plutonium 236 solution is obtained from OAK RIDGE NATIONAL LABORATORY, with the applicable certificates.

• The solution to mark the samples is prepared in nitric acid 7, 8N, heated for two hours at 8°C, to stabilize the plutonium in valence state IV.

• The solution thus prepared is diluted in nitric acid 7, 8N until a radioactivity concentration of approximately 74 mBq/ml (2pCi/ml) is obtained.

• The standard solution is evaluated by means of an alpha count in a proportional continuous gas flow counter; this measurement is verified by another taken with alpha spectrometry.

• The spectrometer is calibrated and the baselines determined in accordance with Procedure No. \_\_\_\_\_ of the Radiological and Environmental Protection office.

### 3.6.3 Measurement of the sample

• The planchet prepared in accordance with 3.5 is introduced into the counter equipment for measurement in accordance with Procedure No. \_\_\_\_\_ of the Radiological and Environmental Protection Office.

• The counting time will never be less than 1440 minutes, depending upon the radioactivity of the sample.

## 3.7 Calculations

3.7.1 Calculation of the Lower Limit of Detection (LLD) according to the Currie Criterion.

The American standards ASTH (Volume 12, 01, 1983, C-100. For determination of plutonium 239 and plutonium 238 with a 95% confidence level, for which 2 x the square root of 2K takes a value of 4.66, and considering a maximum of 10 counts of total baseline, the detection limit of the device is given by the formula:

$$LLD = \frac{4.66}{2.2 \text{ CE}} \left( \frac{C_b}{T_b} \right)^{1/2}$$

where:

$C_b$  = Counts per minute from baseline for radionuclide i.

$T_b$  = Counting time from baseline expressed in minutes.

CE = Count efficiency

With:

An efficiency of 30%  
 Counting time of 1440 minutes  
 A baseline of 10 total counts  
 LLD = 0.29 mBq = 0.008 pCi

### 3.7.2 Calculation of Minimum Detectable Radioactivity (MDR)

This calculation requires a sufficient recount (at least 50 total counts) which will permit a close enough approximation of the Poisson distribution to that of Gauss, for us to use Gaussian statistics.

The Currie criterion for a small number of events is:

$$\text{MDR} = \frac{2.71 + 4.66 \sqrt{C_t}}{T \cdot R \cdot CE \cdot E}$$

where:

$C_t$  = Total counts = 50  
 $T$  = Counting time = 4320 min.  
 $R$  = Chemical yield = 70%  
 $CE$  = 30%

$$\text{MDR} = \frac{2.71 + 4.66 \sqrt{50}}{4320 \times 2.22 \times 0.3 \times 0.7} = 0.017 \text{ mBq/planchet}$$

(0.017 pCi/planchet)

### 3.7.3 Calculation of Errors

3.7.3.1 The alpha spectrometer calibration error is given by the errors of the secondary standards utilized and certified by the National Bureau of Standards (N.B.S.)

Rev \_\_\_ Date 9-2-86

Page 9 of 14

The variance of the different calibration measurements is sufficiently low to permit reproduction of the measurement in 98.6% of the cases.

$$LLD = 0.29 \pm 0.0034 \text{ mBq (0.006 } \pm 0.000093 \text{ pCi)}$$

which involves an error of 1.15% with 95% confidence.

3.7.3.2 The error of the minimum detectable radioactivity will be formed by the accumulation of the following random errors:

- a) Spectrometer calibration error which is considered random.
- b) Radiochemical analysis error.
- c) Micropipette error.
- d) Sample quartering error.
- e) Counting error.

- Calibration error is 0.92%
- The analysis error with Carrier ion exchange is 10%
- The micropipette error is 3%.
- The sample quartering error is 11%
- The counting error with 95% confidence is:

$$2 \sigma = \frac{2 \sqrt{Ct}}{T} = \frac{\sqrt{50}}{4320} = 0.0032$$

which represents 28.8%

Considering that:

$$E_{\text{total}} = \sqrt{(E_a\%)^2 + (E_b\%)^2 + (E_c\%)^2 + (E_d\%)^2 + (E_e\%)^2}$$

The total error = 32.56%

Rev \_\_\_ Date 9-2-86

Page 10 of 14

Minimum detectable radioactivity in a counting time of 4320 minutes with 95% confidence:

MDR =  $0.63 \pm 0.20$  mBq/sample ( $0.017 \pm 0.0054$  pCi/sample)

#### 3.7.4 Calculation of Recovery

The chemical recovery of the method is calculated through the use of the internal standard of Pu 236, and is expressed as a percentage of the added standard and that counted.

$$\% \text{ recovery} = \frac{\text{mBq counted} \times 100}{\text{mBq added}}$$

Sample analysis of marked targets should be employed with a given frequency in a series of analyses.

#### 3.8 Expression of Results

The final results of the sample's Pu 239 + Pu 240 alpha radioactivity is expressed as radioactivity per gram of soil. The results by sample will be expressed in the following manners:

- Case a)        A > LLD

The sample thus has a true radioactivity value, the result being expressed as A2 followed by the corresponding units.

- Case b)         $\leq$  A LLD

The result will be expressed as LLD followed by its units.

## 4. RESPONSIBILITIES

It is the responsibility of the director of the Radiological and Environmental Protection office to:

- Approve the application of these procedures and direct such application when considered appropriate.

- Monitor the proper application of these procedures.

It is the responsibility of the chief of the Radiological Environment Operations Unit to:

- Keep these procedures updated.

- Designate the chief of the laboratory where these procedures are carried out.

- Monitor the proper application of these procedures.

It is the responsibility of the laboratory chief to:

- Supervise the correct application of the procedures by the analysts in his/her charge.

- Prepare updating of these procedures.

- Approve the results by signature.

It is the responsibility of the analysts to:

- Perform the procedures correctly.

- Request the information needed and deliver that obtained in timely fashion and the proper form.

## 5. REFERENCES

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5. Manual of Analytical Methods for Radiobioassay. Los Alamos Scientific Laboratory. July 1983.
6. A. YAMATO. An anion exchange method for the determination of Am 241 and plutonium in environmental and biological samples. Journal of Radioanalytical Chemistry, Vol. 75, Nos. 1-2 (1982) 265-273.
7. Loveridge B.A. Quantitative Electrodeposition [of] plutonium for Alpha and Beta Assay. AER-R 3266 (1960).
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## 6. ENCLOSURES

### 6.1 Additional Treatment of Soil

In cases where the radioactivity of the sample is very low, and the entire sample must be used starting with step i) of the process described in 3.3.2, the following procedure will be adopted:

- a) Dry out the decantation liquids.
- b) Dilute in 300 ml of distilled water.
- c) Add approximately 12 g of oxalic acid and agitate until dissolution is complete.
- d) In the case of calcareous soil, acidify to pH 1.5, in order to produce coprecipitation of the transuranides as calcium oxalate.

If the soil is not calcareous, add 2 ml of calcium solution and acidify to pH 1.5 to produce coprecipitation.

- e) Filter through filter paper and discard the filtrate.
- f) Calcine the filter containing the precipitate in the crucible at 400° for 24 hours.
- g) Dissolve the ashes with HCl concentrate.
- h) Dry.
- i) Continue step a. as described in 3.3.3 of the procedure.

### 6.3 Decontamination

#### Reagents

- Saturated solution of potassium permanganate (64 g/l)
- Sulphuric acid 0, 2N
- Sodium sulphite acid (5%)
- DABEER detergent

Decontamination

There are two procedures:

Procedure A

- Saturated mixture of potassium permanganate and sulphuric acid 0, 2N (V/V) (Solution A).
- Wash the material with Solution A.
- Wash with sodium sulphite acid.
- Wash with distilled water.

Procedure B

- Wash the material while hot, with 50% nitric acid.
- Wash with DABEER detergent.
- Wash with distilled water.



TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Principle of the Method
  - 3.2 Instrumentation and Materials
    - 3.2.1 Instrumentation
    - 3.2.2 Special reagents
  - 3.3 Treatment of the Sample
    - 3.3.1 Pretreatment
    - 3.3.2 Wet incineration
    - 3.3.3 Dissolution and Stabilization
  - 3.4 Radiochemical Separation
    - 3.4.1 Preparation of the Ionic Resin Column
    - 3.4.2 Ion Exchange and Elution
  - 3.5 Treatment of the Americium Portion
    - 3.5.1 Precipitation
    - 3.5.2 Wet Incineration
    - 3.5.3 Dissolution and Preparation
  - 3.6 Purification of the Americium Portion
    - 3.6.1 Preparation of the Ionic Resin Column
    - 3.6.2 Ion Exchange and Elution
  - 3.7 Electrodeposit of the Plutonium and Americium Portions
  - 3.8 Measurement
    - 3.8.1 Equipment
    - 3.8.2 Preparation of Standard Solutions
    - 3.8.3 Sample Measurement
  - 3.9 Calculations
    - 3.9.1 Calculation of Limit of Detection
    - 3.9.2 Calculation of Minimum Detectable Radioactivity
    - 3.9.3 Calculation of Errors
    - 3.9.4 Calculation of recovery
  - 3.10 Expression of Results
4. RESPONSIBILITIES
5. REFERENCES
6. ENCLOSURES
  - 6.1 Decontamination

1. PURPOSE

To describe the method used in the Radiological Environment Operations Unit of the Radiological and Environmental Protection office for determination of plutonium and americium levels in air samples.

2. SCOPE

Will be applied in the asbestos filters used for air sampling.

3. COMPOSITION

3.1 Principle of the Method

Pu239 + Pu240 and Am241 levels in air samples are determined by dissolving the filter in acid solvents, separation by ion exchange to obtain the plutonium portion and the americium portion, electrodeposit of the plutonium portion, precipitation and purification of the americium portion by ion exchange, electrodeposit of the americium portion, measurement of radioactivity by alpha spectrometry of the electrodeposited Pu and Am portions.

3.2 Instrumentation and Materials

3.2.1 Instrumentation and material

- Crucible
- Heating plates
- Ion exchange columns
- Precipitate receptacles
- Teflon rods and receptacles

REV \_\_\_\_ Date: \_\_\_\_

Page 3 of 16

- Porcelain capsules
- Stainless steel planchets, diameter 14 mm
- Electrodeposit cells
- Platinum anode electrodeposit system
- Alpha spectrometer with silicon barrier semiconductor detectors.

### 3.2.2 Special reagents

- Pu-236 marker solution
- Cm-244 marker solution
- DOWEX AG 1X2 (50-100 mesh) resin in chloride form
- BIORAD 1X8 resin in chloride form
- Nitric acid
- Hydrochloric acid
- Hydrofluoric acid
- Oxalic acid
- Hydroxylamine hydrochlorate
- Ammonium hydroxide
- Methanol
- Ammonium thiocyanate
- Ammonium nitrate
- Calcium solution (20 mg/ml)
- Methyl red

## 3.3 Treatment of the Sample

### 3.3.1 Pretreatment

a) Place the entire filter in a 600 ml precipitate receptacle, cover with perforated aluminum foil and calcine in the crucible at 450°C for eight hours.

b) Add the plutonium-236, 78 mBq (2pCi) and curium-244, 78 mBq(2pCi) tracers.

3.3.2 Wet incineration

- a) Dissolve the hot filter in a solution of HF-HNO<sub>3</sub> (V/V) (200 ml of acid solution).
- b) Stir with a teflon rod until the filter is completely dissolved.
- c) Dry.

3.3.3 Dissolution and stabilization

- a) Dissolve the residue with a few drops of HCl concentrate and H<sub>2</sub>O<sub>2</sub>.
- b) Dry.
- c) Dissolve in 500 ml of HNO<sub>3</sub> 7, 8 N.
- d) Stabilize the plutonium at valence IV by heating on the hot plates at 80°C for two hours, covering the sample with a watch crystal.
- c) Cool and let sit until the following day.

3.4 Radiochemical separation

The resin utilized is DOWEX AG 1X2 (50-100 mesh) anionic resin which retains the Pu and Th and the Am, U, Cm and Fe elute in it when it is in nitric acid 7, 8 N medium.

3.4.1 Preparation of the resin

Utilize exchange columns with a switch, with a length of 17 cm and an inside diameter of 11 mm.

- a) Place spun glass at the bottom of the column, moistening it with distilled water.
- b) Fill the entire volume of the column with resin suspended in distilled water (16 ml of volume).
- c) Let the resin settle.
- d) Wash the resin with HNO<sub>3</sub>, 7, 8N until it gives no chloride reaction.

#### 3.4.2 Ion exchange and elution

- a) Pass the nitric acid solution containing the sample through the AG 1X2 anionic resin at a rate not in excess of 2 ml/min.
- b) Wash three times with 50 ml portions of HNO<sub>3</sub>, 7, 8N.
- c) Recover the 650 ml of nitric acid liquid containing the americium portion.
- d) Wash with five volumes of HCl 10 N column to eliminate the thorium.
- e) Discard the washing liquid.
- f) Elute the plutonium fixed in the column with a 1 g solution of hydroxylamine hydrochlorate dissolved in 80 ml of HCl 0.5N

### 3.5 Treatment of the Americium Portion

#### 3.5.1 Precipitation

- a) Dry the americium portion.

- b) Dissolve in 200 ml of distilled water
- c) Heat on the hot plates do facilitate dissolution. Allow to cool.
- d) Add 30 ml of saturated oxalic acid.
- e) Add 2 ml of 20 mg/ml calcium solution.
- f) Precipitate as calcium oxalate at pH 1.2 with concentrated ammonium hydroxide.
- g) Let sit until the next day.
- h) Filter through a paper filter and centrifuge three times for 15 minutes at 3000 rpm in ORTO centrifuge, using 250 cc centrifuge tubes.
- i) Discard the filtrate liquid and the floating matter from each centrifuging.
- j) If the centrifuging optioin has been selected, go to subparagraph c) of section 3.5.3.

#### 3.5.2 Incineration

- a) Incinerate the filter containing the calcium oxalate precipitate in the crucible at 400°C for four hours.

#### 3.5.3 Dissolution and Preparation

- a) Dissolve the ashes with a few drops of HCl concentrate and oxygenated water.
- b) Dry
- c) Dissolve in 14 ml of boiling HNO<sub>3</sub> concentrate, assisting the process with a glass rod. Add 186 ml of CH<sub>3</sub>OH, to make the solution 1M HNO<sub>3</sub>/93% CH<sub>3</sub>OH.

REV \_\_\_ Date 6-2-86

Page 7 of 16

d) Add 3 g of  $\text{NH}_4\text{NO}_3$ , until it is 0.2 N and stir until it dissolves.

e) Wash the glass rod used for stirring with 2 ml of 1M  $\text{HNO}_3$ /93%  $\text{CH}_3\text{OH}$ .

f) Add to the solution 2 ml of Biorad 1X8 resin suspended in a medium of 1M  $\text{HNO}_3$ /93%  $\text{CH}_3\text{OH}$ .

g) Cover with Nescofilm and let sit until the following day.

### 3.6 Purification of the Americium Portion

#### 3.6.1 Preparation of the ionic resin column.

Use columns with a switch, 9 cm long and 13 mm in diameter.

Since the resin medium is methanol, always cover the mouth of the column with Nescofilm to prevent alcohol evaporation to the extent possible.

a) Place spun glass in the bottom of the column, moistening it with distilled water.

b) Fill the column with BIORAD 1X8 resin suspended in distilled water.

c) Allow the resin to settle.

d) Wash the resin with 1M  $\text{HNO}_3$ /93%  $\text{CH}_3\text{OH}$ .

#### 3.6.2 Ion Exchange and Elution

a) Pass solution f (section 3.5.3) containing the sample, through the BIORAD 1X8 anionic resin.

REV \_\_\_ Date 6-2-86

Page 8 of 16

- b) Wash three times with 20 ml portions of 1M HNO<sub>3</sub>/93% CH<sub>3</sub>OH.
- c) Discard the washing liquid.
- d) Wash with five C.V. of 0.1 N HCl/0.5 M NH<sub>4</sub>SCN/80% CH<sub>3</sub>OH.
- e) Discard the washing liquid.
- f) Elute the americium fixed in the column with 50 ml of 1.5 N HCl/ CH<sub>3</sub>OH.

3.7 Electrodeposit of the Pu and Am Portions

- a) Dry the Pu and Am elutions.
- b) Add, at least twice, 2 cc of HCl concentrate.
- c) Evaporate to dryness each time without allowing burning.
- d) Add 2 cc of HCl 2 N and one drop of methyl red.
- e) Neutralize the solution with 50% ammonia solution, just to the point where the red color disappears.
- f) Add at least 2 drops of HCl concentrate to reestablish acidity (pH = 3.5) (appearance of a light pink tone).
- g) Place this solution in the electrodeposit cell, washing the receptacle with distilled water and adding it to the cell.
- h) Connect 400 mA current, and electrodeposit for an hour and a half.
- i) Add 2 cc of 50% ammonia a few minutes before turning off the current.
- j) Wash the cell with distilled water and allow to dry

### 3.8 Measurement

#### 3.8.1 Equipment

The plutonium-239, 240 and americium-241 radioactivity is measured with alpha spectrometry, with Model TA-21-307-100 (ORTEC) silicon barrier semiconductor detectors, with a nominal active area of 300 mm<sup>2</sup>, a resolution of 21 Kev and a dead time of 0.5 microseconds. The power supply is 100 volts.

These detectors are connected to a multichannel ORTEC analyzer and to a DIGITAL PDP-11/23 microcomputer with the RT-11 operating system.

#### 3.8.2 Preparation of the Standard Solutions

a) The plutonium-236 solution is obtained from OAK-RIDGE NATIONAL LABORATORY, with its applicable certificates.

- The sample tracer solution is prepared with nitric acid 8N heated for 2 hours at 80°C in order to ensure that the plutonium is stabilized in valance status IV.

- The solution thus prepared is diluted with nitric acid 0.8N until a radioactivity of 0.8 MBq/ml (2pCi/ml) is obtained.

- The standard solution is evaluated immediately with an alpha count in a proportional continuous gas flow counter, verifying the other measurement taken with alpha spectrometry.

b) The curium-244 solution is obtained in the LABORATORIE DE METROLOGIE DES RAYONNEMENTS IONISANTS (CEA), with its applicable certificates.

- The initial solution was prepared with curium nitrate diluted in nitric acid 1N, with an initial radioactivity of  $3.6 \times 10^4$  Bq/g.

REV \_\_\_ Date 6-2-86

Page 10 of 16

• This initial solution is diluted in nitric acid 1N until a radioactivity of ?? mBq.ml is obtained.

c) Spectrometer calibration and determination of baselines is done in accordance with office procedure No.

### 3.8.3 Sample Measurement

• The planchet prepared in accordance with 3.7 is introduced into the counter for measurement according to procedure No. of the Radiological and Environmental Protection office.

• The counting time will never be less than 2880 minutes.

### 3.9 Calculations

3.9.1 Calculation of Lower Limit of Detection (LLD) according to the Currie Criterion

American ASTH standards (Volume 12, 01, 1983, C-100)

For determination of Plutonium-239, plutonium-238 and americium-241 with a 95% confidence level, for which 2 x the square root of 2K takes the value of 4.66, and considering a maximum of 10 counts of the total baseline, the lower limit of detection of the device is given by the formula:

$$LLD = \frac{4.66}{2.2 CE} \left( \frac{C_r}{T_r} \right)^{\frac{1}{2}}$$

$C_r$  = Baseline counts per minute for the i isotope.

$T_r$  = Counting time of the baseline expressed in minutes

CE = Counting efficiency

with:

An efficiency of 30%

A counting time of 1440 minutes

A baseline of 10 total counts

LLD = 0.30 mBq (0.008 pCi)

REV \_\_\_\_ Date 8-2-86

Page 11 of 16

3.9.2 Calculation of Minimum Detectable Radioactivity (MDR).

This calculation requires a sufficient recount (at least 50 total counts) which will permit a close enough approximation of the Poisson distribution to that of Gauss, for us to use Gaussian statistics.

The Currie criterion for a small number of events is:

$$MDR = \frac{2.71 + 4.66 \sqrt{C_t}}{T \cdot R \cdot CE \cdot F}$$

- C<sub>t</sub> = Total counts
- R = Chemical yield = 70%
- CE = 30%
- T = Counting time

$$MDR = \frac{2.71 + 4.66 \sqrt{50}}{4320 \times 2.22 \times 0.3 \times 0.7} = 0.63 \text{ mBq/planchet}$$

(0.017 pCi/planchet)

3.9.3 Calculation of Errors

3.9.3.1 The alpha spectrometer calibration error is given by the errors of the secondary standards utilized and certified by the N.B.S.

The variance of the different calibration measurements is sufficiently low to permit reproduction of the measurement in 98.6% of the cases.

LLD = 0.296 ± 0.0034 mBq (0.008 ± 0.000092 pCi) which involves an error of 1.15% with 95% confidence.

3.9.3.2 The error of the minimum detectable radioactivity will be formed by the accumulation of the following random errors:

REV \_\_\_ Date 6-2-86

Page 12 of 16

- a) Spectrometer calibration error which is considered random.
- b) Radiochemical analysis error.
- c) Micropipette error.
- d) Counting error

- Calibration error is 0.92%
- The analysis error with Carrier ion exchange is 10%
- The micropipette error is 3%.
- The counting error with 95% confidence is 28.8%

$$\text{Total error} = \sqrt{E_a^2 + E_b^2 + E_c^2 + E_d^2}$$

$$\text{Total error} = 30.65\%$$

Minimum detectable radioactivity in a counting time of 4320 minutes with 95% confidence:

$$\text{MDR} = 0.63 \pm 0.19 \text{ mBq/sample } (0.017 \pm 0.0053 \text{ pCi/sample})$$

#### 3.9.4 Calculation of Recovery

The chemical recovery of the method is calculated through the use of the internal standard of Pu 236, and is expressed as a percentage of the added standard and that counted.

$$\% \text{ recovery} = \frac{\text{Bq counted} \times 100}{\text{Bq added}}$$

Sample analysis of marked targets should be employed with a given frequency in a series of analyses.

### 3.10 Expression of Results

The final results of the sample's Pu 239 + Pu 240 and americium 241 alpha radioactivity is expressed as radioactivity per cubic meter of air. The results by sample will be expressed in the following manners:

- Case a)  $A > \text{LLD}$

The sample thus has a true radioactivity value, the result being expressed as A2 followed by the corresponding units.

- Case b)  $\leq A \text{ LLD}$

The result will be expressed as LLD followed by its units.

## 4. RESPONSIBILITIES

It is the responsibility of the director of the Radiological and Environmental Protection office to:

- Approve the application of these procedures and direct such application when considered appropriate.
- Monitor the proper application of these procedures.

It is the responsibility of the chief of the Radiological Environment Operations Unit to:

- Keep these procedures updated.
- Monitor the proper application of these procedures
- Designate the chief of the laboratory where these procedures are carried out.

REV \_\_\_\_ Date 6-2-86

Page 14 of 16

It is the responsibility of the laboratory chief to:

- Supervise the correct application of the procedures by the analysts in his/her charge.

- Prepare updating of these procedures.

- Approve the results by signature.

It is the responsibility of the analysts to:

- Perform the procedures correctly.

- Request the information needed and deliver that obtained in timely fashion and the proper form.

## 5. REFERENCES

a) Ballestra, Holm and Fukai

"Low Level Determination of Transuranic Elements in Marine Environmental Samples". Symposium on the determination of radionuclides in environmental and biological materials (Oct. 1978).

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"An Improved Radiochemical Procedure for Low Level Measurement of Americium in Environmental Matrices". Talanta, Vol. 29 (1982).

c) Holm, Ballestra, Fukai

"A Method for Ion Exchange Separation of Low Levels of Americium in Environmental Materials". Talanta, Vol. 26 (1979)

d) Yamamoto, Komuru, Sakanque.

"A Simple Sequential Separation Method of Pu and Am by Anion Exchange and Extraction Chromatography". Radiochemical Acta 29 (1981).

e) C. Gaseé Leonarte

"Determination of Transuranides in Air". 1984.

f) Manual of Analytical Methods for Radiobioassay. Los Alamos (1983)

g) A. Yamato

"An Anion Exchange Method for the Determination of Am-241 and Plutonium in Environmental and Biological Samples".

Journal of Radioanalytical Chemistry Vol. 75 1982)

h) "Reference Methods for Marine Radioactivity Studies II". AIEA 175.

## 6. ENCLOSURES

### 6.1 Decontamination of Glass and Teflon Material

#### Reagents

- Saturated potassium permanganate solution (64 g/l).
- Sulphuric acid 0.2N.
- Acid sodium sulphite (5%)
- DABBER detergent

#### Decontamination

- Wash the material with DABBER detergent
- Mix the saturated potassium permanganate solution with the sulphuric acid 0.2N (V/V) (Solution A)

Procedure A

- Wash the material with solution A
- Wash with acid sodium sulphite
- Wash with distilled water

Procedure B

- Wash with hot 50% nitric acid
- Wash with DABBER detergent
- Wash with distilled water

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DIVISION: TRANSURANIDE IMPACT AND GEOCHEMISTRY

Pages 13

REVISION No. 0

Date: 11-10-87

SPECIFIC PROCEDURE No. MA/14 Comments\_\_\_ Approval \_\_\_ Execution\_\_\_

TITLE: PROCEDURES FOR DETERMINATION OF PLUTONIUM-239 + PLUTONIUM  
240 IN VEGETATION SAMPLES.

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Revision No.	0	1	2	3	4
Prepared by	Emma Iranzo Martín /signature/				
Date	11-10-87				
Reviewed by	Emilio Iranzo González /signature/				
Date	11-10-87				
Supervised by	[illegible]				
Quality Assurance	/signature/				
Date	11-24-87				
Approved by	F. Mingot /signature/				
Date	1/21/88				

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Rev 0 Date 11-10-87

TITLE:  
PROCEDURE FOR DETERMINATION OF  
PLUTONIUM-239+240 IN VEGETATION  
SAMPLES

Page 1 of 13

TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Principle of Method
  - 3.2 Instrumentation and Materials
    - 3.2.1 Instrumentation
    - 3.2.2 Special Reagents
  - 3.3 Treatment of the Sample
    - 3.3.1 Pretreatment
    - 3.3.2 Wet incineration
  - 3.4 Radiochemical Separation
    - 3.4.1 Preparation of the ionic resin column
    - 3.4.2 Ion Exchange and Elution
  - 3.5 Purification and Electrodeposit
  - 3.6 Measurement
  - 3.7 Calculation
4. RESPONSIBILITIES
5. REFERENCES
6. ENCLOSURES

IMPACT OF TRANSURANIDES  
C.I.E.M.A.T.

Rev 0 Date 11-10-87

TITLE:  
PROCEDURE FOR DETERMINATION OF  
PLUTONIUM-239+240 IN VEGETATION  
SAMPLES

Page 2 of 13

1. PURPOSE

To describe the method used in the Transuranide Impact and Geochemistry Operations Unit for determination of plutonium in vegetation samples.

2. SCOPE

Method is applicable to cultivated and wild vegetation.

3. COMPOSITION

3.1 Principle of the Method

Levels of Pu 239 and Pu 240 in vegetation samples are determined through dissolution of the ashes of the samples with acid mixtures, separation of the specified radionuclides by ion exchange, purification and separation by electrodeposit and final measurement by alpha spectrometry.

The method uses Pu-236 as a marker.

3.2 Instrumentation and Materials

3.2.1 Material and equipment

- Ion exchange columns of 8 ml volume.
- Stainless steel planchets of 14 mm diameter.
- Electrodeposit cells.
- Electrodeposit device with platinum anode.

- Alpha spectrometer with silicon barrier semiconductor detectors.
- Drying stove
- Crucible
- Hot plate

3.2.2 Special Reagents

- Pu 236 marker solution
- DOWEX AG 1 x 2 (50-100 mesh) anionic resin in chloride form
- Nitric acid
- Hydrochloric acid
- Hydrofluoric acid
- Hydroxylamine hydrochlorate
- Oxygenated water (30%)
- Ammonium oxalate
- Ammonium hydroxide

3.3 Treatment of the Sample

3.3.1 Pretreatment

- Carbonization. Place the sample (3-5 kg) in the drying stove, gradually increasing the temperature to 250°C.
- Incineration. Place the carbonized sample in the crucible at 500°C for 12 hours.

3.3.2 Wet Incineration

- a) Weigh 0.5-1 g of ash on precision scales.
- b) Add 50 ml of an acid mixture of 50% v/v of nitric acid (65%) and hydrofluoric acid (48%)
- c) Dry in a teflon receptacle at a constant temperature of 90°C.

- d) Repeat the process
- e) Add 25 ml of nitric acid concentrate (65%).
- f) Dry.
- g) Repeat the process.
- h) Add 25 ml of nitric acid 7.8N and 3 ml of oxygenated water to the residue.
- i) Place the receptacle on a hot plate and digest with addition of oxygenated water (30%) in 1 ml portions until the solution is clear.
- j) Complete the volume with nitric acid 7.8N up to 100-200 ml, depending upon the weight of the sample being analyzed.
- k) Add 74 mBq (2.0 pCi) of the marker solution (Pu-236).
- l) Heat for 90 minutes at 80°C, covering the receptacles with a watch crystal.
- m) Leave overnight at room temperature.

### 3.4 Radiochemical Separation

#### 3.4.1 Preparation of the ionic resin column

- a) Place spun glass in the column tube and moisten it with distilled water.
- b) Fill the column with resin suspended in distilled water.
- c) Permit the resin to settle, adding whatever amount may be necessary to completely fill the column.

d) Wash the resin with nitric acid 7.8N solution until it does not give a chloride reaction.

#### 3.4.2 Ion exchange and elution

- a) Pass the nitric solution containing the sample through the ionic exchange resin at a flow rate not to exceed 1-2 ml/min.

Rev 0 Date 11-10-87

Page 5 of 13

- b) Wash with 5 volumes of the nitric acid 7.8N column.
- c) Discard the washing liquids.
- d) Add 10 ml of HCl concentrate. Discard the effluent.
- e) Add about 0.25 g of hydroxylamine hydrochlorate to the column.
- f) Elute the plutonium held in the column with three 5 ml portions of hydrochloric acid.

### 3.5 Purification and electrodeposit

- a) Evaporate the eluted matter under an infrared lamp until dry.
- b) Dissolve the residue in 1 ml of hydrochloric acid 12N.
- c) Evaporate under an infrared lamp until dry.
- d) Add 3 ml of 4% ammonium oxalate and 1 ml of hydrochloric acid 1N.
- e) Lightly heat under infrared lamp; approximately one minute.
- f) Transfer the solution to the electrolytic cell, washing the vessel several times with distilled water which is added to the cell until it totals 10 ml.
- g) Electrodeposit at a current intensity of 200 mA for three hours.
- h) After three hours, without disconnecting the current, add 2 ml of ammonium hydroxide (50% by volume) to bring the electrolyte's pH to 7.5, and verify the same with litmus paper.
- i) Separate the electrolytic cell from the stand, previously disconnecting the current.
- j) Wash the cell with distilled water.
- k) Separate the planchet from the cell and wash it with distilled water.
- l) Allow the planchet to dry and flame it.

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Rev 0 Date 11-10-87

TITLE:  
PROCEDURE FOR DETERMINATION OF  
PLUTONIUM-239+240 IN VEGETATION  
SAMPLES

Page 6 of 13

### 3.6 Measurement

#### 3.6.1 Equipment

The radioactivity of the electrodeposited Pu 239 + Pu 240 is measured by alpha spectrometry with silicon barrier semiconductor detectors Model TR-21-307-100 (ORTEC), with a nominal active area of 300 mm<sup>2</sup>, a resolution of 21 Kev and a dead time of 0.5 μsec. The operating power supply is 100 volts.

These detectors are connected to an ORTEC multichannel analyzer and to a DIGITAL PDP-11/23 microcomputer with the RT-11 operating system.

#### 3.6.2 Preparation of the Standard Solution

The plutonium 236 solution is obtained from OAK RIDGE NATIONAL LABORATORY, with the applicable certificates.

- The solution to mark the samples is prepared in nitric acid 7.8N, heated for two hours at 80°C, to stabilize the plutonium in valence state IV.

- The solution thus prepared is diluted in nitric acid 7.8N until a radioactivity concentration of approximately 74 mBq/ml (2pCi/ml) is obtained.

- The standard solution is evaluated by means of an alpha count in a proportional continuous gas flow counter; this measurement is verified by another taken with alpha spectrometry.

• The spectrometer is calibrated and the baselines determined in accordance with Procedure No. of the Radiological and Environmental Protection office.

### 3.6.3 Measurement of the sample

• The planchet prepared in accordance with 3.5 is introduced into the counter equipment for measurement in accordance with Procedure No. of the Radiological and Environmental Protection Office.

• The counting time will never be less than 1440 minutes, depending upon the radioactivity of the sample.

### 3.7 Calculations

3.7.1 Calculation of the Lower Limit of Detection (LLD) according to the Currie Criterion.

American standards ASTH (Volume 12, 01, 1983, C-100. For determination of plutonium 239 and plutonium 238 with a 95% confidence level, for which  $2 \times$  the square root of  $2K$  takes a value of 4.66, and considering a maximum of 10 counts of total baseline, the detection limit of the device is given by the formula:

$$LLD = \frac{4,66}{2,2 \text{ CE}} \left( \frac{C_f}{T_f} \right)^{\frac{1}{2}}$$

where:

$C_f$  = Counts per minute from baseline for radionuclide i.

$T_f$  = Counting time from baseline expressed in minutes.

CE = Count efficiency

With:

An efficiency of 30%  
Counting time of 1440 minutes  
A baseline of 10 total counts  
LLD = 0.29 mBq = 0.008 pCi

### 3.7.2 Calculation of Minimum Detectable Radioactivity (MDR)

This calculation requires a sufficient recount (at least 50 total counts) which will permit a close enough approximation of the Poisson distribution to that of Gauss, for us to use Gaussian statistics.

The Currie criterion for a small number of events is:

$$\text{MDR} = \frac{2,71 + 4,66\sqrt{\text{Ct}}}{\text{T} \cdot 2,22 \cdot \text{E} \cdot \text{R}}$$

where:

C<sub>t</sub> = Total counts = 50  
T = Counting time = 4320 min.  
R = Chemical yield = 70%  
CE = 30%

$$\text{MDR} = \frac{2,71 + 4,66\sqrt{50}}{4320 \times 2,22 \times 0,3 \times 0,7} = 0.63 \text{ mBq/planchet}$$

(0.017 pCi/planchet)

### 3.7.3 Calculation of Errors

3.7.3.1 The alpha spectrometer calibration error is given by the errors of the secondary standards utilized and certified by the National Bureau of Standards (N.B.S.).

Rev 0 Date 11-10-87

Page 9 of 13

The variance of the different calibration measurements is sufficiently low to permit reproduction of the measurement in 98.6% of the cases.

$$\text{LLD} = 0,29 \pm 0,0034 \text{ mBq} (0,008 \pm 0,00092 \text{ pCi})$$

which involves an error of 1.15% with 95% confidence.

3.7.3.2 The error of the minimum detectable radioactivity will be formed by the accumulation of the following random errors:

- a) Spectrometer calibration error which is considered random.
- b) Radiochemical analysis error.
- c) Micropipette error.
- d) Sample quartering error.
- e) Counting error.

- Calibration error is 0.92%
- The analysis error with Carrier ion exchange is 10%
- The micropipette error is 3%.
- The sample quartering error is 11%
- The counting error with 95% confidence is:

$$2 \sigma = \frac{2 \sqrt{Ct}}{T} = \frac{\sqrt{50}}{4320} = 0,0032$$

which represents 28.8%

Considering that:

$$E_{\text{total}} = \sqrt{(E_a\%)^2 + (E_b\%)^2 + (E_c\%)^2 + (E_d\%)^2 + (E_e\%)^2}$$

The total error = 32.56%

Minimum detectable radioactivity in a counting time of 4320 minutes with 95% confidence:

$$\text{MDR} = 0.63 \pm 0.20 \text{ mBq/sample (0.017} \pm 0.0054 \text{ pCi/sample)}$$

#### 3.7.4 Calculation of Recovery

The chemical recovery of the method is calculated through the use of the internal standard of Pu 236, and is expressed as a percentage of the added standard and that counted.

$$\% \text{ recovery} = \frac{\text{mBq counted} \times 100}{\text{mBq added}}$$

Sample analysis of marked targets should be employed with a given frequency in a series of analyses.

#### 3.8 Expression of Results

The final results of the sample's Pu 239 + Pu 240 alpha radioactivity is expressed as radioactivity per gram of soil. The results by sample will be expressed in the following manners:

- Case a)  $A > \text{LLD}$

The sample thus has a true radioactivity value, the result being expressed as A2 followed by the corresponding units.

- Case b)  $\leq A \text{ LLD}$

The result will be expressed as LLD followed by its units.

IMPACT OF TRANSURANIDES  
C.I.E.M.A.T.  
Rev 0 Date 11-10-87

TITLE:  
PROCEDURE FOR DETERMINATION OF  
PLUTONIUM-239+240 IN VEGETATION  
SAMPLES

Page 11 of 13

#### 4. RESPONSIBILITIES

It is the responsibility of the director of the Radiological and Environmental Protection office to:

- Approve the application of these procedures and direct such application when considered appropriate.
- Monitor the proper application of these procedures.

It is the responsibility of the chief of the Radiological Environment Operations Unit to:

- Keep these procedures updated.
- Designate the chief of the laboratory where these procedures are carried out.
- Monitor the proper application of these procedures.

It is the responsibility of the laboratory chief to:

- Supervise the correct application of the procedures by the analysts in his/her charge.
- Prepare updating of these procedures.
- Approve the results by signature.

It is the responsibility of the analysts to:

- Perform the procedures correctly.
- Request the information needed and deliver that obtained in timely fashion and the proper form.

IMPACT OF TRANSURANIDES  
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Rev 0 Date 11-10-87

TITLE:  
PROCEDURE FOR DETERMINATION OF  
PLUTONIUM-239+240 IN VEGETATION  
SAMPLES

[note: Page 12 of 13 is missing from the assigned copy]

Page 13 of 13

#### Decontamination

There are two procedures:

##### Procedure A

- Saturated mixture of potassium permanganate and sulphuric acid 0, 2N (V/V) (Solution A).
- Wash the material with Solution A.
- Wash with sodium sulphite acid.
- Wash with distilled water.

##### Procedure B

- Wash the material while hot, with 50% nitric acid.
- Wash with DABEER detergent.
- Wash with distilled water.

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DIVISION: TRANSURANIDE IMPACT AND GEOCHEMISTRY

Pages 14

REVISION No. 0

Date: 1-20-1988

SPECIFIC PROCEDURE No. MA/16 Comments\_\_\_ Approval \_\_\_ Execution\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF Pu-239+Pu-240 IN BIOLOGI-  
CAL SAMPLES AND TISSUES.

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Revision No.	0	1	2	3	4
Prepared by	Alicia Alvarez				
	/signature/				
Date	1-20-1988				
Reviewed by	EmmaIranzo				
	/signature/				
Date	5-23-88				
Supervised by	F. Alamillos				
Quality	/signature/				
Assurance					
Date	5-31-1988				
Approved by	F. Mingot				
	/signature/				
Date	6/?/88				

Form P/01

TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Principle of Method
  - 3.2 Instrumentation and Materials
    - 3.2.1 Instrumentation
    - 3.2.2 Reagents
  - 3.3 Treatment of the Sample
    - 3.3.1 Pretreatment
    - 3.3.2 Wet Attack
    - 3.3.3 Addition of Marker
  - 3.4 Radiochemical Separation
    - 3.4.1 Preparation of the Ion Exchange Columns
    - 3.4.2 Ion Exchange and Elution
  - 3.5 Electrodeposit
  - 3.6 Measurement
    - 3.6.1 Equipment
    - 3.6.2 Preparation of Standard Solution
    - 3.6.3 Calibration in Efficiencies
    - 3.6.4 Determination of Baseline
    - 3.6.5 Energy Calibration
    - 3.6.6 Sample Measurement
  - 3.7 Calculation
    - 3.7.1 Calculation of Lower Limit of Detection (LLD) according to the Currie Criterion
    - 3.7.2 Calculation of Chemical Yield
    - 3.7.3 Calculation of Minimum Detectable Concentration
    - 3.7.4 Radioactivity calculation
    - 3.7.5 Calculation of count-based uncertainties

3.8 Expression of Results

4. RESPONSIBILITIES

5. REFERENCES

1. PURPOSE

To describe the method used in the Transuranide Impact and Geochemistry Operations Unit for determination of plutonium in biological samples.

2. SCOPE

Method is applicable to animal tissue and milk samples.

3. COMPOSITION

3.1 Principle of the Method

Levels of Pu 239 and Pu 240 in biological samples are determined through extraction of the plutonium contained in the samples with acid mixtures, separation by means of ion exchange resins, preparation of the source by electrodeposit and final measurement by alpha spectrometry.

3.2 Instrumentation and Materials

3.2.1 Instrumentation

- Ion exchange columns of 35 and 5 ml capacity.
- Stainless steel planchets of 2.5 cm diameter.
- Electrodeposit cells.
- Autotransformer.
- Platinum electrode with Hg-Cu contact.
- Alpha spectrometer with silicon barrier semiconductor detectors.

- Crucible
- Stove

3.2.2 Reagents

- Pu 236 marker solution
- Pu 242 marker solution
  - AG 1 x 8 (20-50 mesh) and  
AG 1 x 8 (50-100 mesh) anionic resin
  - Hydrochloric acid
  - Nitric acid
  - Sulphuric acid
  - Hydrogen peroxide
  - Sodium nitrite
  - Ammonium Iodide
  - Ammonium hydroxide
  - Sodium sulphate
  - "Thymol blue" indicator

3.3 Treatment of the Sample

3.3.1 Pretreatment

- a) Dry in the stove at 120°C, increasing the temperature slowly and progressively, to avoid projections.
- b) Calcine at 550°C overnight to eliminate organic matter.

3.3.2 Wet Attack

- a) Weigh from 1 to 2 g of ash in 250 ml Pyrex receptacles.

b) Add 100 ml of HNO<sub>3</sub> concentrate and dry, occasionally adding drops of H<sub>2</sub>O<sub>2</sub>. Repeat this step until white ash is obtained.

d) Dissolve the final residue in HNO<sub>3</sub> 8N and cool to room temperature.

### 3.4 Radiochemical separation

#### 3.4.1 Preparation of the ion exchange resin columns.

##### COLUMN I

a) Fill a glass column of 1.1 cm diameter x 30 cm, with approximately 30 ml of AG1 x 8 moist resin (20-50 mesh). Wash the resin successively with HCl 1N, HNO<sub>3</sub> 8N and deionized water with a volume equivalent to 5-10 [times] the volume of the resin.

##### COLUMN II

b) Prepare an ion exchange column of 0.5 cm diameter x 10 cm, with 3 ml of AG1 x 8, 50-100 mesh resin. Wash the resin with 20 ml of HCl concentrate.

### 3.4.2 Ion Exchange and Elution

- a) Pass the nitric solution containing the sample through the Column I resin at a drip rate not to exceed 2 ml/min.
- b) Wash the column walls with 3-5 ml of HNO<sub>3</sub>, 8M. Repeat the washing 3 times.
- c) Pass 200 ml of HNO<sub>3</sub>, 8N through the column and discard the washing liquid.
- d) Wash the walls of the column with HCl concentrate.
- e) Pass 200 ml of HCl concentrate through the column and discard the washing liquid.
- f) Elute the plutonium with 100 ml of an acid solution of ammonium iodide in HCl (1.5 g of INH<sub>4</sub> dissolved in 5 ml of H<sub>2</sub>O and dilute to 100 ml with HCl concentrate). Catch the elution in a 250 ml receptacle.
- g) Add 5 ml of HNO<sub>3</sub> concentrate to the elution, stir and evaporate on a hot plate until dry, without bringing it to boiling.
- h) Dissolve the residue in 1-2 ml of HCl concentrate, and evaporate until dry. Repeat this step once.
- i) Wash the walls of the container with 3-5 ml of HCl concentrate, heat almost to boiling, transfer it to a clean centrifuge tube, washing the receptacle three times with 2 ml of HCl concentrate and unite all the washing liquids.

j) Add approximately 150 mg of sodium nitrite, mix, let sit for 15 minutes, centrifuge and transfer the floating matter to Column II.

k) Stop draining the sample through the column, wash the walls twice with 2-3 ml portions of HCl concentrate. Wash the column with 20 ml of HCl concentrate. Discard the washing liquid.

l) Elute the plutonium of the column with 20 ml of the ammonium iodide-HCl solution (prepared as in subparagraph f)).

m) Collect the elute in a 50 ml receptacle, add 2 ml of HNO<sub>3</sub> concentrate, mix, evaporate until dry, without bringing to a boil.

### 3.5 Electrodeposit

a) Add 1 ml of Na<sub>2</sub>SO<sub>4</sub> 0.3M to the residue

b) Evaporate until dry.

c) Add 0.3 ml of H<sub>2</sub>SO<sub>4</sub> concentrate.

d) Heat the receptacle, keeping it in motion, until the residue is completely dissolved. Heat only to the extent necessary; a large amount of the H<sub>2</sub>SO<sub>4</sub> should not be permitted to evaporate.

e) Add 4 ml of distilled water and two drops of 0.2% thymol blue.

f) Add drops of NH<sub>4</sub>OH concentrate until the color turns yellow-orange.

g) Transfer the solution to the electrodeposit cell and wash the remainder with a total of 5 ml 1% H<sub>2</sub>SO<sub>4</sub>.

h) Adjust the pH to 2.1-2.4 with drops of NH<sub>3</sub> concentrate, or if the final point is exceeded, with 20% H<sub>2</sub>SO<sub>4</sub>.

i) Connect a current of 1.2A for one hour. Add 1 ml of  $\text{NH}_3$  one minute before turning off the current.

j) Wash the planchet with a diluted ammonia solution (10 ml of  $\text{NH}_3$  in one liter of  $\text{H}_2\text{O}$ ).

k) Dry the planchet by heating it gently for a few minutes on a hot plate.

### 3.6 Measurement

#### 3.6.1 Equipment

The radioactivity of the electrodeposited Pu 239 + Pu 240 is measured by alpha spectrometry with silicon barrier semiconductor detectors Model 576 and 576A (ORTEC), with a nominal active area of  $300 \text{ mm}^2$ , a resolution of 21 Kev and a dead time of 0.5  $\mu\text{sec}$ . The operating power supply is 100 volts.

These detectors are connected to an ORTEC multichannel analyzer and to a DIGITAL professional 380 microcomputer.

#### 3.6.2 Preparation of the Standard Solution

The plutonium 236 has been furnished by the AERE-Harwell laboratories (Great Britain). The solution has been diluted and calibrated in the CIEMAT (Ionizing Radiation Metrology Division).

#### 3.6.3 Calibration in Efficiencies

A master specimen of known radioactivity is electrodeposited in the same geometry as the sample to be measured.

This master is measured in each detector. The ratio between between the disintegrations per minute of the master and the existing counts per minute is the efficiency.

#### 3.6.4 Determination of Baseline

The baseline in the spectral zone corresponding to the Pu-239 + Pu 240 is determined by measuring the counts of a "target" prepared with the same analytical process used for the samples.

#### 3.6.5 Calibration in Energies

Calibration in energies is accomplished using master specimens calibrated and prepared by the National Laboratory of Los Alamos (USA). The available sources are Am-241, Pu-238, Pu-239 and Pu-242.

#### 3.6.3 Measurement of the sample

- The planchet prepared in accordance with 3.5 is introduced into the counter equipment for measurement in accordance with Procedure No. of the PRYMA Institute.

- The counting time will never be less than 1440 minutes, depending upon the radioactivity of the sample.

### 3.7 Calculations

#### 3.7.1 Calculation of the Lower Limit of Detection (LLD) according to the Currie Criterion.

American standards ASTH (Volume 12, 01, 1983, C-100. For determination of plutonium 239 and plutonium 238 with a 95% confidence level, for which  $2 \times$  the square root of  $2K$  takes a value of

4.66, and considering a maximum of 10 counts of total baseline, the detection limit of the device is given by the formula:

$$LLD = \frac{4,66}{CE} \left( \frac{C_t}{T_t} \right)^{\frac{1}{2}}$$

where:

$C_t$  = Counts per second from baseline for radionuclide i.

$T_t$  = Counting time from baseline expressed in seconds.

CE = Count efficiency expressed in cps/Bq

### 3.7.2 Calculation of Chemical Yield

The chemical yield or recovery is the ratio between the radioactivity of the marker added and that found.

The calculation takes the following form:

$$Y = \frac{\text{mBq counted}}{\text{mBq added}}$$

### 3.7.3 Calculation of Minimum Detectable Radioactivity (MDR)

According to the ASTH standards, the minimum detectable concentration is calculated by dividing the value of the LLD by the chemical yield and the quantity of sample as follows:

$$AMD = \frac{4,66}{Y V CE} \left( \frac{C_t}{T_t} \right)^{\frac{1}{2}}$$

where:

Y = Chemical yield

V = Quantity of sample used in making the analysis (kilos or liters).

### 3.7.4 Calculation of Radioactivity

When, in the spectral zone corresponding to Pu-239 Pu-240 emission, a sufficient number of counts is obtained to consider (according to established criteria) that radioactivity exists, calculation is performed in the following manner:

$$A = A_{\text{marker}} \cdot \frac{C_{\text{Pu-239 + Pu-240}} - C_{B1}}{C_{\text{marker}} - C_{B2}} \cdot \frac{1}{V}$$

where:

$A_{\text{marker}}$  = Marker radioactivity added (Bq)

$C_{\text{Pu-239+Pu-240}}$  = Total counts due to peak Pu-239

$C_{B1}$  = Total counts of the Pu-239 spectral zone target

$C_{B2}$  = Total counts of the marker emission spectral zone target

### 3.7.5 Calculation of Uncertainties due to Counting

For expression of the results, the IAEA recommends calculating the statistical uncertainty as a function of unity:

$$\epsilon = \sqrt{\left( \frac{\sqrt{C_{\text{Pu-239}} + C_{B1}}}{C_{\text{Pu-239}} - C_{B1}} \right)^2 + \left( \frac{\sqrt{C_{\text{marker}} + C_{B2}}}{C_{\text{marker}} - C_{B2}} \right)^2}$$

### 3.8 Expression of Results

The final result of the sample's Pu 239 + Pu 240 alpha radioactivity is expressed as:

Liquid samples: Radioactivity/l

Tissue samples: Radioactivity/wet kg

The result per sample will be expressed in the following manners:

- Case a)  $A > \text{LLD}$

The sample thus has a true radioactivity value, the result being expressed as:

$A \pm 2\sigma A$

- Case b)  $\leq A \text{ LLD}$

The result will be expressed as LLD followed by its units.

## 4. RESPONSIBILITIES

It is the responsibility of the director of the Radiological and Environmental Protection office to:

- Approve the application of these procedures and direct such application when considered appropriate.
- Monitor the proper application of these procedures.

It is the responsibility of the chief of the Radiological Environment Operations Unit to:

- Keep these procedures updated.
- Designate the chief of the laboratory where these procedures are carried out.

- Monitor the proper application of these procedures.

It is the responsibility of the laboratory chief to:

- Supervise the correct application of the procedures by the analysts in his/her charge.

- Prepare updating of these procedures.
- Approve the results by signature.

It is the responsibility of the analysts to:

- Perform the procedures correctly.
- Request the information needed and deliver that obtained in timely fashion and the proper form.

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TITLE: CALIBRATION PROCEDURE FOR THE PHOSWICH SYSTEM FOR  
DETECTION OF ACTINIDES IN THE LUNGS

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PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

3 of 20

TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Detection Enclosure
  - 3.2 Detection System
    - 3.2.1 Phoswich Detectors
    - 3.2.2 Description (components and functions of the associated electronic chain)
4. CALIBRATIONS
  - 4.1 Calibration Equipment
    - 4.1.1 Calibration Sources
    - 4.1.2 Description of the Mannequin
  - 4.2 Calibration System
    - 4.2.1 Energy Calibration
    - 4.2.2 Efficiency Calibration
    - 4.2.3 Recording of Calibration Factors
    - 4.2.4 Calibration Frequency
5. OBTAINING THE PARAMETERS OF REFERENCE
  - 5.1 Determination and Control of Detector Resolution
  - 5.2 Verification of Energy Calibration
  - 5.3 Verification of Efficiency Calibration
  - 5.4 Environmental Background Determination and Control
6. QUALITY ASSURANCE
7. DETECTION THRESHOLD
  - 7.1 Established Criteria
8. QUALIFICATIONS
9. RESPONSIBILITIES
10. REFERENCES
11. APPENDICES

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

4 of 20

1. PURPOSE

To describe the methods used for calibration of the Phoswich system for detecting actinide photon emitters in the lungs.

2. SCOPE

The procedure applies to the Phoswich Corporal Radioactivity Counter (CRC), belonging to the CIEMAT Radiological Protection Unit for conducting in vivo measurements of radionuclides emitting photons of energy below 200 KeV.

3. COMPOSITION

3.1 CRC Detection Enclosure

The detection enclosure used (Appendix I) is a shielded chamber having the following inside dimensions: 243 cm square and 197 cm high and of low floor, constructed of the following materials:

- Steel plate 130 mm thick (prenuclear quality steel)
- Lead plate 5 mm thick
- Cadmium plate 0.7mm thick
- Copper plate 0.38 mm thick

The chamber is in overpressure relative to the area where it is located. It has a system of forced ventilation through absolute filters, which provides 30 renewals/hour under normal operating conditions and 60 renewals/hour for cleaning operations.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

5 of 20

### 3.2 Detection System

The detection system is comprised of the following components:

#### 3.2.1 Phoswich Detectors

The PHOSWICH (Phosphor Sandwich) detector (Appendix II) consists of two flashing crystals connected optically to a single photomultiplier tube which catches the light from both, producing a single pulse.

The combination of crystals used in the C.R.C. is NaI(Tl)-CsI(Tl). The INa(Tl) crystal is a 0.2 cm flasher with a fluorescence decay period of 0.25  $\mu$ sec, which acts as a detector crystal. The CsI(Tl) crystal is a 5.8 cm thick flasher with a 1.1  $\mu$ sec decay period, which is used in anticoincidence to eliminate noise due to the Compton effect.

This system is used to detect low energy X rays and  $\gamma$  rays, when we want to suppress the continuous Compton effect which is produced in that energy zone. This suppression is achieved through the use of coincidence techniques, which permit discrimination of the pulses produced in each crystal, and in particular, eliminate those events which have deposited all their energy in the first crystal and have an interaction with the second crystal.

#### 3.2.2 Description (Components and Functions) of the Associated Electronic Chain

The low energy emitter detection system, which is diagramed in Appendix III, consists of:

- 2 Harshaw Type 20MBHS2M/5B-CX Phoswich detectors, 12.7 cm in diameter, connected optically to RCA-S83006E type photomultiplier tubes; serial numbers CT-122 and DO-240.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

6 of 20

- 2 ORTEC high tension sources, Models 456 and 556, which supply voltages varying between 50 and 3000 V and currents of from 0 to 10 mA.

- 2 CANBERRA Model 2005 preamplifiers. These are high input and low output impedance adapters, which permit the passage of a voltage impulse from each of the detectors to a linear amplifier without producing non-linear effects in its amplitude.

- 1 CANBERRA Model 1456A addition amplifier. This electronic module, connected to the output of the preamplifiers, adds up to 4 independent unipolar or bipolar signals; it includes gain control over one input and admits voltage impulses of between 0 and  $\pm 10V$ .

- 1 ORTEC Model 460 pulse amplifier. It accepts input pulses for any polarity from the addition amplifier and expands their amplitude by means of an adjustment in the gain factor within the range of 3 to 1000. Its function is to provide a delay line for all outgoing pulses.

- 1 ORTEC Model 427A pulse retarder. The pulse amplitude retarder module can delay a linear or logic signal from 0 to 4.75  $\mu\text{sec}$ , in increments of 0.25  $\mu\text{sec}$ . The output pulse is delayed 3.25  $\mu\text{sec}$ , thus causing the pulse carrying the energy information to arrive at the linear port in timely fashion.

- 1 ORTEC Model 458 pulse form analyzer. This module measures the drop time of the input pulse coming from the amplifier and generates, as an output, a linear pulse with an amplitude proportional to the drop time. It also serves as a single channel analyzer which permits generation of logic pulses, based on the drop time of the input pulse. One of these pulses is produced when the drop time is within a preselected range. The output pulse width is 0.4  $\mu\text{sec}$ , and is delayed 3  $\mu\text{sec}$  from the input signal.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

7 of 20

- 1 ORTEC Model 550 single channel analyzer. This module produces a logic output pulse for any type of unipolar or bipolar input which exceeds a preselected voltage in the discrimination window. The basic function of this module is to determine whether the amplitude of an arbitrary input signal (in our case, bipolar) is greater than the discrimination level (energy range of interest < 200 KeV).
- 1 LASL brand pulse shaper. An electronic module which admits logic pulse inputs, producing a 3.2 V amplitude output pulse, the duration of which is regulated by a 30-160  $\mu$ sec variable potentiometer. It extends the duration of the pulse, so that the signal coming from the single channel analyzer coincides, time-wise, with the signal coming from the pulse shape analyzer.
- 1 ORTEC Model 418A coincidence module. The coincidence module permits analysis of 5 inputs with three possible operating modes: Coincidence, Anticoincidence and Off. It is essentially an input pulse adder and level discriminator, producing an output pulse when the permitted inputs occur within the preselected resolution time. The input resolution time is determined by the width of the pulses.
- 1 ORTEC Model 426 linear port. An electronic module which is used to inhibit a signal in accordance with some pre-specified conditions. The signal duration is variable between 0.3 and 4  $\mu$ sec. Its mission is to self-activate on arrival of a positive logic signal, and permit output of the pulse at the preselected time making it optimum for subsequent use.
- 2 CANBERRA Model 8075 analog-digital converters. An electronic module which converts the amplitude of the impulses analyzed into numerals proportional to that amplitude.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

8 of 20

. 1 CANBERRA Series 35 Plus, Model 3503 multichannel analyzer, with 8192 channels. It performs microprocessor analyses such as area calculations, peak searches, normalization, integration, data blending and energy calibration.

#### 4. CALIBRATIONS

This is the entirety of operations and calculations directed at ensuring that the information obtained in each measurement can be interpreted in terms which permit identification and quantification of the radioactivity of a given radionuclide deposited in different organs or tissues.

Calibration is detection system-specific and, therefore, might be changed replacement, or behavioral modification of any of the elements of the measurement equipment.

Calibration for low energies (<200 KeV) is conditioned by the fact that the response of the detector is highly contingent on its distance from the source, as well as on the absorption characteristics of any materials between them. This makes it necessary to calibrate the equipment separately for different thoracic thicknesses and composition percentages of muscle and fat. It is also necessary to perform calibrations for material deposited in the lung, as well as in the lymph nodes, as a function of the chronological characteristics of the contamination.

##### 4.1 Calibration Equipment

##### 4.1.1. Calibration Sources

. The precise sources of reference used as a quality control to verify system stability are:

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

9 of 20

Radio-nuclide	Ref No.	R (Mev)	Radio-activity (Bq10E4)	Calibra-tion date
<sup>241</sup> Am	70U58	0.018 0.060	3.99	01.09.85
<sup>57</sup> Co	7T453	0.122	4.43	01.09.85

The active organs (lungs) used for efficiency calibration are:

Organ	Radionuclide	Radioactivity (Bq*10E4)	Reference
Lungs	<sup>241</sup> Am	2.15	630(R-L)
"	<sup>239</sup> Pu	7.01	632(R-L)
Lymph Nodes	"	10.36	543 L
"	"	2.85	543 S1
Lungs	"	2.85	543 S2
"	U-Nat.	1.18	661(R-L)
"	U-Nat (enr.3%)	0.05	663(R-L)

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

10 of 20

4.1.2 Description of the Mannequin

The mannequin utilized was designed by the Lawrence Livermore National Laboratory (LLN), (Appendix IV). It is a modular mannequin, having characteristics identical with those of the human master specimen defined by the ICRP. It comprises a skeleton of material equivalent to bone, removable organs and a series of thoracic plates (layers B and C) equivalent to tissue with different muscle/fat ratios (see accompanying table) which can be placed upon the torso to simulate different thoracic thicknesses. There are inert organs with recesses into which tubes of known radioactivity can be inserted for calibration and lungs contaminated with AM-241, Pu-239, Natural Uranium and slightly (3%) enriched natural uranium.

Thicknesses (cm) $\pm$ 0.2	Ref. Layers
1.67	Core
2.57	B150-1 C137-1
3.27	B150-2 C137-2
3.57	B150-3 C137-3
4.27	B150-4 C137-4

Core: Intrinsic LLN mannequin

B150 Layers: Polyurethane with 2.1% CaCO<sub>3</sub>. Material equivalent to 50% muscle and 50% fat.

C137 Layers: Polyurethane with 4.3% CaCO<sub>3</sub>. Material equivalent to 100% muscle.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

11 of 20

4.2 Calibration System

4.2.1 Energy Calibration

Establishes a relationship between the pulse height scale of the multichannel analyzer (MCA) and the energy of the photons emitted by a given source.

The following are used for energy calibration of the Phoswich detectors:

- a) Precise Am241 sources referring to an X ray of 17.8 KeV and a  $\gamma$  emission of 59.5 KeV, and Co57 sources referring to a  $\gamma$  emission of 122.1 keV.
- b) A perfectly reproducible geometry consisting of placing the sources at the base of a plexiglass trapezoid equidistant from each detector. The detectors are each placed into orifices cut into the trapezoid.
- c) The response must be linear, acting upon the zero and the gain until a distribution of 0.5 KeV per channel, and the correct position for the  $\gamma$  emission of 122.1 keV is obtained.
- d) A minimum of 5000 counts are accumulated in the peak channel, in order for the photopeaks to appear well defined and to be able thus to calculate with the greatest possible precision, the centroid of the photopeak of interest.

4.2.2 Efficiency Calibration

This is radionuclide-specific and comprises the following operations:

- a) Obtain the parameters of reference, as indicated in section 5.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

12 of 20

- Record the resolution of each of the detectors with a precise Am-241 source

- Verify the energy calibration by recording the net recount for the  $\gamma$  emission of a precise Am-241 source, in the region of interest defined for this radionuclide

- Obtain the environmental background without the mannequin for the region of interest defined for this radionuclide.

b) With the mannequin in the measurement position (\*), charged with the specific radionuclide and maximum filtration, define the region of interest (ROI) characteristic of the photopeak selected. The criterion for defining the ROI is to take an area surrounding the peak channel until the gaussian extreme becomes confused with the background.

(\*) The Measurement geometry for calibration of pulmonary detection is the supine decubitus position, with a detector in contact over each hemithorax.

c) With the mannequin in the measurement position, and for each thoracic composition and thickness, obtain a count with the lungs inert and another count with the lungs radioactive, for the ROIs defined.

d) Calculate the net counts in the selected ROI for each thoracic thickness and composition. The net counts are obtained as the difference between the total counts in the contaminated mannequin and in the inert mannequin.

e) Calculate the efficiency for the different thoracic thicknesses and compositions, as the quotient between the net counts of the photopeak of a given energy and the number of photons of that energy emitted by the source.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

13 of 20

For thoracic thicknesses and compositions differing from those calibrated, efficiency is calculated by interpolation from the experimental values obtained.

f) Calculation of thickness/efficiency curves for each thoracic composition.

4.2.3 Calibration Factor Records

The calibration parameters are recorded in a specific and referenced document. The technical documentation of all evaluations will contain a certification of the calibration applied. The parameters of reference obtained during the calibration process will be maintained in the the system's quality assurance books, always reflecting the deviations between the true and reference values.

4.2.4 Calibration Frequency

Primary recalibrations will be performed whenever the quality assurance program reflects a substantial modification in the system's stability.

Efficiency calibration should be valid for periods of at least three years, provided no essential system component is replaced.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

14 of 20

5. OBTAINING THE REFERENCE PARAMETERS

Prior to performing the comprehensive efficiency calibration, a series of the detection system's behavioral characteristics are evaluated. These will be utilized as a reference for monitoring the system's stability, and therefore of the validity of the calibration.

5.1 Detector Resolution Determination and Control

Detector Resolution Determination consists of:

1. Using a precise Am241 source positioned at the base of a plexiglass trapezoid equidistant from each detector.

2. Measuring for sufficient time to obtain at least 5000 counts in the photopeak channel so as to obtain statistical acceptability.

3. Calculate the resolution, using the equation:

$$R = \frac{\text{FWHM}}{E_0} \times 100$$

where:

FWHM = Width at half the maximum height of the photopeak (expressed in energy).

$E_0$  = The energy of the photopeak (in this case, 59.5 KeV)

4. The resolution of Detector No. 1 is calculated by suppressing the signal from Detector No. 2.

5. The resolution of Detector No. 1 is calculated by suppressing the signal from Detector No. 1

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

15 of 20

6. The resolution of the whole is accomplished with the sum of the two detectors.

7. Record the values obtained (Appendix V).

8. These operations will be conducted monthly.

5.2 Verification of Energy Calibration

1. Take as a reference, a precise Am 241 source referred to the X ray of 17.8 KeV and the  $\gamma$  emission of 59.5 KeV.

2. Use a constant geometry. The source will be placed at the base of a plexiglass trapezoid equidistant from each detector.

3. Measure for sufficient time to obtain at least 5000 counts in the photopeak channel, so as to obtain statistical acceptability.

4. Take into account the energy calibration and verify that the position of the centroids and the distance in channels referred to the 17.8 KeV and 59.5 KeV are kept within the levels defined in Section 6.

5. Record the values obtained (Appendix VI).

6. These operations will be conducted daily.

5.3 Verification of the Efficiency Calibration

1. Take as a reference a precise Am241 source referred to the  $\gamma$  emission of 59.5 KeV.

2. Use a constant geometry. The source will be placed at the base of a plexiglass trapezoid equidistant from each detector.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

16 of 20

3. Measure for sufficient time to obtain at least 5000 counts in the photopeak channel, so as to obtain statistical acceptability.

4. Determine the net photopeak count in the region of interest defined for this radionuclide.

5. Record the values obtained (Appendix VII)

6. This control will be conducted daily.

5.4 Environmental Background Determination and Control

1. Daily determination of the environmental background inside the chamber with reproducible geometry.

2. Counting time of 54,000 seconds, so as to obtain statistical acceptability.

3. Record the background daily in the ROIs defined for the contaminants which it is sought to evaluate.

4. Compare the daily backgrounds with the mean monthly background.

6. QUALITY ASSURANCE

During calibration, a series of parameters are obtained, which will be used later to verify the validity of the calibration in use, and therefore, to guarantee system stability and the quality of the results.

Any time an evaluation is to be made during a work session, the corresponding verifications of environmental background, energies and efficiencies described in Section 5 will be made.

The validity of the calibration will be verified by comparing the results of these determinations with the parameters of reference.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

17 of 20

- A resolution variation of more than 30% compared to the resolution of reference may lead to recalibration of the system, or failing that, application of a correction factor.
- Any slippage (>5%) of the energies observed upon verification of the energy calibration, will be corrected immediately by adjusting the zero and the gain.
- A significant (>5%) variation in the efficiency may lead to recalibration of the system, or failing that, application of a correction factor. The mean results of the monthly values obtained in the efficiency checks will also be recorded, in order to permit tendency analysis.
- A significant (>5%) increase of the environmental background in the regions of interest established for any radionuclide, will lead to system review.

7. DETECTION THRESHOLD

7.1 Established Criteria

The minimum detectable radioactivity (MDR) describes the detection equipment's capability to distinguish between counts corresponding to contaminated and non-contaminated persons. The MDR is established to be:

$$\text{MDR} = \frac{2.714.64 S_b}{E_r * T_c}$$

where:

- $S_b$  = Standard deviation of background
- $E_r$  = Efficiency
- $T_c$  = Counting time

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

18 of 20

As the first term is negligible compared to the second, this equation is actually equivalent to 4.64 times the standard deviation of the background, expressed in terms of dose, in accordance with ANSI N13.30.

In any case, but very especially in the Phoswich detection system for low energy emitters, the background counts, the efficiency values, and hence, the MDRs, depend heavily upon the individual's physical constitution.

The dependency of the efficiencies calculated with regard to individual physical constitution is significant in the detection of low energy emitters in the lungs, due to the strong absorption this type of radiation undergoes in passing through the thorax.

#### 8. QUALIFICATIONS

The individual responsible for the Corporal Radioactivity Counter is the person designated by the Dosimetry Service who will see to the proper technical maintenance of the equipment assigned to this detection system and for the correct application of these procedures.

#### 9. RESPONSIBILITIES

It is the responsibility of the Chief of the UOPRI to:

- Review the updates to these procedures, and direct them when considered appropriate.
- Ensure the correct application of these procedures.

PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

19 of 20

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PHOSWICH DETECTION SYSTEM  
CALIBRATION PROCEDURE

0

SPECIFIC PR-X7-08

11-29-91

20 of 20

11. APPENDICES

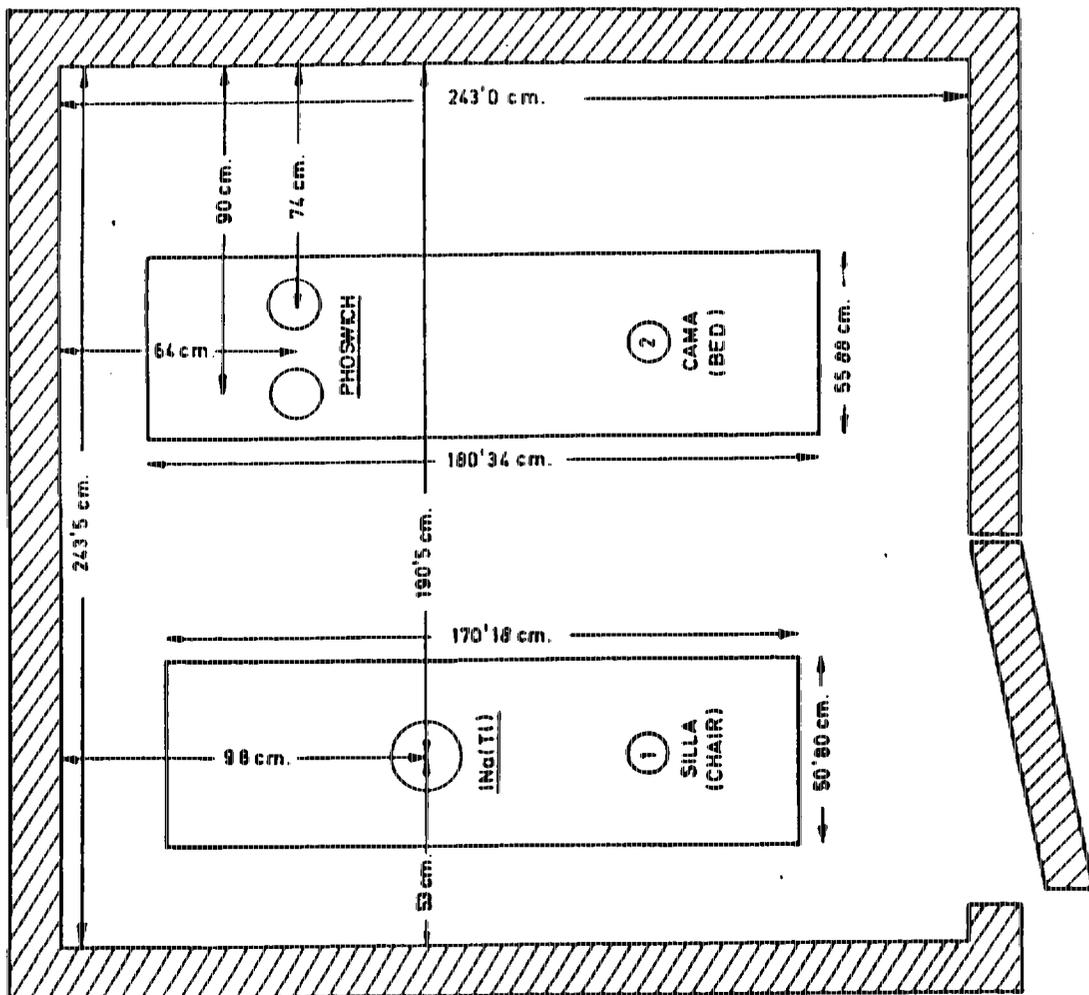
- APPENDIX I CRC Detection Enclosure Plan
- APPENDIX II Phoswich Detector Diagram
- APPENDIX III Schematic Diagram of the Phoswich Low Energy Detection System
- APPENDIX IV Diagram of the LLN Mannequin
- APPENDIX V Obtaining the Parameters of Reference. Resolution.
- APPENDIX VI Obtaining the Parameters of Reference. Verification of Energy Calibration.
- APPENDIX VII Obtaining the Parameters of Reference. Verification of Efficiency Calibration.

APPENDIX I  
CRC DETECTION ENCLOSURE PLAN

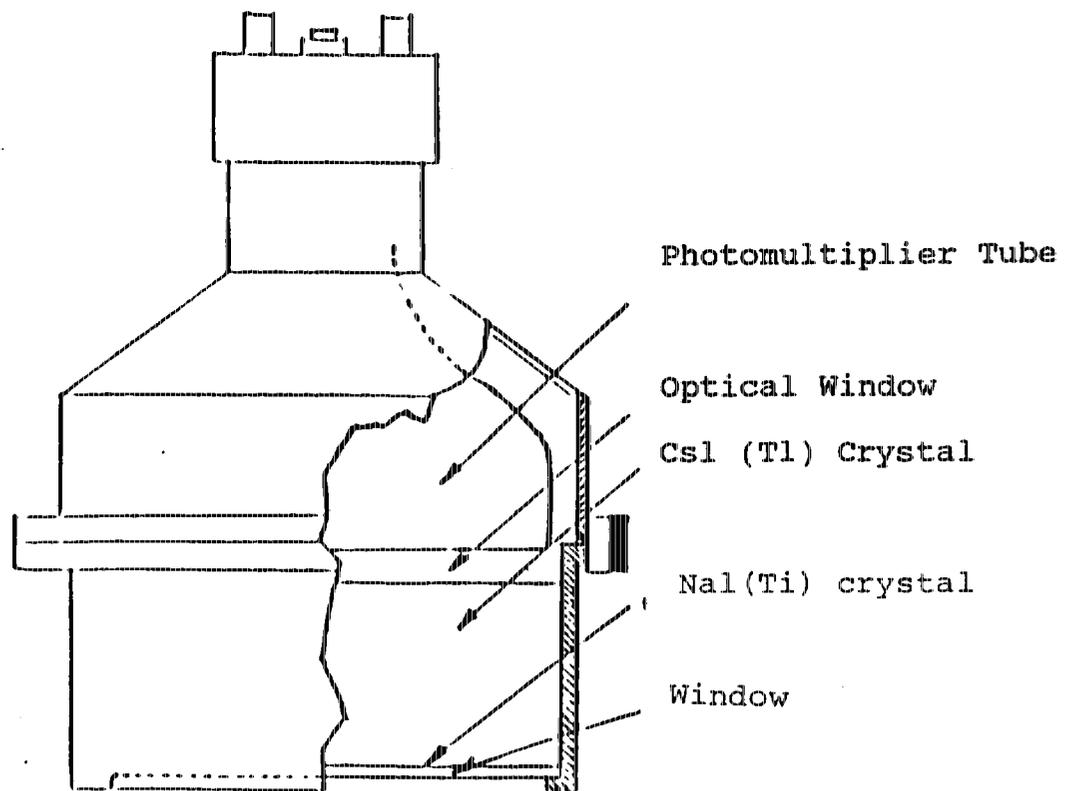
## SHIELDING ROOM

SHIELD FROM OUTSIDE TO INSIDE :

13 cm.	IRON
05 cm.	LEAD
01 cm.	CADMIUM
01 cm.	COPPER



APPENDIX II  
PHOSWICH DETECTOR DIAGRAM

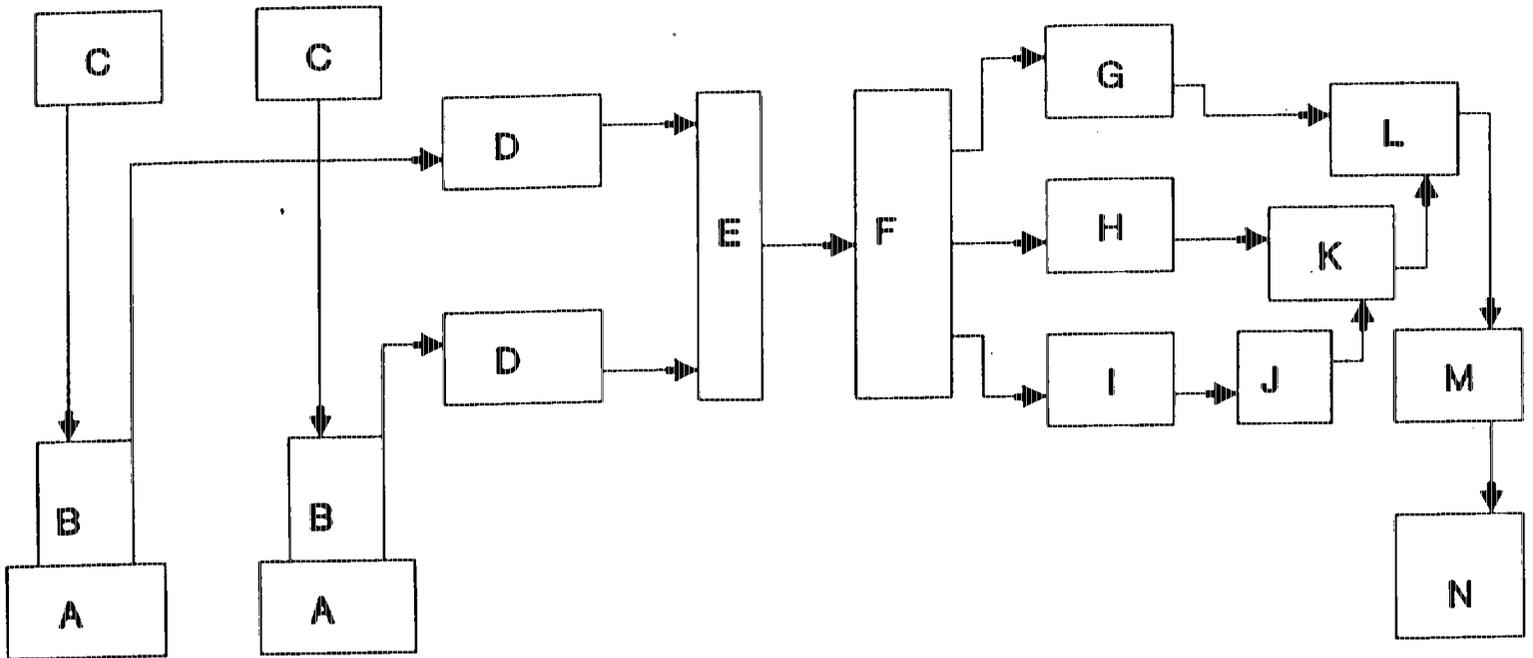


"Phoswich" Detector Diagram

APPENDIX III

SCHEMATIC DIAGRAM OF PHOSWICH LOW ENERGY DETECTION SYSTEM

SCHEMATIC DIAGRAM OF PHOSWICH LOW ENERGY DETECTION SYSTEM



A - Harshaw Type 20MBHS2M/5B-CX  
Phoswich detectors

I - ORTEC 550 single channel

B - RCA-S83006E type photomulti-  
pliers

analyzer

C - ORTEC 456,556 High Voltage

J - LASL pulse shaper

D - CANBERRA 2005 preamplifiers

K - ORTEC 418A Coincidence  
module

E - CANBERRA 1465A adding ampli-  
fier

L - ORTEC 426 linear port

F - ORTEC 460 pulse amplifier

M - CANBERRA 8075 A-D con-  
verter

G - ORTEC 427A pulse delayer

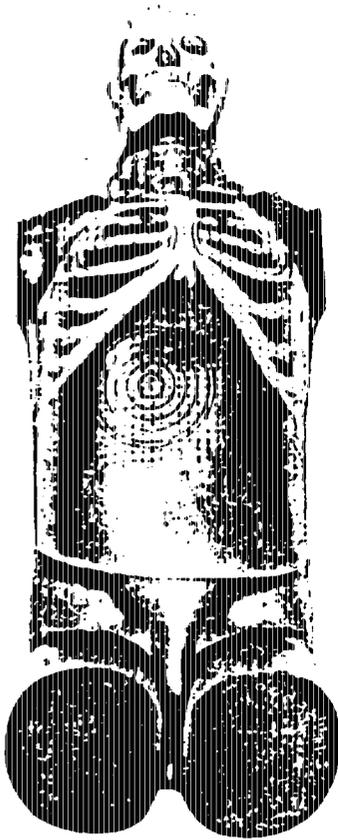
N - CANBERRA 35 Plus multi-  
channel analyzer

H - ORTEC 458 pulse shape  
analyzer

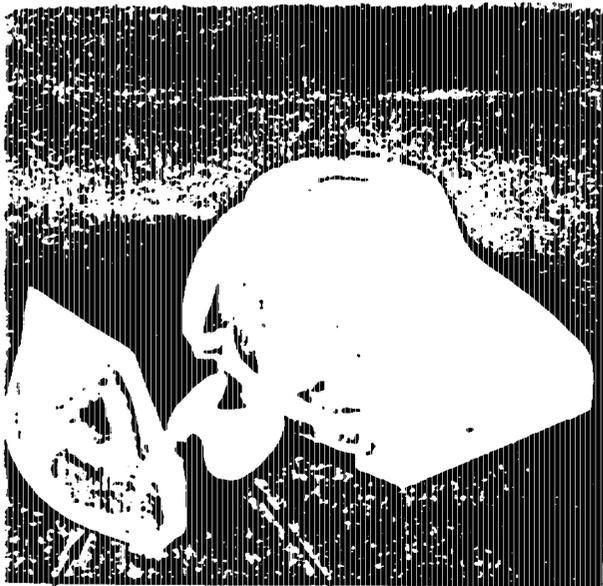
APPENDIX IV

DIAGRAM OF THE LLN MANNEQUIN

# THE FISSION-PRODUCT PHANTOM



FISSION-PRODUCT PHANTOM



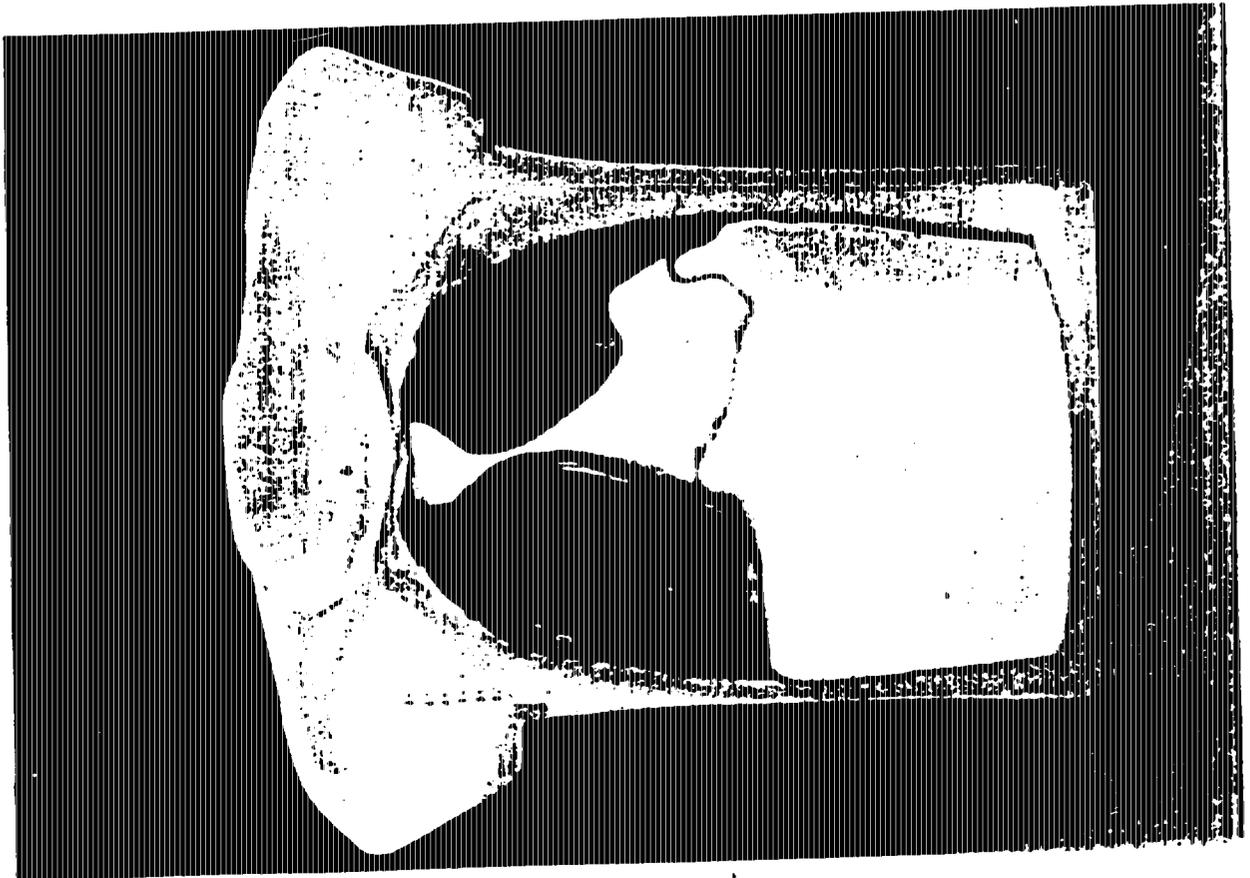
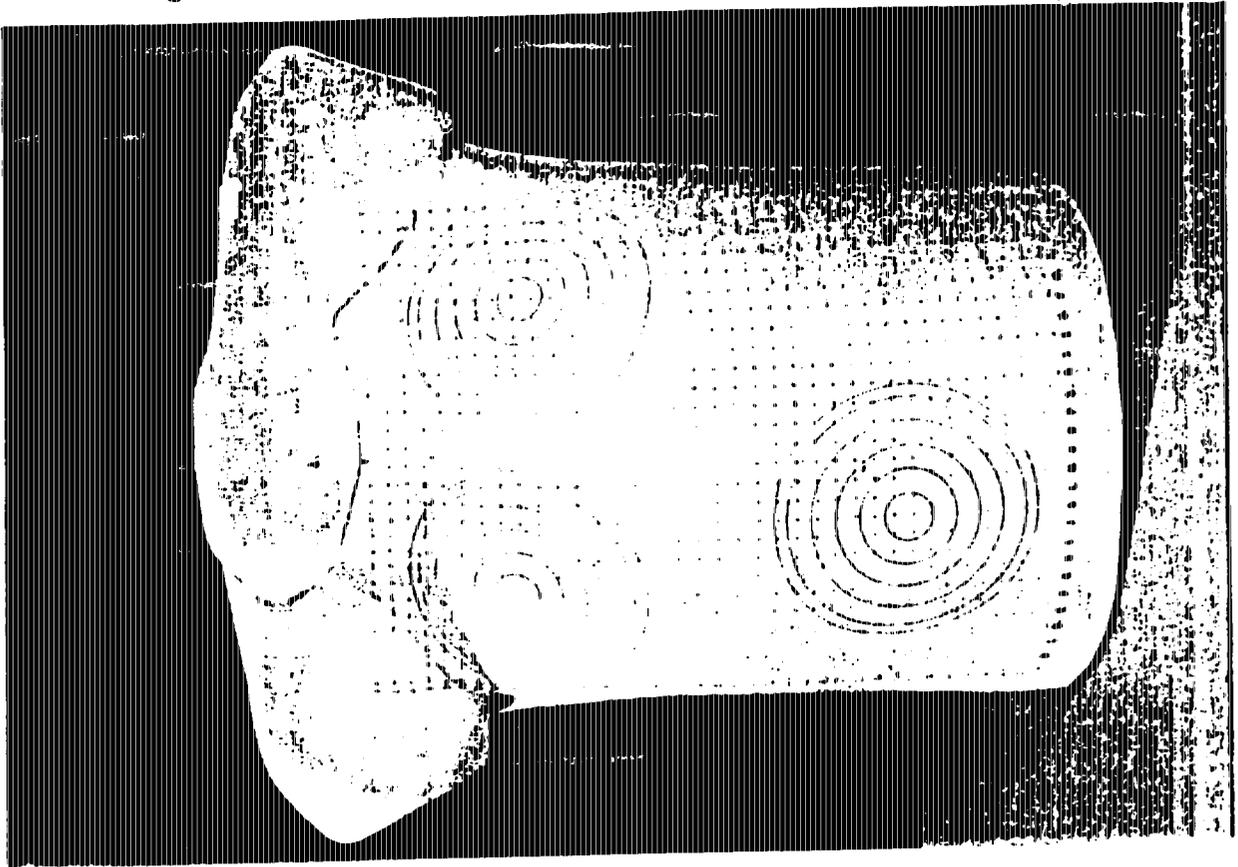
THYROID

— ANTERIOR TISSUES

THYROID PHANTOM  
WITHOUT HEAD, SHOWING  
INNER ASSEMBLY



THYROID PHANTOM  
WITH HEAD



APPENDIX V

OBTAINING THE RESOLUTION REFERENCE PARAMETERS

MONITORING OF CRC OPERATING CONDITIONS

RESOLUTION OF PHOSWICH DETECTORS

DETECTOR IDENTIFICATION	NOMINAL RE-SOLUTION %	CALIBRATION RESOLUTION R1%
-------------------------	-----------------------	----------------------------

No. 1 CT-122	16.32	
--------------	-------	--

No. 2 DQ-240	12.9	
--------------	------	--

No (1+2)

Calibration date:    /    /

AM-241 59.9 keV Photopeak of Reference

No of counts, peak channel  $\approx$  5000

DATE	DETECTOR 1		DETECTOR 2		DETECTORS R1+R2	
	R2%	R2/R1%	R2%	R2/R1%	R2%	R2/R1%
	RESOL.		RESOL.		RESOL.	

APPENDIX VI  
OBTAINING PARAMETERS OF REFERENCE  
VERIFICATION OF ENERGY CALIBRATION



APPENDIX VII  
OBTAINING THE PARAMETERS OF REFERENCE  
VERIFICATION OF EFFICIENCY CALIBRATION



JUNTA DE ENERGIA NUCLEAR ["NUCLEAR ENERGY COUNCIL"] PAGES 14  
OFFICE: RADIOLOGICAL AND ENVIRONMENTAL PROTECTION REVISION 0  
DIVISION: RADIOLOGICAL ENVIRONMENT O.U. Date 6/9/86  
SPECIFIC PROCEDURE No. MA/01 Comments\_\_\_ Approval\_\_\_ Execution\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF Pu-239 IN URINE SAMPLES  
BY ALPHA SPECTROMETRY

X CONTROLLED COPY No. 10 ASSIGNED TO MS EMMA IRANZO MARTIN  
\_\_\_\_ NON-CONTROLLED COPY

Revision No.	0	1	2	3	4
Prepared by	Asunción Espinosa				
Date	6-9-86				
6-9-86	/signature/				
Reviewed by	E. Iranzo Gonzalez				
Date	6-20-86				
9-19-86	/signature				
Supervised for	F. Alamillos				
Quality Assurance					
Date	10-20-86				
10-20-86	/signature/				
Approved by	F. Mingot				
Date	[illegible]-1987				
10-22-1986					

Form P/01

JEN DOCUMENT No. \_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
REV\_0 Date 6-9-86 Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

Page 1 of 14

## TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Collection of the Sample
  - 3.2 Preparation of the Sample
  - 3.3 Wet Incineration and Dissolution
  - 3.4 Purification by Ion Exchange
  - 3.5 Elution of Plutonium
  - 3.6 Electrodeposit of Plutonium
  - 3.7 Measurement
    - 3.7.1 Equipment
    - 3.7.2 Efficiency Calibration. Preparation of Standard
    - 3.7.3 Measurement of Sample
  - 3.8 Calculations
    - 3.8.1 Calculation of Lower Limit of Detection
    - 3.8.2 Calculation of Minimum Detectable Radioactivity
    - 3.8.2 Calculation of Errors
    - 3.8.4 Calculation of Recovery
  - 3.9 Expression of results
4. RESPONSIBILITIES
5. REFERENCES, REAGENTS AND MATERIAL
  - 5.1 References
  - 5.2 Reagents and Material
6. ENCLOSURES

JEN DOCUMENT No. \_\_\_\_  
REV\_\_0\_\_ Date 6-9-86

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

Page 2 of 14

1. PURPOSE

To describe the method used in the laboratories of the Radiological Environment O.U. of the Radiological and Environmental Protection office, for determination of Pu-239 in urine samples.

2. SCOPE

The method is applied to 24-hour urine samples of possibly contaminated persons coming from internal contamination monitoring programs.

### 3. COMPOSITION

#### 3.1 Collection of the Sample

Sample A: Twenty-four hour urine collected in liter bottles slightly acidulated with nitric acid. No micturition must be lost, and the standards given in the cited analysis reference must be scrupulously adhered to in order to eliminate contamination.

Sample B: Urine from one micturition for creatinine analysis.

#### 3.2 Preparation of the Sample

- Measure the volume and the pH of the urine.
- Transfer the urine into a 2 liter graduate.
- Add 40°BE concentrated (63%) nitric acid until it reaches 1N.
- Add prepared and stabilized Pu-242 standard solution over nitric acid 8N (approximately 4 dpm).
- Place the graduate with the urine in a water bath.
- Heat, with magnetic agitation for 30 minutes, keeping the water temperature between 70 and 80°C during the agitation time.
- Add 1 ml of phosphoric (orthophosphoric) acid to each urine sample.
- Add 24°BE concentrated (35.05%) ammonium hydroxide until a voluminous precipitate is formed, acidify the urine, verifying its acidity with universal litmus paper.
- Add 0.1 ml of a saturated solution of calcium nitrate (to correct any lack of calcium in the urine) and again add ammonium hydroxide until a precipitate forms.
- Agitate the sample for one hour, allow to settle for approximately 30 minutes, verifying that the precipitation is complete by adding ammonium hydroxide; if it is not, add 25 ml of ammonium hydroxide and agitate for another 30 minutes.
- The precipitation process will not last under 1 hr, 30 minutes.

- Remove the graduate from the water bath, extract the stirring rod and cover with a watch crystal.
- Allow the precipitate to settle overnight at room temperature.
- Decant and discard the effluent.
- Transfer the precipitate to a 90 cc centrifuge tube, by flushing it with distilled water.
- Centrifuge for a minimum of 15 minutes until a precipitate firmly adhering to the bottom of the tube is obtained.
- Discard the flushing liquid
- Wash the graduate twice with 10-15 cc of nitric acid 2N and add the acid to the the centrifuge tube, mixing it with the precipitate.
- Continue the washing of the graduate two or three times and mix it with the precipitate.
- Add 2 or 3 drops of octyl alcohol before proceeding to evaporation.
- Place the centrifuge tube containing the solution of the precipitate on an aluminum block and evaporate dry overnight at 95-100°C.

### 3.3 Wet Incineration and Dissolution

3.3.1 • Incinerate the residue from the evaporations in the same tube, with concentrated nitric acid at 300°C on an aluminum heating block, until white ashes are produced (the nitric acid will be added un 2-3 cc increments, cooling before each addition).

3.3.2 Allow to cool at room temperature.

3.3.3 Add 40-50 ml of nitric acid 8N to dissolve the ashes. Leave at room temperature overnight to enhance the dissolution.

### 3.4 Purification by Ion Exchange

#### 3.4.1 Preparation of the anionic resin column

- Use the glass column in accordance with the enclosure.
- Place spun glass in the tube of the column, moistening it with distilled water.
- Fill the tube of the column with Dowex A.G. 1X2, 50-100 mesh anionic resin, suspended in distilled water.
- Wash the resin with at least five portions (VC) of nitric acid 8N until the washing liquid does not give a chloride reaction with silver nitrate.
- Let the nitric acid pass completely through before going to the next step.

#### 3.4.2 • Pass the ash solution contained in the centrifuge tube through the resin column at a flow rate of 1-2 ml per minute.

- Wash the centrifuge tube three times with 10 ml of nitric acid 8N each time, allowing it to pass through completely before each washing. (Any AMERICIUM and URANIUM the sample may contain will be in the liquid coming from the passage of the sample solution through the column, plus the two nitric acid washings. These liquids may be saved for subsequent radiochemical analyses.)
- Slowly add 3 ml of concentrated hydrochloric acid to the column. (Any Th the sample may contain will be in these liquids)
- Allow the acid to pass completely through.

### 3.5 Plutonium Elution

- Add hydroxylamine hydrochlorate (solid product, approximately 0.25 g) to the resin.
- Elute with hydrochloric acid 0.5n (two portions of 5 ml each time), catching the elution in a 25 ml receptacle.
- Dry the elution by evaporation under an infrared lamp.

### 3.6 Plutonium Electrodeposit

- Add 1 ml of concentrated hydrochloric acid to the evaporation residue of the elution.
- Dry by evaporation under an infrared lamp.
- Add 1 ml of hydrochloric acid 1N and 3 ml of 4% ammonium oxalate.
- Heat gently under an infrared lamp for approximately one minute.
- Transfer the solution to the electrolytic cell, washing the receptacle several times with distilled water, which is added to the cell until 10 ml has been completed.
- Electrodeposit at 200 mA for 3 hours.
- At the end of 3 hours, without disconnecting the current, add 2 ml of 50% (v/v) ammonium hydroxide to bring the electrolyte to pH 7.5, verifying it with litmus paper.
- Remove the cell from the stand, disconnecting the current previously.
- Wash the cell with distilled water.
- Remove the planchet from the cell and wash it with distilled water.
- Allow the planchet to dry and flame it.

#### 3.7.1 Equipment

The radioactivity of the electrodeposited Pu-239+Pu-240 is measured by means of alpha spectrometry, with ORTEC Model TR-21-300-100 silicon barrier semiconductor detectors having a nominal active area of 300 mm<sup>2</sup>, a resolution of 21 KeV and a dead time of 0.5 S. The operating power supply is 100 volts.

These detectors are connected to an ORTEC multichannel analyzer and a DIGITAL PDP-11/23 microcomputer with the RT-11 operating system.

3.7.2 Efficiency Calibration. Preparation of Standard

The plutonium-242 solution is obtained from Oak Ridge National Laboratory, with its respective certificates.

- The solution to mark the samples is prepared in nitric acid 8N, by heating at 80°C for 2 hours to ensure that the plutonium is stabilized in valence state IV.
- The solution thus prepared is diluted in nitric acid 8N until a radioactivity of approximately 8.8 dpm (147 mBq) is obtained.
- The standard solution is evaluated by means of an alpha count in a continuous gas flow counter, verifying the measurement with other alpha spectrometry.
- The spectrometer is calibrated, and baselines determined according to procedure no. \_\_\_\_\_ of the Radiological and Environmental Protection office.
- The counting time will never be less than 2880 minutes.

3.8 Calculations3.8.1 Calculation of the Lower Limit of Detection (LLD)

• According to the Currie Criterion. The American standards ASTH (Volume 12, 01, 1983, C-100) for determination of plutonium 239 and plutonium 238 with a 95% confidence level, for which 2 x the square root of 2K takes a value of 4.66, and considering a maximum of 10 counts of total baseline, the detection limit of the device is given by the formula:

$$LLD = \frac{4,66}{2,2 E} \left( \frac{C_f}{\tau_f} \right)^{\frac{1}{2}}$$

JEN DOCUMENT No. \_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0 Date 6-9-86

Page 8 of 14

$C_b$  = Counts per minute from baseline for isotope i.

$T_b$  = Counting time from baseline expressed in minutes.

CE = Count efficiency

With an efficiency of 30%, a counting time of 1440 minutes and a baseline of 10 counts. The lower limit of detection will be:

$$LLD = 0.29 \text{ mBq} = 0.008 \text{ pCi}$$

### 3.8.2 Calculation of Minimum Detectable Radioactivity (MDR)

This calculation requires a sufficient recount (at least 50 total counts) which will permit a close enough approximation of the Poisson distribution to that of Gauss, for us to use Gaussian statistics.

• The Currie criterion for a small number of events is:

$$MDR = \frac{2.71 + 4.66 \sqrt{C_b}}{T \times 2.22 \times E \times R}$$

where:

T = Counting time

$C_b$  = Total counts

R = Recovery

Therefore, for

R = 70%

E = 30%

We obtain

$$MDR = \frac{2.71 + 4.66 \sqrt{50}}{4320 \times 2.22 \times 0.3 \times 0.7} = 0.629 \text{ mBq/sample}$$

(0.017 pCi/sample)

## 3.8.3 Calculation of Errors

3.8.3.1 The alpha spectrometer calibration error is given by the errors of the secondary standards utilized and certified by the N.B.S.

The variance of the different calibration measurements is sufficiently low to permit reproduction of the measurement in 98.6% of the cases.

$$\begin{aligned} \text{LLD} &= 0.296 \pm 0.0034 \text{ mBq} \\ \text{LLD} &= 0.008 \pm 0.000092 \text{ pCi} \end{aligned}$$

which involves an error of 1.15% with 95% confidence.

3.8.3.2 The error of the minimum detectable radioactivity will be formed by the accumulation of the following random errors:

- a) Spectrometer calibration error which is considered random.
- b) Radiochemical analysis error.
- c) Micropipette error.
- d) Sample quartering error.
- e) Counting error.

- Calibration error is 0.92%
- The analysis error with Carrier ion exchange is 10%
- The micropipette error is 3%.
- The sample quartering error does not exist in our case, since we are taking the entire sample.
- The counting error with 95% confidence is:

$$2\sigma = 2 \frac{\sqrt{N}}{t} = 2 \frac{\sqrt{50}}{4320} = 0.0032 = 28.26 \%$$

$$\text{MDR} = 0.017 \pm 0.0048 \text{ pCi} \quad \text{MDR} = 0.629 \pm 0.17 \text{ mBq}$$

where N = No of counts  
t = counting time

JEN DOCUMENT No. \_\_\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0 Date 6-9-86

$\% \text{Error total} = \sqrt{(\text{Error \% a})^2 + (\text{Error \% b})^2 + (\text{Error \% c})^2 + (\text{Error \% d})^2}$  Page 10 of 14

Error total % = 30.14%

Minimum detectable radioactivity in a counting time of 4320 minutes with 95% confidence:

$$\text{MDR} = 0.629 \pm 0.19 \text{ mBq/sample}$$

or, expressed in pCi

$$\text{MDR} = (0.017 \pm 0.0051 \text{ pCi/sample, considering the}$$

total error.

The calculated experimental error is 28%, which, as can be observed, is near the calculated theoretical error.

### 3.8.4 Calculation of Recovery

The chemical recovery of the method is calculated through the use of the internal standard of Pu 242, and is expressed as a percentage of the added standard and that counted.

$$\text{Recovery} = \frac{\text{dpm counted} \times 100}{\text{dpm added}} = \%R$$

On occasion, a target analysis with the standard may be made and apply to it the total recovery of a series of analyses.

Sample analysis of marked targets should be employed with a given frequency in a series of analyses.

### 3.9 Expression of Results

The final results of the sample's Pu 239 + Pu 240 alpha radioactivity is expressed as radioactivity excreted per day. For that purpose, an estimate of the urine which the person monitored must excrete daily is made through analysis of the creatinine in the B urine sample. The following cases may result:

- a) The volume of urine analyzed agrees with the estimate. In this case, the radioactivity counted will be that excreted per day.
- b) The volume of urine excreted does not agree with that calculated through creatinine analysis. In this case, a check will be made to determine if the person suffers from any renal disorder; if no dysfunction exists, the calculated volume will be taken as the daily excretion.

The results by sample will be expressed in the following manners:

- Case a)  $A > \text{LLD}$

The sample thus has a true radioactivity value, the result being expressed as  $A \pm 2\sigma$  followed by the corresponding units.

- Case b)  $\leq A \text{ LLD}$

The result will be expressed as  $\leq \text{LLD}$  followed by its units.

#### 4. RESPONSIBILITIES

It is the responsibility of the director of the Radiological and Environmental Protection office to:

- Approve the updating of these procedures and direct such updating when considered appropriate.

- Monitor the proper application of these procedures.

It is the responsibility of the chief of the Radiological Environment Operations Unit to:

- Keep these procedures updated.
- Monitor the proper application of these procedures.
- Review the updates of these procedures.

JEN DOCUMENT No.\_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
REV\_0\_ Date 6-9-86 METRY

Page 12 of 14

It is the responsibility of the Chief of the Subunit assigned to the Palomares studies to:

- Keep these procedures updated.
- Monitor the correct application of these procedures by the analysts who perform them.
- Designate the analysts.
- Approve the results by signature.
- Submit the results obtained.

It is the responsibility of the analysts to:

- Perform the procedures correctly.
- Request the information needed and deliver that obtained in timely fashion and the proper form.

## 5. REFERENCES

- 1<sup>o</sup> Report compiled by: W.D. Moss and E.E. Campbell. Los Alamos Scientific Laboratory, Report written: February 8, 1966:
- 2<sup>o</sup> Determination of plutonium in urine by anion exchange.  
Evan Campbell and Moss. Health Physics Vol. 11 p.737-742, 1.966
- 3<sup>o</sup> G.H. Kramer. AECL-6879-1980
- 4<sup>o</sup> N.A. TALVETIE= Anal. Chem. 43-1971
- 5<sup>o</sup> Lloyd A. Currie. Limits for Qualitative Detection and Quantitative Determination Analytical Chemistry. Vol. 40-586-593 (1968)
- 6<sup>o</sup> Manual of Analytical Methods for Radiobioassay. Los Alamos, July 1983

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TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0 Date 6-9-86

Page 13 of 14

## 5.2 Reagents and Material

Phosphoric acid

Octyl alcohol

Hydrochloric acid 0.5N

Nitric acid 2N

Nitric acid 8N

Hydroxylamine hydrochlorate

Ammonium hydroxide 2%

Calcium nitrate

Ammonium oxalate 4%

Dowex 1 x 2 50-100 mesh resin

2 liter glass graduates

90 cc Teflon centrifugal tubes

Teflon magnetic stirring rods

Magnetic agitators with built-in thermostat

Water bath, as in drawing (Figure )

Aluminum heating blocks (Figure )

Teflon electrodeposit cells

Infrared lamp

Stainless steel planchets, diam. 1.2 cm., electrodeposit 0.7 diam.

Current rectifier

Electrodeposit equipment (Figure )

## 6. ENCLOSURES

a) Cited circular for analysis, with sample taking recommendations.

b) Form for annotation of data on the sample and other particulars.

c) Figures

d) Material decontamination.

Decontamination of glass and Teflon material coming from plutonium determination

Reagents

- Saturated potassium permanganate solution (64 g/l)
- Sulphuric acid (5%)
- Acid sodium sulphite (5%)
- Dabeer detergent

Decontamination

- Wash the material with Dabeer detergent
- Mix the saturated potassium permanganate solution and the sulphuric acid 0.2N (V/V) SOLUTION A

Procedure A

- Wash material with Solution A.
- Wash material with acid sodium sulphite.
- Wash with distilled water.

Procedure B

- Wash, while hot, with 50% nitric acid.
- Wash with Dabeer detergent.
- Wash with distilled water.

Ciemat

SHEET 1 OF 19

SPECIFIC PROCEDURE No. PR-X3-04

REVISION 0

TITLE: SEQUENTIAL DETERMINATION OF Am-241 IN 24-HOUR URINE SAMPLES  
BY ALPHA SPECTROMETRY

ENVIRONMENTAL IMPACT G. PUBLISHING AND DISTRIBUTION SERVICE

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PREPARED BY: (\*)

Asunción Espinosa /signature/ December 30, 1991

REVIEWED BY: (\*)

José Gutierrez /signature/ February 10, 1992

QUALITY ASSURANCE CONCURRENCE: (\*)

Margarita Segarra /signature/ March 4, 1992

APPROVED BY: (\*)

Francisco Mingot /signature/ EFFECTIVE DATE 3/5/1992

(\*) NAME(S), SIGNATURE(S), DATE(S)

FORM P 01-1

CIEMAT      TITLE:      SEQUENTIAL DETERMINATION OF  
                 Am-241 IN 24-HOUR URINE SPECIMENS BY  
                 ALPHA SPECTROMETRY  
                 SPECIFIC PROCEDURE No PR-X3-04

REVISION 0  
Date 12-5-91

SHEET 2 OF 19

RECORD OF REVISIONS

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			ESPINOSA	GUTIERRREZ	SEGARRA	MINGOT

JEN DOCUMENT No. \_\_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
REV\_0 Date 6-9-86 METRY

SHEET 3 OF 19

### TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Preparation of the Sample
    - 3.1.1 Precipitation of americium with calcium oxalate
    - 3.1.2 Purification of americium by anion exchange
    - 3.1.3 Elution of the americium of the column
    - 3.1.4 Electrodeposit
  - 3.2 Measurement
    - 3.2.1 Equipment
    - 3.2.2 Efficiency Calibration.
    - 3.2.3 Measurement of Sample
  - 3.3 Calculations
    - 3.3.1 Calculation of Lower Limit of Detection
    - 3.3.2 Calculation of Radioactivity
    - 3.3.3 Calculation of Errors
  - 3.4 Expression of results
4. REAGENTS AND MATERIAL
5. RESPONSIBILITIES
6. REFERENCES

JEN DOCUMENT No. \_\_\_

REV 0 Date 6-9-86

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-239 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 4 OF 19

## 1. PURPOSE

To describe the method used in the Environmental Impact and Geochemistry Operations Unit (Laboratory of Bioelimination of Ground Transuranides) of the Environmental Institute, for determination of Am-241 in urine samples.

## 2. SCOPE

Radiochemical analysis is made of 24-hour urine samples of people who may have suffered internal contamination.

## 3. COMPOSITION

### 3.1 Preparation of the Sample

- The 24-hour urine sample collected in accordance with procedure MA/01 is treated in accordance with this same procedure to eliminate the Pu-239+240.
- The nitric acid liquids coming from passage through the Bio Rad AG 1x2 column and the wash liquids from the column (paragraph 3.4.2 of procedure MA/01) are collected to determine their Am-241 contents.

JEN DOCUMENT No. \_\_\_\_

REV\_0 Date 6-9-86

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 5 OF 19

- . Add the Am-243 marker in nitric acid 1N medium, approximately 4 dpm.
- . Evaporate until almost dry.
- . Add approximately 50 ml of distilled water, and measure the pH.
- . If necessary, adjust the pH with ammonium hydroxide, to  $1.5 \pm 0.2$ .
- 3.1.1 Precipitation of the americium with calcium oxalate
  - . Add 50 ml of the saturated calcium oxalate solution to the sample adjusted to pH 1.5 (if the pH were higher, the phosphates would precipitate out). The precipitate formed must be milky.
  - . Allow to sit.
  - . Decant the precipitate.
  - . Centrifuge the residue three times at 3000 revolutions per minute for 15 minutes each time, washing with distilled water, and discarding the washing liquids.
  - . Dissolve the precipitate in 7 ml of boiling concentrated nitric acid, stirring vigorously with a Teflon rod.
  - . Add 93 ml of methanol and cover with flexible moldable translucent paper, allowing it to sit until the following day.

JEN DOCUMENT No. \_\_\_\_  
REV\_0\_ Date 6-9-86

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 6 OF 19

### 3.1.2 Purification of the americium by anion exchange

#### Preparation of the column

- Fill the glass column (1.2 cm inner diameter and 8 ml [sic] long, provided with a switch) with Bio-Rad AG-1x8, 100-200 mesh resin suspended in distilled water and previously agitated for 15 minutes.
- Wash the column with 1M nitric acid until it stops giving a chloride reaction with silver nitrate (a minimum of 5 column volumes).
- Wash the column twice with nitric acid 1M, 93% methanol.
- Pass the sample through the column at a flow rate of from 1 to 1 ml/minute.
- Wash the tube which contained the sample three times with 50 ml nitric acid 1M 93% of methanol and pass it through the column.

### 3.1.3 Americium Elution

- The americium is eluted in the column with 10 volumes of the column of nitric acid 1 M.

#### 3.1.4 Electrodeposit of the Sample

- Evaporate the elution of the column until dry under an infrared lamp.
- If residue remains in the bottom of the receptacle, it is dissolved in nitric acid 1M (93% methanol) and step 3.1.2 is repeated.
- When the elution receptacle has no residue, the radionuclide is electrodeposited.
- Add 2 ml of concentrated hydrochloric acid to the elution receptacle and evaporate dry. Repeat this step at least three times.
- Prepare the electrodeposit cell with a stainless steel planchet 12 mm in diameter.
- Add 2 ml of hydrochloric acid 2N to the elution receptacle.
- Add 1 drop of 5% methyl red.
- Neutralize the solution with 50% ammonium hydroxide until the color changes.
- Add at least 2 drops of hydrochloric acid concentrate, to reestablish acidity.
- Transfer the electrolyte solution to the electrodeposit cell.
- Wash the receptacle with distilled water and transfer it to the cell.

- Mount the cell on the electrodeposit device with platinum anode.
- Keep the electrodeposit at 400 milliamperes for one hour, 30 minutes.
- Add 2 ml of 50% ammonium hydroxide solution.
- Disconnect the device and wash the cell with distilled water.
- Dry the planchet under infrared lamp.
- Flame the planchet with a Bunsen burner between the flame's oxidizing and reductor zone.

### 3.2 Measurement

#### 3.2.1 Equipment

The radioactivity of the electrodeposited Am-241 is measured by means of alpha spectrometry, with silicon barrier semiconductor detectors having a nominal active area of 300 mm<sup>2</sup>, a resolution of 21 KeV and a dead time of 0.5 S. The operating power supply is 100 volts.

These detectors are connected to a multichannel analyzer and a DIGITAL PDP-11/23 P.C. with an operating system.

JEN DOCUMENT No. \_\_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
REV\_0 Date 6-9-86 METRY

SHEET 9 of 19

### 3.7.2 Efficiency Calibration

Calibration of the spectrometer and determination of baselines is done in accordance with the procedure using secondary standard planchets containing the Pu-238, Pu-239+240, Pu-242, Am-243 and Am-241 masters which are standardized in a proportional gas counter using the primary NBS standards.

By means of the secondary standards a spectrum of each isotope is obtained in all detectors using 479 channels, which cover the energy ranges of 4 to 6 Mev.

### 3.2.3 Sample Measurement

The planchet prepared in accordance with 3.1.4 is introduced into the counting equipment for measurement.

The counting time will be 5760 minutes.

### 3. Calculations

#### 3.3.1 Calculation of the Lower Limit of Detection (LLD)

• According to the Currie Criterion. The American standards ASTH (Volume 12, 01, 1983, C-100) for determination of AM-241 with a 95% confidence level, for which 2 x the square root of 2K takes a value of 4.65. The detection limit of the device is given by the formula:

$$LLD = \frac{4,65}{2,2E} \left( \frac{C_f}{T_f} \right)^{1/2}$$

$C_f$  = Counts per minute from baseline for isotope i.

$T_f$  = Counting time expressed in minutes.

CE = Count efficiency

#### 3.8.2 Calculation of Minimum Detectable Radioactivity

This calculation requires a sufficient recount of at least

JEN DOCUMENT No. \_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
REV\_0\_ Date 6-9-86 Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 11 OF 19

50 total counts which will permit a close enough approximation of the Poisson distribution to that of Gauss, for us to use Gaussian statistics.

. The Currie criterion for a small number of events is:

$$\text{MDR} = \frac{2,71 + 4,65 \sqrt{C_t}}{T \times 2,22 \times E \times R}$$

where:

T = Counting time

C<sub>t</sub> = Total counts

R = Recovery

With a mean recovery for our method of:

R = 70%

An equipment efficiency of:

E = 30%

and a counting time of 5760 minutes

we obtain:

$$\text{MDR} = \frac{2,71 + 4,65 \sqrt{50}}{5760 \times 2,22 \times 0,3 \times 0,7} = 0,481 \text{ mBq/sample}$$
$$= 0,013 \text{ pCi/sample}$$

(0.013pCi/sample)

### 3.3.3 Calculation of Errors

3.3.3.1 The alpha spectrometer calibration error is given by the errors of the secondary standards utilized and certified by the N.B.S.

The variance of the different calibration measurements is sufficiently low to permit reproduction of the measurement in 98.6% of the cases.

3.3.3.2 The error of the minimum detectable radioactivity will be formed by the accumulation of the following random errors:

- a) Spectrometer calibration error which is considered random.
- b) Radiochemical analysis error.
- c) Micropipette error.

- d) Sample quartering error.
- e) Counting error.

- Calibration error is 0.92%
- The analysis error with Carlier ion exchange is 10%
- The micropipette error is 3%.
- The sample quartering error does not exist in our

case, since we are taking the entire sample.

- The counting error with 95% confidence is:

$$2\sigma = 2 \frac{\sqrt{N}}{t} = \frac{2 \sqrt{50}}{5760} = 0.0025 = 19,2\%$$

$$\text{MDR} = 0,013 \pm 0,0025 \text{ pCi}$$

$$\text{MDR} = 0,481 \pm 0,0914 \text{ mBq}$$

where: N = No of counts

t = counting time

$$\text{Total error} = \sqrt{(\text{Error}\%a)^2 + (\text{Error}\%b)^2 + (\text{Error}\%c)^2 + (\text{Error}\%e)^2}$$

expressed on %

$$\text{Total error} = 22\%$$

JEN DOCUMENT No. \_\_\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0 Date 6-9-86

SHEET 14 OF 19

Minimum detectable radioactivity in a minimum counting time of 5760 minutes with 95% confidence.

$$\text{MDR} = 0.481 \pm 0.0914 \text{ mBq/sample}$$

or, expressed in pCi

$$\text{MDR} = (0.013 \pm 0.0025 \text{ pCi/sample})$$

The calculated experimental error is 28%, which, as can be observed, is near the calculated theoretical error.

#### 3.8.4 Calculation of Recovery

The chemical recovery of the method is calculated through the use of the internal standard of Am 243, and is expressed by the ratio between the dpm counted and the dpm added.

$$\text{Recovery} = \frac{\text{dpm counted} \times 100}{\text{dpm added}} = R$$

JEN DOCUMENT No.\_\_\_\_  
REV\_\_0\_\_ Date 6-9-86

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 15 of 19

It is advisable to conduct periodic sampling of urine targets which have had a determined amount of Am-241 added so as to calculate the mean recovery of the method. The mean of our sample is not under 70%.

#### 3.4 Expression of Results

The final results of the sample's Am-241 alpha radioactivity is expressed as radioactivity excreted per day. For that purpose, an estimate of the urine which the person monitored must excrete daily is made through analysis of the creatinine in a urine sample. The following cases may result:

- a) The volume of urine analyzed agrees with the estimate. In this case, the radioactivity counted will be that excreted per day.
- b) The volume of urine excreted does not agree with that calculated through creatinine analysis. In this case, a check will be made to determine if the person suffers from any renal disorder; if no dysfunction exists, the calculated volume will be taken as the daily excretion.

JEN DOCUMENT No. \_\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0\_ Date 6-9-86

SHEET 16 of 19

The results by sample will be expressed in the following manners:

- Case a)  $A > \text{LLD}$

The sample thus has a true radioactivity value, the result being expressed as  $A \pm E_{\text{total}}$  followed by the corresponding units.

- Case b)  $\leq A \text{ LLD}$

The result will be expressed as  $\leq \text{LLD}$  followed by its units.

#### 4. REAGENTS AND MATERIAL

Concentrated nitric acid, Probus

Nitric acid 7.8N

Nitric acid 2N

Nitric acid 1N

Nitric acid 1N 93% methanol

Concentrated Hydrochloric acid, Merck

Hydrochloric acid 2N

Calcium oxalate

Ammonium hydroxide

JEN DOCUMENT No. \_\_\_\_

TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

REV\_0\_ Date 6-9-86

SHEET 17 of 19

Ammonium hydroxide 50%

Dowex 1X8 (100 - 200 mesh) exchange resin

Methyl red 5%

Reflon rods

Precipitate receptacles, 250 ml

Centrifuge tubes, 100 ml

Ion exchange glass columns, diam 1.3 cm (w/ switch)

Disposable electrodeposit cells.

Stainless steel planchets

#### 5. RESPONSIBILITIES

It is the responsibility of the chief of the Environmental Impact and Geochemistry Unit to:

- Keep these procedures updated.
- Monitor the proper application of these procedures.
- Review the updates of these procedures

It is the responsibility of the chief of the Ground Transuranides Laboratory:

JEN DOCUMENT No.\_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
REV\_0 Date 6-9-86 METRY

SHEET 18 OF 19

- Keep these procedures updated.
- Monitor the proper application of these procedures by the analysts which perform them.
- Designate the analysts.
- Approve the results by signature.
- Submit the results obtained.

It is the responsibility of the analysts (defined as those persons who execute these procedures as their job, or a portion thereof) to:

- Perform the procedures correctly.
- Request the information needed and deliver that obtained in timely fashion and the proper form.

JEN DOCUMENT No. \_\_\_\_\_ TITLE: PROCEDURE FOR DETERMINATION OF  
REV\_0\_ Date 6-9-86 Pu-23 IN URINE SAMPLES BY ALPHA SPECTRO-  
METRY

SHEET 19 OF 19

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MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER  
SPECIFIC PT-X7-13 7-30-92 0

3 20

TABLE OF CONTENTS

1. PURPOSE
2. SCOPE
3. COMPOSITION
  - 3.1 Detection System
  - 3.2 Calibration Method
  - 3.3 Conduct of Pulmonary Measurements
    - 3.3.1 Checking the detection system
    - 3.3.2 Determination of thoracic thickness
    - 3.3.3 Measurement of subject
  - 3.4 Evaluation of Radioactivity Deposited in the Lungs
    - 3.4.1 Evaluation of the corporal background
    - 3.4.2 Calculation of deposited radioactivity
    - 3.4.3 Correction of pulmonary radioactivity
  - 3.5 Treatment of Errors
  - 3.6 Minimum Detectable Radioactivity
4. QUALITY ASSURANCE
  - 4.1 Quality Assurance of the Detection System
  - 4.2 Quality Control for Conduct of Measurements
5. TREATMENT OF DATA
6. QUALIFICATIONS
7. RESPONSIBILITIES
8. REFERENCES
9. APPENDICES

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER 0  
SPECIFIC PT-X7-13 7-30-92

4 20

1. PURPOSE

To describe the methods applied for measurement, operations and calculations to permit obtaining of dosimetric information of actinide lung contamination.

2. SCOPE

Application of the Phoswich Corporal Radioactivity Counter (CRC) system belonging to the CIEMAT Radiological Protection Unit for "in vivo" measurement of radionuclides emitting photons of energy less than 200 keV.

3. COMPOSITION

3.1 Detection System

Specific Procedure PR-X7-08, "Calibration of the Phoswich Detection System" applies.

3.2 Calibration Method

Specific Procedure PR-X7-08, "Calibration of the Phoswich Detection System" applies.

3.3 Conduct of Pulmonary Measurements

3.3.1 Checking the detection system

In accordance with Specific Procedure PR-X7-08, system stability and validity of the calibrations in use is checked before each measurement, generating a daily report on Form PR-X7-13-01: "Verification of the Detection System" (Appendix I). The program daily checks the conditions of measurement in order to detect any variations due to thermal drift or electronic maladjustments associated with the detectors, and compares the results obtained with the values predefined in the calibrations.

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER  
SPECIFIC PT-X7-13 7-30-92 0

5 20

The functions of the program are:

\* Verification of energy calibration: The official applies Section 5 of Specific Procedure PR-X7-08.

\* Verification of efficiency and resolution calibration: the official applies Section 5 of Specific Procedure PR-X7-08.

\* Monitoring environmental background: Daily 1800 sec background before and after each measurement session, verifying that the value obtained is within the permitted range of error.

If these verifications indicate a system malfunction, several courses of action may be taken, depending on the circumstances:

- A variation of 10% of the energy slippage and of increased daily environmental background will be corrected by adjusting circuit parameters, with the corresponding subsequent study.

- A resolution variation of more than 30% or a small (<15%) loss of detection efficiency necessitates application of a correction factor at the time of estimating the quantity of the radionuclide measured. However, this cause leads to the subsequent recalibration of the system.

### 3.3.2 Prior preparation

When persons come into contact with the Corporal Radioactivity Counter for the first time, a personal record is opened, using Form PR-X7-13-02: "Record of Registration in the CRC" (Appendix II), and they must be included in some of the control programs defined in Specific Procedure PR-X7-10: "Conduct of Internal Dosimetric Controls"

- Monitoring external contamination

If the presence of any type of external contamination is suspected, the procedure requires:

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER 0  
SPECIFIC PT-X7-13 7-30-92

6 20

\* Monitoring of external contamination by a radiological protection technician. The skin of the person to be measured will be bathed and scrubbed, so that there will be total certainty that any contamination measured comes from internally deposited radionuclides.

In the event of any incident, the radiological protection technician will make a contamination check of both clothing and personal effects.

- Information and safety instructions for the subject

The person to be examined will be given special clothing and footwear for access to the counting chamber. At the time of each measurement, Form PR-X7-13-03: "CRC Measurement Monitoring Report" (Appendix C) will be used to record name, company, region examined, monitoring program, weight, height and age.

The subject will be asked if he/she has had any previous body radiation count. If the answer is affirmative, the subject will be given a brief summary of the counting process safety conditions, while being installed on the counter. If it is the first time the subject has reported for measurement, the installation technician must briefly explain to him/her the purposes, method and safety precautions of the corporal counting process.

Prior to any "in vivo" measurement, the technician must ensure that the person who is to be examined has a perfect understanding of the entire measurement process. He/she must be told how to leave the counting chamber, and shown how the audio and video communications operate. This equipment includes the voice-activated intercom system and closed circuit television which permits the technician to observe the subject in the counting position.

As for the purposes of the measurement, the subject will be told whether it is sought to identify and

quantify the radioactivity deposited internally due to an explosion, or to confirm that any internally deposited radiation is below the detection limits.

### 3.3.3 Determination of thoracic thickness

#### a) Experimental: by ultrasound

The thoracic thickness is measured with an ultrasound apparatus designed especially for this purpose (HEWLETT-PACKARD 7215A ECHOENCEPHALOSCOPE). This equipment consists of a high frequency electric power generator and a transmitter, which is a probe of piezoelectric material which transforms the electric energy into ultrasound energy.

Since the semireduction thickness for x-rays having 17 Kev of energy in muscle and fatty tissue is 7 mm, it is essential that the thoracic thickness be measured accurately; that is, it is necessary to know the detector-to-lung distance in order to be able to apply the corresponding calibration factors.

The following procedure is followed:

- . Materials needed
- \* Aquasound transmission gel
- \* Kleenex type tissues
  
- . Prior to the ultrasound measurement
- \* Connection of the equipment and the necessary adjustments must be completed at least 5 minutes before the measurement.
  
- . Preparation of the subject to be examined
- \* The person sits on a chair
- \* The subject is warned that the transmission gel will be slightly cold when applied to the skin.
  
- . Measurement
- \* Apply the gel to the ultrasound transmitter or probe.
- \* Place the probe gently on the chest, in the positions indicated in Form PR-X7-13-04: "Determination of Thoracic Thickness" (Appendix IV).

- \* Observe the pulses which appear on the screen when NEAR COARSE is varied.
- \* Adjust the SCREEN CENTER until the pulse coincides with the vertical center line.
- \* Repeat this operation in the twelve positions defined in Form PR-X7-13-04.
- \* Calculate and record the result on Form PR-X7-13-04.
  
- . After measurement
- \* Use a tissue to remove the gel traces from the subject's chest.
- \* The person may go inside the chamber for measurement.

b) Theoretical calculation of thoracic thickness (T.T.)

Routine calculation of thoracic thickness in men employs an empirical function which establishes a correlation between the subject's weight, height and age, by means of the equation:

$$T.T.(mm) = 1.24 \times [\text{Weight (Kg)} / (\text{Height})^2(m)] - 0.03 \text{ Age (yrs)} - 0.8$$

Determination of thoracic thickness in women is given by the above equation (thoracic thickness calculated for a man of the same age, weight and height) but increased by 20%. This calculation is verified by an indeterminate number of measurements conducted using AEA technology.

3.3.4 Measurement of the subject

- \* Ask what music he/she wants to listen to during the measurement.
- \* Escort the subject into the counting chamber.
- \* Place him/her on the bed in the decubitus dorsal position.
- \* Position the detectors just above the area corresponding to the lungs. The detectors are positioned by moving the arms of the structure until they reach a position as close as possible to the pulmonary area, consistent with easy breathing by the subject.

Once both the person and the detectors are properly positioned, the detector arms are firmly secured, ensuring that the subject is as comfortable as possible. He/she will be cautioned not to change position under the detectors and remain supine throughout the counting period.

\* Close the door of the shielded chamber and activate the counting, normally setting a counting time of 1800 seconds.

\* At the end of the measurement, transfer the spectrum to, and save it in, the computer associated with the detector.

\* Open the door of the shielded chamber. Tell the subject not to move until told otherwise, and proceed to loosen the arms of the structures of the two detectors, and swing them back away from the bed upon which the person just measured is lying. When this has been done, the subject may be permitted to get up and leave the chamber.

### 3.4 Evaluation of Radioactivity Deposited in the Lungs

#### 3.4.4 Evaluation of the corporal background

In order to evaluate deposited radioactivity in the lungs, the contribution made by the corporal background must be subtracted.

The physical constitution of the individual is very significant in the detection of low energy emitters in the lungs, due to the heavy absorption this type of radiation undergoes in passing through the individual's thorax.

The main factor responsible for individual differences is the contribution of K-40 (natural radioactivity of the human body) and typical levels of Cs-137 < 199 Bq due to fall-out. Thus measurement of an individual always involves the use of his own personal background; that is measurement of the individual before his exposure to any type of internal contamination.

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER 0  
SPECIFIC PT-X7-13 7-30-92

10 20

In this case, if the subject's background (personal background) is known, any statistically significant increase in the defined region of interest will mean radioactivity from the element, provided that, at the time of measurement, the subject presents no other contamination from more powerful emitters.

If the corporal background is not known (estimated personal background), a method has been established which consists of analyzing reference spectra obtained from measurement of "target" individuals, that is, persons not exposed to radiation. We thus obtain a type spectrum which represents the contribution of the K-40 and Cs-137 as a function of the thoracic thickness of the individuals. Therefore, the background of any subject can be expressed through a regression linear function of the form:

Estimated personal background =  $A + B \times$  (thoracic thickness)

where A and B are coefficients to be determined.

### 3.4.2 Calculation of deposited radioactivity

Analysis of the resulting spectrum:

a) Existence of photopeak and single contaminant

If there is a photopeak, the radioactivity is calculated with the following equation:

$$\text{Radioactivity in lungs (Bq)} = A_n / T_c \times E_r \quad [1]$$

where  $A_n$  are the net counts in the photopeak of interest, calculated from:

$$A_n = I_1 - F_1$$

where

$I_1$  = Count spectrum, less the daily environmental background count.

$F_1$  = Actual or estimated personal corporal background (Section 3.4.1)

$T_c$  = Counting time

$E_r$  = Counting efficiency in the corresponding region of interest, expressed as a function of the thoracic thickness.

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER

SPECIFIC                      PT-X7-13                      7-30-92                      0

11 20

The calibration factors in effect are summarized below (Table 1)

Table 1  
Calibration Factors as a Function of Thoracic Thickness

Thoracic Thickness (cm)	Pu-239 (cpm/kBq)	Am-241 (cpm/kBq)	U-238(b) (cpm/mg)
1.6	5.44	1103.04	6.43
1.8	4.10	1012.16	6.08
2.0	3.18	937.23	5.74
2.2	2.53	874.24	5.45
2.4	2.05	801.44	5.00
2.6	1.69	773.87	4.98
2.8	1.41	733.12	4.79
3.0	1.20	697.11	4.61
3.2	1.02	665.03	4.46
3.4	0.88	636.24	4.31
3.6	0.77	610.24	4.18
3.8	0.68	586.62	4.06
4.0	0.60	565.06	3.95
4.2	0.53	545.29	3.85
4.4	0.48	527.06	3.75

ROI (a)                      [10 - 25]                      [37 - 74]                      [36 - 110]

(a) Region of interest in keV

(b) The U-238 was determined from Th-234 gamma emission, assuming that the radioactivity of U-238 and Th-234 are in a 1:1 ratio.

b) Existence of photopeak and simultaneous presence of two contaminants (the case of Plutonium and Americium)

In the event that there is contamination with interference of two contaminants, as in the case of plutonium and americium, the theoretical relationship between the intensities of the two regions of interest must be established, both for pure americium and for plutonium, so as to estimate the contribution of each.

This relationship is established by means of a factor R (quotient of the net areas of the two regions) which is a function of the type  $R = s \times e^{bx}$ , a and b being coefficients to be determined, and x the thoracic thickness.

The determination of the net counts of a contaminant  $An'$  (roi1) is made by subtracting the contribution of the other (roi2), as follows:

$$An' (roi1) = An (roi1) - An(roi2) * R(roi1/roi2)$$

$$An' (roi2) = An (roi2) - An(roi1) * R(roi2/roi1)$$

These calculations are repeated successively until the results are stabilized. The radioactivity deposited in the lungs is subsequently calculated from equation [1].

#### c) Nonexistence of photopeak

In the event measurement detects no type of contamination, we proceed to calculate the minimum detectable radioactivity (MDR) for the radionuclide of interest.

#### 3.4.3 Correction of Pulmonary Activity

When the radioactivity of the contaminant is distributed uniformly in the lungs, liver and bones (Am-241-critical organs) the results obtained from the measurement and analysis methods described heretofore will reflect the combined contents in the lungs and liver in the count, due to the proximity of the two organs. At normal working levels it is not possible to appreciate this proportion, but in this case, too, the interference of the radioactivity in the bones is not very significant. The correction would have to be made for each particular situation and circumstances, and therefore, cannot be documented in general procedures.

### 3.5 Treatment of Errors

In most cases, errors in calculation of the radioactivity deposited in the lung can be included in two categories:

1. Those which by their nature are truly due to chance. The main component is associated with counting statistics and the consequent uncertainties in analysis of a spectrum with its constituent elements for a normal contact period of 30 minutes. Other variations due to chance are those relating to the position of the detector relative to the subject, but their contribution is truly insignificant (1%).

2. Those which, in the case of individual measurements, are considered systematic errors. Included in this case are:

- Systematic errors in calibration which originate from the fact that the internal geometry of a calibration mannequin cannot duplicate that of a human being. This type of error cannot be evaluated, but we follow the recommendations of ANSI N13.30, which suggest that the standard deviation between a target person and the mannequin is approximately 25%.

- Systematic errors in calibration provoked by an incorrect estimate of thoracic thickness. In general, they are not very significant, because both in the mathematical expression used for the calculation and in the experimental measurement of men, we find a margin of error of  $\pm 2.8$  mm, which leads to an uncertainty of only 5% in the calibration factor. By doubling the thoracic thickness error for the case of measurement of women, where the method of estimation is less satisfactory, we obtain an uncertainty of 10% for the calibration factor, which is less than the statistical counting error.

- Systematic errors if the pulmonary distribution of the contaminant in a subject differs markedly from the uniform distribution in the calibration lungs. It is recommended that this error be disregarded because the uncertainty incurred in the radioactivity of the lungs used for calibration is a systematic error source of greater scope.

All these considerations are taken into account in calculating the errors which the program intended for the purpose effects.

### 3.6 Minimum Detectable Radioactivity

The value of the Minimum Detectable Radioactivity (MDR) indicates the capacity of the detection equipment to distinguish between counts corresponding to individuals who are internally contaminated and those who are not.

Due to variations which exist in the size and shape of the body among the general populace and to the variable distribution of radioactivity within the organism and each organ, the values of the MDR are given for a uniform description and for a "model-man" (ICRP 23).

Thus the uncertainties in measurement will be less for thin and small persons and greater for tall and heavy persons, due to the differences in autoabsorption. The MDR can be calculated by using the following formula in accordance with the Currie criterion and the recommendations of ANSI N13.30:

$$\text{MDR} = \frac{2.71 + 4.65 S_b}{E_1 \times T_c}$$

where:

$S_b$  = Standard deviation of the corporal background, using the criteria given in sections 3.4.1 and 3.5.

$E_1$  = Calibration factor or efficiency, in the appropriate units

$T_c$  = Counting time.

The MDRs for a mean thoracic thickness of 3 cm and for the effective calibrations are summarized below (Table 2).

Table 2

Minimum Detectable Radioactivity  
(for a mean thoracic thickness of 3 cm)

Radionuclide	Energy (keV)	MDR
Pu-239	17	3.5 KBq
Am-241	59.5	12 Bq
U-238 (Th-234)	63 92	3.5 mg

4. QUALITY ASSURANCE

All CRC operations are carried out in accordance with the internal CIEMAT quality assurance standards and the norms of the "American National Standards Institute" (ANSI): "Standard internal dosimetry for mixed fission products and activation products" (ANSI 1978) and "Practice for occupational and radiation exposure records systems" (ANSI 1972).

4.1 Detection system quality assurance

There are three basic types of daily verifications (energy, efficiency and control of environmental sources) when that type control is carried out. The criteria are as set forth in Procedure PR-X7-08 and in Section 3.3.1 of this procedure.

Detector resolution is something which is not specifically verified every day. Any significant change in resolution will give rise to a counting efficiency loss and should be reflected in the daily verification of the detection system. Periodic tests of the resolution of the detectors and their associated photomultiplier tubes should be conducted every 6 months.

These verifications provide very valuable information of possible signs of deterioration in the detectors. All these data are recorded in the CRC operation logbooks and stored in the computer on a daily basis.

#### 4.2. Quality Assurance for Measurements

The procedures to guarantee quality assurance are included in each measurement process.

All measurements taken with the corporal radioactivity counter must be documented and stored. The documentation must be sufficiently detailed so that a person qualified in this work can reconstruct the calculations and confirm that the efficiency and energy calibrations did operate and were maintained on the applicable day. Fortunately, the natural K-40 existing in each person's body acts as an internal standard of identification of any change in the energy calibration and also gives us an approximate indication as to whether the counting efficiency is satisfactory.

The results of the measurements are verified in detail on a weekly basis by the official responsible for the CRC, in order to ensure that the data have been filed properly and backup copies have been made of everything recorded in the computer during the week.

#### 5. TREATMENT OF DATA

The computer support of the CRC consists of several microcomputers which can be interconnected with the CIEMAT computer network.

Each measurement made in the CRC gives rise to a spectrum or histogram of the number of energy events for a given detector and geometry. The multichannel analyzers which collect this information provide, in turn, information which can be presented in different terms.

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER 0  
SPECIFIC PT-X7-13 7-30-92

17 20

A series of programs convert the input data into a common format, so that all data, regardless of source, can be handled by the same group of programs.

The basic functions are described below:

\* Spectrum transfer program

- Transmission of spectra from the MCA to the PC, description and smoothing of it, recording of regions of interest and tabulation of results, for additional analysis of a single datum or a group of data.

\* Detection system verification program

- Determination of centroids, resolution and area of the photo-peaks used in calibration and comparison of the values obtained daily with those described in the calibration file, rejecting the measurement if the variations exceed a predefined percentage. Generation of a report.

\* Updating program of backgrounds: environmental, mannequin and corporal

- Storage of environmental and mannequin background spectra, with the capability of determining the statistical variation of the chamber's own background over a given period, and for the desired region of interest.
- Determination of net personal background of an individual, as a function of his/her thoracic thickness. Filing of the personal spectrum.
- Determination of corporal backgrounds estimated from a linear regression between background values and thoracic thicknesses of a set of files stored, with the corporal values of "target" persons.

\* Calculation and data storage program

- Rapid calculation of results, inter-relating the above described files with a file of calculated efficiencies for each radionuclide calibrated.
- Preparation of a final report using Form PR-X7-13-05: "Technical CRC Analysis Report" (Appendix V).
- Storage of data in a data base file. Each spectrum is associated with its unique serial number; the alphanumeric designators and the calculated results give

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER 0  
SPECIFIC PT-X7-13 7-30-92

18 20

rise to a file which is stored in the computer's data base. This data base can now be accessed through several different programs to display, print out, make calculations, or carry out all types of statistical analyses.

It must be carefully verified that all the descriptive information is correct before posting it to a safety file. All of these data are fundamental if the results are to be available to authorized personnel desiring to access them.

Once the entire process of processing the results has been completed, the interested party and the agency chief will be informed of the results of monitoring conducted, in accordance with Procedure PR-X7-10 (Forms PR-X7-10-04 and PR-X7-10-05).

In the event of any positive count of a routine nature, or due to some incident, the chief of the UOPRI will be notified immediately, and an evaluation will be made of the dose received by the individuals affected. From that time forward, a monitoring measurement program will be required for the person occupationally exposed to the radiation.

If any external agency contracts the services of the CRC, a report will be submitted on Form PR-X7-13-06: "Report of internal contamination to external clients" with the data of the measurements and contamination levels detected, and dose evaluation if necessary according to Procedure PR-X7-10 (Forms PR-X7-10-04 and 05).

## 6. QUALIFICATIONS

The official responsible for the Corporal Radioactivity Counter is the person designated by the chief of the Internal Radiological Protection Unit, who will see to the proper technical maintenance of the equipment assigned to this detection system and the proper application of this procedure.

## 7. RESPONSIBILITIES

It is the responsibility of the chief of the UOPRI:

- To review the updates to this procedure and direct them when considered suitable.
- To ensure the correct application of this procedure.

MEASUREMENT OF ACTINIDES IN THE LUNGS IN THE  
CORPORAL RADIOACTIVITY COUNTER

SPECIFIC

PT-X7-13

7-30-92

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SPECIFIC PT-X7-13 7-30-92

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9. APPENDICES

- APPENDIX I Verification of the Detection System
- APPENDIX II CRC Record Form
- APPENDIX III CRC Measurement Monitoring Report
- APPENDIX IV Thoracic Thickness Determination
- APPENDIX V CRC Analysis Technical Report
- APPENDIX VI Report of Internal Contamination of External Clients

APPENDIX I

VERIFICATION OF THE DETECTION SYSTEM

CIEMAT  
CORPORAL RADIOACTIVITY COUNTER

Form PR-X7-13 -01

Phoswich System  
Condition Verification

Date

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Energy Variation

Precise Am-241 Source

Deviation =

Counting Time = 300 s

First Centroid =

Calibration Centroid =

Second Centroid =

Calibration Centroid =

Deviation = %

Difference =

Calibration Difference =

Error = %

---

Efficiency Variation

Measurement Count

Calibration Count =

Error = %

FWHM =

---

Environmental Background Variation

Counting Time =

Zone

Calibr.

CPM

Daily

CPM

Error

Count

Count

(34-74)

(74-148)

(72-220)

(170-250)

APPENDIX II

CRC RECORD FORM



APPENDIX III

MONITORING REPORT FOR EACH  
MEASUREMENT

CIEMAT

Form PR-X7-13-03

CORPORAL RADIOACTIVITY COUNTER

RECORD OF MEASUREMENT

DATE:

REPORT No. CIEMAT/SPR/DIC/ /

FULL NAME:

CRC No:

COMPANY / (C.O.):

POSITION:

CONTROL PROGRAM:

REGION EXAMINED:

AGE: Years

WEIGHT: Kg

HEIGHT: cm

THORACIC THICKNESS: cm

No OF FILED SPECTRUM: XX CRC No. Measurement No.

APPENDIX IV

THORACIC THICKNESS DETERMINATION

CORPORAL RADIOACTIVITY COUNTER

ULTRASOUND DETERMINATION OF THORACIC THICKNESS

NAME:

CRC No:

DATE:

WEIGHT (Kg):

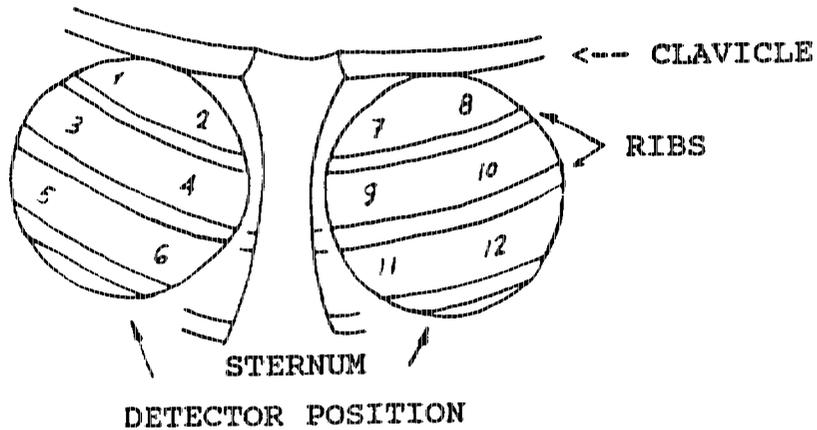
HEIGHT (m):

AGE:

THEORETICAL THICKNESS 9cm):

REMARKS:

ULTRASOUND POSITIONS



RIGHT LUNG

LEFT LUNG

Positions:

Position:

- 1:                    2:
- 3:                    4:
- 5:                    6:

- 7:                    8:
- 9:                    10:
- 11:                   12:

TOTAL of the 12 positions:

MEAN VALUE:

= THORACIC THICKNESS (cm)

APPENDIX V

CRC ANALYSIS TECHNICAL REPORT



APPENDIX VI

REPORT OF INTERNAL CONTAMINATION  
OF EXTERNAL CLIENTS

CIEMAT

Form PR-X7-13-06

Internal Dosimetry  
Corporal Radioactivity Counter

REPORT OF INTERNAL CONTAMINATION

Detection System:

Ref:

Type monitoring:

Date:

MONITORING PROGRAM:

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Code	Surname	First Name	Measure- ment Date	Radio- activity measured (mg U238)	Statis- tical error (mg U238)	MDR (mg U238)
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Signed: