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DEPOSITION OF PLUTONIUM IN THE LUNG OF A WORKER FOLLOWING AN ACCIDENTAL INHALATION EXPOSURE

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Abstract

The deposition of PuO_2 in the lungs of an occupationally exposed worker is characterized by assay for plutonium in excreta samples and from *in vivo* measurements of ^{241}Am in the thoracic region. Chelation therapy by intravenous injection of 1 gm Ca-DTPA was initially performed shortly after the incident and repeated using 0.5 gm of the chelate four additional times in subsequent days post intake. Analysis of the air sampler filter retrieved from the site of the exposure identified the isotopic composition and particle size of the plutonium material inhaled by the worker.

Chelation with Ca-DTPA did not significantly reduce the magnitude of the lung or systemic deposition as determined from assay of plutonium in urine samples collected from the worker. *In vivo* measurements for ^{241}Am verify the retention of the inhaled material in the lung and also indicate the ingrowth of an amount of ^{241}Am as a daughter product of the ^{241}Pu initially inhaled.

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DISCLAIMER

Little, if any, plutonium has been cleared from the body in the period following chelation treatment. Translocation of activity from the lung to the skeleton is examined by performing in vivo measurements for ^{241}Am in the head (skull).

INTRODUCTION

A worker was exposed to a small quantity of plutonium while performing a routine pressing, sintering, and density measurement of plutonium dioxide pellets in a glovebox. A fatigue crack in a glove allowed a small quantity of plutonium dioxide powder to be released to the room when the worker removed his arm from the port. Contamination was found on the worker's right hand, nostrils, and protective clothing as well as the room floor and the exterior surface of the glovebox. Following removal of the protective clothing and containment of the skin contamination, the worker was transported to the whole body counting unit for *in vivo* measurements.

Preliminary *in vivo* measurements of 1.3 nCi ^{241}Am in the anterior thorax indicated an internal deposition of plutonium. Although plutonium was detected in spot urine samples collected immediately following the exposure incident, samples obtained several hours later were negative implying that some or all of the initial plutonium activity may have been due to external contamination. The concentration of plutonium in the first fecal sample, collected the day following the exposure, was approximately 30 pCi/gm.

INTRODUCTION (contd.)

Decorporation therapy consisted of a series of five injections with Ca-DTPA administered intravenously, one gram being given shortly after the exposure, and 0.5 gram of the chelate given in four subsequent days. The insoluble nature of the inhaled material was supported by the lack of enhanced urinary plutonium excretion following chelation therapy.

Samples of the filter from the continuous air monitor and the material being sintered in the glovebox were analyzed by a chemical separation of plutonium and americium followed by alpha and mass spectroscopy to determine the percentage isotopic composition of the material to which the worker was exposed.

MEASUREMENTS

The isotopic composition of the plutonium and americium activity on the air sampler filter was measured by alpha, mass, and gamma spectrometry. The composite results of the atom percent and the alpha percent abundance of the plutonium isotopes and ^{241}Am are given in Table 1. The errors associated with each measurement represent two standard deviations

MEASUREMENTS (contd.)

at the 95% confidence level. The ratio of the total alpha activity to ^{241}Am is approximately 11 to 1.

Measurements of ^{241}Am located in the anterior thorax region of the worker are used to determine the plutonium deposition in the lung since the ratio of plutonium to americium in the inhaled material is presumably known from the air filter analysis.

The *in vivo* measurements are routinely performed using either of two scintillation detector systems. Each system is equivalent in overall sensitivity for ^{241}Am in the lung but employ different types of detectors. The basic lung counting array uses four NaI(Tℓ) detectors connected in parallel; one pair in contact with the anterior thorax and the other pair against the posterior thorax with a layer of fabric from a canvas cot between the detector and subject (Pa77).

The second *in vivo* measurement system employed with this evaluation was a special system using two dual-crystal phoswich detectors [NaI(Tℓ)/CsI(Tℓ)] placed in direct contact with the anterior thorax of the worker. Although only two detectors are employed in the special system, the

MEASUREMENTS (contd.)

operating characteristics (i.e., lower background and improved sensitivity) are equivalent to the four detector array (Ne78).

The phoswich detectors provide a measure of the uranium and neptunium L X-rays at approximately 17 KeV from the decay of ^{241}Am and plutonium which the four detector array cannot adequately detect. In addition, using a live-time computer analysis program, the phoswich detector provides a mechanism to reduce the low energy contribution to the ^{241}Am spectrum from Compton scattered photons due to the presence of higher energy, gamma-emitting nuclides in the body such as ^{40}K or ^{137}Cs for example (Ne78).

In addition to measurements of ^{241}Am in the thorax, the phoswich detector array is uniquely employed to detect activity that may have translocated from the lung to the skeleton. *In vivo* measurements of ^{241}Am in the head (skull) can be used to estimate whether translocation from the lung has occurred (Co77). The detection limit for ^{241}Am in the skull, using two detectors and a 30-minute counting period is 30 pCi at the 95% confidence level.

MEASUREMENTS (contd.)

Results of 28 sequential *in vivo* thorax measurements for ^{241}Am deposited in the lung of the exposed worker are listed on Table 2 and illustrated on Figure 1. The net energy spectrum, Figure 2, from an *in vivo* thorax measurement made on day 493 post intake, shows a prominent peak in the ^{241}Am region. An overall systematic error of approximately 10% for each measurement was determined from repetitive counts prior to which the detectors were repositioned on the worker's thorax.

Measurements of the worker's chest wall thickness have been made using an ultrasound unit operated with a phased array transducer. The chest wall thickness for the worker was 3.26 cm which is significantly greater than that calculated from the relationship of Ramsden and Speight (Ra67) which employs a ratio of weight to height to estimate chest wall thickness for routine survey counts. The effect of this additional attenuating tissue between the lung and the detectors has been included in the evaluation of the worker's internal deposition of ^{241}Am.

An evaluation of the systemic deposition is performed using measurements of plutonium in excreta samples and com-

MEASUREMENTS (contd.)

paring the elimination with time to the metabolic models for the nuclide (La56, He57). Both urine and fecal samples were obtained from the worker following the exposure. The results of the plutonium analysis in these samples are given on Tables 3 and 4. No estimate of error is listed with each analytical value because the autoradiographic procedure used to detect the plutonium alpha particles in the electrodeposited samples following chemical separation does not provide a mechanism for tracer analysis. However, based upon batch results and the evaluation of samples in which known concentrations of calibrated NBS traceable plutonium activity have been added, the overall estimate of error is approximately ±100% at the limits of detection (.05 dpm).

ANALYSIS

Results of sequential *in vivo* measurements of ^{241}Am in the thorax indicate no significant translocation of the material at 500 days post intake. Since the isotopic composition of the inhaled material includes ^{241}Pu, the *in vivo* ^{241}Am measurements require simple in-growth corrections. However, the air sampler filter may not adequately represent the isotopic

ANALYSIS (contd.)

composition of the material which the worker inhaled so that the correction applied to the in vivo measurement may not be adequate. A least square regression line fitted to the corrected thorax measurements has a zero slope which indicates that the isotopic composition of ^{241}Am and ^{241}Pu on the air filter is an adequate estimate to that inhaled by the worker.

No detectable amount of ^{241}Am was measured in the head (skull) count. Translocation of activity from the lung to the lymph nodes would not be detected from an in vivo thorax measurement since the deposition sites cannot be differentiated with large radius scintillation detectors. Translocation of activity from the lung to the lymph nodes by phagocytosis could occur. However, at this time approximately 500 days post intake, the activity deposited in the lung appears to have an effective half-time much greater than that assumed for typical Class Y material.

Assuming 1.3 nCi of ^{241}Am is deposited in the lung for the year and the plutonium alpha to ^{241}Am ratio is 12.5, the dose to the lung would be slightly greater than 15 rem using the ICRP dose calculations.

ANALYSIS (contd.)

Results of radiochemical analysis for plutonium in urine and fecal samples indicate that there is essentially no systemic deposition of activity in the worker. The insoluble nature of the inhaled material, as determined from *in vivo* measurements and the apparent lack of efficacy of the chelation therapy immediately following intake, is supported, in retrospect, by the urine and fecal plutonium excretion results.

Approximately 15 nCi of plutonium-alpha mixture was initially inhaled as a result of the accidental exposure. Assuming a maximum plutonium concentration in feces (following seven days post intake) of 0.01 pCi/gm, the total activity excreted, including the first week post intake, was less than 100 pCi. Less than 10 pCi of plutonium was excreted in urine which includes the samples collected during the period of chelation therapy. That is to say, less than 1% of the inhaled plutonium material has been removed from the body in the 500 days post intake. This fact characterizes the inhaled material as extremely insoluble and supports the results obtained by *in vivo* measurements.

CONCLUSION

The material to which the worker was exposed was calcined at approximately 600° and was being subjected to high pressure pretreatment prior to sinterability testing. It appears that this combination of procedures produced a plutonium dioxide particulate with an aerodynamic diameter suitable for alveloar deposition. The results of *in vivo* thorax measurements indicate that the amount of inhaled activity deposited in the lung has remained constant for approximately 500 days. Implications of the analytical measurements performed thus far characterize the inhaled material as extremely insoluble with an effective half-life in the lung much greater than typical Class Y plutonium material.

The clearance half-time for high-fired, insoluble plutonium oxide in the human lung is typically less than 500 days. Metabolic models published by the I.C.R.P. indicate a range for the clearance half-time from 50 days for soluble material to 500 days for highly insoluble oxides. It has been concluded that, for this case, sequential thorax measurements support essentially an infinitely-long lung clearance half-time for the compound inhaled by this worker.

CONCLUSION (contd.)

The I.C.R.P. metabolic model predicts that the initially soluble fraction of inhaled Class Y material which enters the bloodstream from the naso-pharynx and traceo-bronchial regions should be approximately 2.5% of the long-term material (i.e., $T_{1/2}$ = 500 days) deposited in the lung. Taking advantage of the fact that, in this case, decorporation therapy was initiated immediately following uptake, we can assume that the first 24-hour urine sample represents most of the initially soluble material entering the bloodstream from the naso-pharynx and tracheo-bronchial region. This actual short-term fraction is unusually small and represents only 0.004% of the long-term material measured in the lung. Assuming that urinary plutonium excretion on the days of chelation represent the fraction of activity entering the blood from the lung, a clearance half-time of 35 years is calculated. Using the fecal excretion of plutonium at approximately 10 days post intake, a clearance half-time of 40 years is predicted. This agreement is probably fortuitous.

The above data is consistent with in vivo measurements for americium in the thorax. No decrease or clearance can be ob-

CONCLUSION (contd.)

served within the precision of the measurements (i.e., approximately ±20%). A clearance half-time of 1000 days (i.e., twice that for Class Y material) would have decreased the initial amount of inhaled material by 25% within 500 days, a fact unsupported by the results of *in vivo* counting.

Further studies will include a continuation of *in vivo* thorax measurements for ^{241}Am and collection of additional excreta samples for plutonium assay in order to determine the presence of any systemic organ distribution. A follow-up study of the solubility and particle size of the material on the air sampler filter is also considered.

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TABLE 1

ISOTOPIC COMPOSITION OF ALPHA ACTIVITY ON AIR FILTER

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
Atom Percent	0.065 ± 0.0034	86.4 ± 0.3	11.6 ± 0.3	1.4 ± 0.1	0.24 ± 0.03	0.25 ± 0.04
Alpha Percent	11.6 ± 0.6	53.8 ± 0.2	26.1 ± 0.7		0.01 ± 0.0012	8.4 ± 1.3

Error represents 2 S.D. at 95% confidence level.

TABLE 2

RESULTS OF IN VIVO MEASUREMENTS FOR ^{241}AM IN THE THORAX

Days Post Intake	^{241}Am Detected (nCi)	^{241}Am Ingrowth Calculated (nCi)	^{241}Am Deposition Calculated (nCi)
1	1.3	.001	1.29
2	1.1	.002	1.09
3	1.3	.003	1.29
7	1.3	.007	1.29
17	1.2	.016	1.18
24	1.3	.023	1.27
31	1.3	.029	1.27
38	1.1	.036	1.06
39	1.1	.036	1.06
45	1.1	.042	1.05
52	1.3	.048	1.25
59	1.4	.055	1.34
80	1.2	.074	1.12
111	1.3	.103	1.19
122	1.3	.113	1.18
132	1.5	.122	1.37
136	1.4	.126	1.27
150	1.6	.138	1.46
178	1.3	.163	1.13
254	1.1	.231	.86
279	1.4	.253	1.14
311	1.5	.281	1.21
339	1.5	.305	1.19
366	1.6	.328	1.27
402	1.8	.358	1.44
430	1.6	.381	1.21
465	1.7	.411	1.28
493	1.6	.434	1.16

TABLE 3

MEASUREMENTS OF PLUTONIUM IN EXCRETA[1]

(URINE)

Days Post Intake	Type[2]	Volume (mℓ)	Plutonium pCi/ℓ[3]	Comment
0	Spot	250	2.7	Field collection
0	Spot	380	-[4]	Following decontamination
1	Spot	100	-	Prior to chelation
1	24-hr total	1500	0.8	1 gm Ca-DTPA
2	Spot	100	-	Prior to chelation
2	24-hr sim	900	0.3	0.5 gm Ca-DTPA
3	24-hr sim	950	0.1	0.5 gm Ca-DTPA
4	24-hr sim	1250	-	
5	24-hr sim	700	0.1	
7	24-hr sim	700	0.1	0.5 gm Ca-DTPA
8	24-hr sim	1550	0.1	
10	24-hr sim	1200	0.1	0.5 gm Ca-DTPA
11	24-hr sim	1000	0.2	
12	24-hr sim	1200	-	
18	24-hr sim	1200	0.1	
25	24-hr sim	1100	-	
32	24-hr sim	1400	-	
39	24-hr sim	1100	-	
91	24-hr sim	1000	-	
112	24-hr sim	1100	-	
114	24-hr sim	650	-	
119	24-hr sim	800	-	
226	24-hr sim	500	-	

1. Autoradiography employed to detect alpha particles in sample following plutonium chemical separation procedure.
2. Collection of simulated 24-hour sample includes all urine passed during the periods one-half hour before retiring and one-half hour after rising for two consecutive days.
3. Error associated with these values is estimated to be ±100% at these magnitudes.
4. Less than MDA (0.025 dpm/sample).

TABLE 4

MEASUREMENTS OF PLUTONIUM IN EXCRETA[1]

(FECAL)

Days Post Intake	Type	Plutonium Activity (pCi/gm)
1	24-hr total	30.54[2]
3	24-hr total	0.06
4	24-hr total	0.03
5	24-hr total	0.02
6	24-hr total	0.04
280	24-hr total	-[3]
282	24-hr total	-

1. Alpha spectrometric measurement for ^{238}Pu, $^{239,240}Pu$ following chemical separation.
2. Error associated with these measurements is approximately $\pm$100%.
3. Less than MDA (0.2 dpm/sample).

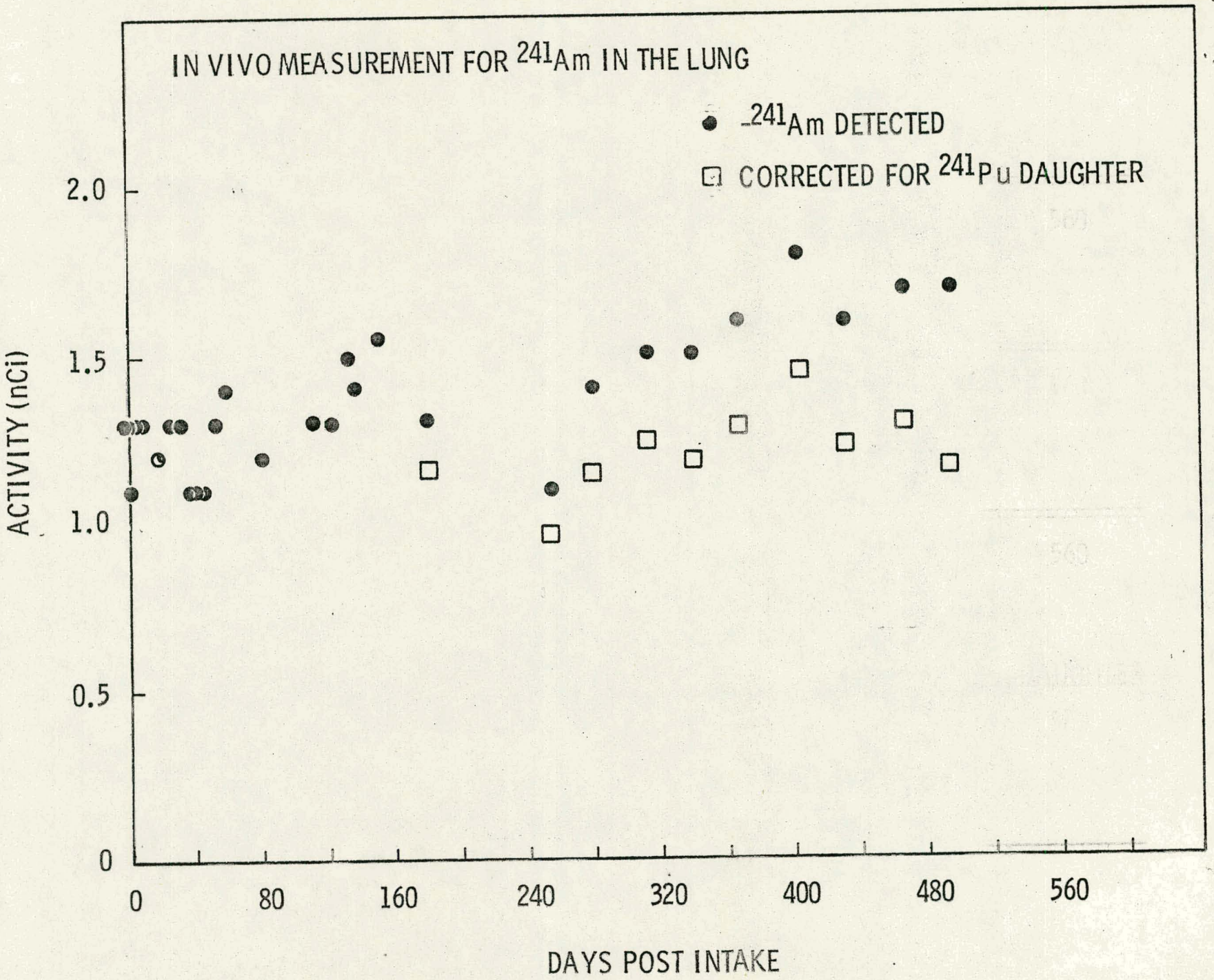
IN VIVO MEASUREMENT FOR ^{241}Am IN THE LUNG
^{241}Am DETECTED
CORRECTED FOR ^{241}Pu DAUGHTER
ACTIVITY (nCi)
2.0
1.5
1.0
0.5
0
0
80
160
240
320
400
480
560
DAYS POST INTAKE

Figure 1

IN VIVO MEASUREMENT OF THORAX — TWO DUAL CRYSTAL PHOSWICH DETECTORS

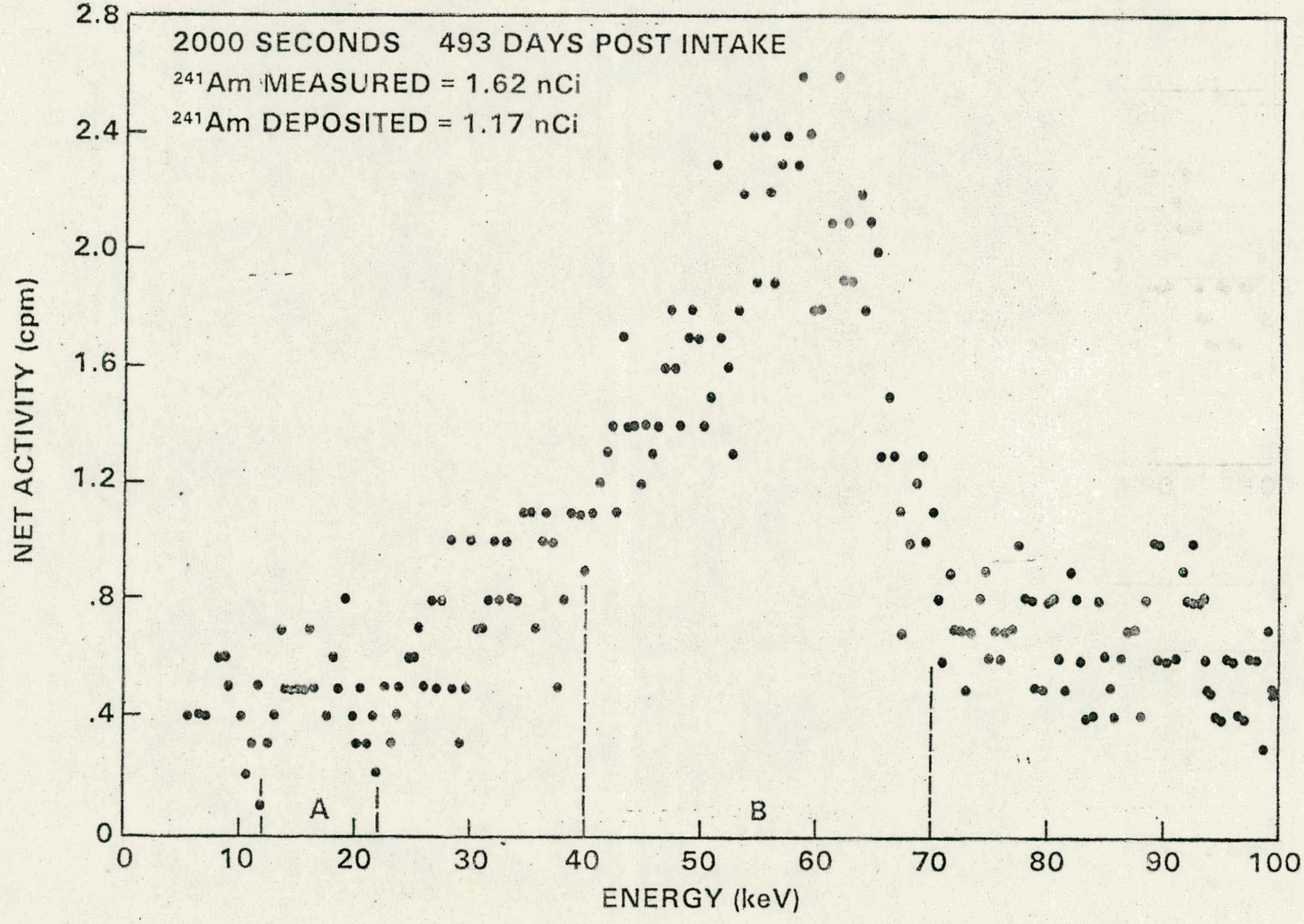

Figure 2.

LIST OF FIGURES