LECTURE 7

IN VIVO COUNTING OF PLUTONIUM IN DOGS*INTRODUCTION

Instrumentation for measuring plutonium in dogs and rodents was developed in our laboratory in about 1956. The dog scanning instrument was discussed in an earlier lecture. These instruments used a detector system of 2 in. by 1 mm thick, thallium-activated sodium iodide scintillation crystals. Using single channel analyzers, we looked primarily at the 17 keV X rays from plutonium. These instruments were very useful but we encountered difficult calibration problems associated with the high attenuation of the low energy X rays. We have not solved all of these problems, but considerable progress has been made in adapting a counter designed for human chest counting to in vivo counting of dogs. To understand the problems, factors important to successful in vivo counting of plutonium will be reviewed.

X- and Gamma-Ray Emissions from Plutonium

Gamma-ray emissions of 100 keV and at the 384 keV complex lack intensity for the detection of plutonium at maximum permissible body burden levels. For such low levels of plutonium, the significant radiations are the L X rays from uranium daughters. The 13.6, 17, and 20.2 keV X rays result from the internal conversion of gamma rays from excited states in the uranium daughters. For pure ^{239}Pu , about 4% of the alpha particle events produce X rays. Other X rays may arise when other plutonium isotopes are present.

 ^{241}Am as a Tracer for Pu

A small amount of ^{241}Am may also occur from the beta decay of ^{241}Pu , which has a 10-year half life. The ^{241}Am emits a 60 keV gamma which can be used as a tracer for plutonium in

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most cases; however, ^{241}Am also gives rise to L X rays which complicates the interpretation of X-ray data. The 60 keV photon is emitted at a rate of about 35.9% of the ^{241}Am alpha particle events. The 60 keV gamma of ^{241}Am is not as severely affected by variations in tissue distribution, tissue absorption, or surface contamination as are the L X rays. Other minor low energy radiations present include 39 keV (^{241}Am) and 52 keV (^{239}Pu).

Important to the detection and measurement of plutonium by in vivo counting of these photon emissions is knowledge of the isotopic composition of the contaminating material. It will change with irradiation time of the plutonium (Figure 7.1). The percentage of ^{239}Pu decreases with irradiation time as the higher isotopes increase. The composition will also change with time after removal from the neutron field (Figure 7.2). The buildup of ^{241}Am increases the total X-ray activity. If

FIGURE 7.1. Change in Isotopic Composition of Plutonium with Irradiation Time⁽¹⁾

FIGURE 7.2. Change in Activity of a Typical Plutonium Sample as a Function of Time⁽¹⁾

there is no separation of americium from plutonium, the buildup of ^{241}Am in the lung can be appreciable (Figure 7.3). The importance of these factors to the interpretation of in vivo counting data has stimulated further work in this area.

For our animal studies we have used scintillation counters rather than proportional counters. The relatively high efficiency of scintillation counters is an attractive feature. In addition they are more easily used for unanesthetized animals than proportional counters.

FIGURE 7.3. Ratio of $^{239}\text{Pu} + ^{240}\text{Pu}/^{241}\text{Am}$ in the Lung (Pulmonary + Lymph) as a Function of Time for an Initial Pulmonary Deposition of 13 nCi of Plutonium ($^{239}\text{Pu} + ^{240}\text{Pu}$) and 1 nCi of ^{241}Am for the Assumed ^{241}Pu Weight Percents in the Initial Deposition - for Oxide Inhalation Only⁽²⁾

BIOLOGICAL RELATIONSHIP BETWEEN Pu AND Am

An important aspect of our use of in vivo counters for our experimental animals is a study to determine whether ^{241}Am can be used as a tracer for plutonium. This study was also motivated by the need for this information to evaluate the results of in vivo counting of plutonium in humans. The objective was to determine whether plutonium and americium separated in the body after inhalation of various plutonium compounds containing ^{241}Am in varying amounts. This study was in two parts, radiochemical and biological. To obtain the biological information it was necessary to develop analytical techniques for plutonium isotopes and ^{241}Am in biological tissues.

Determination of Plutonium-to-Americium Ratios in Biological Samples

In the evaluation of the relative behavior of plutonium and americium in exposed animals, a very precise method of measurement is required. Two techniques were developed and are illustrated with spectra in Figure 7.4. In each, the plutonium and americium were not chemically separated from each other since this could result in preferential loss of one radioisotope, but direct counting techniques were employed which allowed their simultaneous measurement. In one case, following wet ashing, all salts were converted to chlorides and the plutonium and americium were electroplated from an ammonium chloride electrolyte. This method had typical plating recoveries exceeding 99% when there were no interfering trivalent elements present in the dissolved sample. Following electroplating, alpha pulse height analysis was used to measure the ratio of 5.15 MeV alphas to the 5.48 MeV alphas. The 5.15 MeV alphas are emitted by ^{239}Pu and ^{240}Pu while the 5.48 MeV alphas are emitted by both ^{241}Am and ^{238}Pu . The additional peak at 4.18 is due to ^{238}U which is added as a

carrier for electrodeposition. To use this method for plutonium-ameridium analysis, it is necessary that the ratios of the plutonium isotopes be known.

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FIGURE 7.4. *Plutonium-Ameridium Measurements in Biological Materials⁽³⁾*

Photon spectrometry, the second method, illustrated in Figure 7.4, is preferable where it is applicable. Here, the plutonium X rays and the low energy americium gamma rays are measured with a recently developed lithium-drifted silicon

diode having approximately 1 keV resolution. The excellent resolution permits separation of the 13 keV, 17 keV, and 20 keV X-ray lines from plutonium decay, from the 26 keV and 60 keV gamma rays from ^{241}Am decay. The greater penetration of X rays allows direct counting of wet-ashed samples if corrections for self-absorption effects are made. These effects depend on the sample mounting technique and a very uniform mount must be used. Calibration curves for the corrections are made by preparing standards of known plutonium-ameridium content but with different weights of tissue ash.

The primary limitation of the X-ray spectrometry method is that present silicon diode detectors are small and the X-ray fluorescence yields for ^{239}Pu low. Therefore, the overall counter efficiency is rather low. Where sufficiently high levels of exposure have occurred, however, the technique offers the advantage that no chemical separation is required for measuring both absolute amounts of plutonium and ameridium and their ratios. Samples which could be analyzed by both methods gave results which were in good agreement.

Pu-Am Ratios in Dogs After Inhalation of Plutonium

The relative retention, translocation, and excretion of plutonium and ameridium are being studied under several conditions by comparing the ratio of ^{238}Pu and ^{239}Pu to ^{241}Am in dog tissues with the ratio in the aerosol sample collected at the time of an inhalation exposure. The following conditions are being studied:

- Dogs exposed up to 10 years ago to relatively pure $^{239}\text{PuO}_2$.
- Dogs exposed 3 months previously to one of four different types of PuO_2 , relative pure ^{239}Pu .
- Dogs exposed to $^{239}\text{Pu}(\text{NO}_3)_4$ and died within a year.
- Dogs exposed to $^{239}\text{PuF}_4$ containing higher isotopes of Pu and ^{241}Am . Three of six dogs were treated with DTPA.
- Dogs exposed to PuO_2 prepared by calcining oxalate enriched with higher isotopes of plutonium.

Preliminary results have been obtained. ^{239}Pu - ^{241}Am ratios in tissues of dogs sacrificed 3 months after inhalation of one of four different $^{239}\text{PuO}_2$'s are given in Table 7.1. These ratios are compared with the ratios in the sample of aerosol collected at the time of exposure. In dogs which inhaled these oxides, there was no evidence for separation of plutonium from ^{241}Am .

TABLE 7.1. 239 - ^{240}Pu -to- ^{241}Am Ratios in Dog Tissues 90 Days After Inhalation of Plutonium

Plutonium Compound	Aerosol	Lung	Tracheobronchial Lymph Nodes	Liver
$\text{Pu}(\text{NO}_3)_4$	38	70	50	12
PuF_4	6.5	5.5	5.5	--
	6.5	8.3	6.6	--
PuO_2				
1000° Oxalate	14	13	12	--
350° Oxalate	13	11	13	--
450° Metal	31	32	34	--
123° Metal	31	32	30	--
PuO_2 (Enriched)	1.9	1.9	1.8	--

The averaged results of a $^{239}\text{Pu}(\text{NO}_3)_4$ experiment are given in Table 7.1. In all dogs analyzed to date there was an increased ^{239}Pu - ^{241}Am ratio in lungs and lymph nodes and a decreased ratio in liver relative to the aerosol inhaled. This is tentatively interpreted as being due to relatively greater translocation of ^{241}Am to the liver from the lung than ^{239}Pu . The high ratios in the lymph nodes could be a result of ^{241}Am translocation from the lymph nodes, or the translocation may have occurred while the ^{241}Am was still in the lung before it had a chance to accumulate in the lymph nodes.

Data for inhaled $^{239}\text{PuF}_4$ are also shown in Table 7.1. The plutonium-ameridium ratios in the tissues were about the same as for aerosol. Therefore, separation of plutonium from ameridium appeared to be very slight.

Similar results showing no separation of plutonium from ameridium were obtained for PuO_2 which was highly enriched with ^{240}Pu , ^{241}Pu and ^{241}Am when the aerosol was inhaled.

Dogs which had been exposed to $^{239}\text{PuO}_2$ prepared by calcining the oxalate at 350° yield data for long times after exposure, up to 78 months. These data are shown in Table 7.2. The ratios of ^{239}Pu - ^{241}Am in the tissues were not greatly different from the aerosol samples. Possibly the liver shows a reduced ratio--greater accumulation and/or retention of ^{241}Am than ^{239}Pu . Additional data will be obtained from dogs exposed 5 to >8 years ago.

TABLE 7.2. $^{238}, ^{239}, ^{240}\text{Pu}$ -to- ^{241}Am Ratios in Dog Tissues After Inhalation of PuO_2

Dog Number	Time After Exposure (Months)	$^{238}, ^{239}, ^{240}\text{Pu}$ -to- ^{241}Am Ratios*			
		Aerosol	Lung	Lymph Nodes	Liver
268	40	12.6	12.8	10.5	
257	44	13.0	15.8	17.2	
259	54	13.6	13.4	11.4	
246	54	13.1	11.8	9.8	
216	60	13.1	15.4	15.6	7.5
85	66	13.1	15.4		10.6
86	67	13.1	6.3	13.3	
81	73	13.1	11.8	14.3	
212	78	13.2	14.3	9.0	

* $^{238}\text{Pu} = 1.1\%$

In these studies, there was little indication that ^{239}Pu and ^{241}Am were significantly separated in the body after inhalation of $^{239}\text{PuO}_2$ containing amounts of ^{241}Am . However, since ^{241}Am was clearly translocated to the liver more rapidly than ^{239}Pu after inhalation of $\text{Pu}(\text{NO}_3)_4$, it cannot be concluded that ^{241}Am is a valid tracer for plutonium in all cases.

IN VIVO COUNTING OF PLUTONIUM

Counter Design

The counter consists of 52 detectors, each composed of a photomultiplier optically coupled to a 1 mm thick NaI (Tl) crystal, Figure 7.5. The cleaved crystals are 1-3/4 in. in diameter, mounted on a quartz base and with a 1 mil aluminum window. These detectors are arranged into four arrays of 13 each. Each array is housed in a 6 in. x 12 in. by 7 in. deep stainless steel, light-tight box with a 10 mil mylar window. The anodes of the phototubes are connected in parallel and the signal is fed to a pulse-height analyzer. Counting is done in a lead cave or in a steel room. Dogs were counted in the position shown in Figure 7.5 or placed in plastic box on which two banks of crystals were mounted. This is shown in Figure 7.6.

Dog Phantom

A dog phantom was used to calibrate the counter. The phantom was fabricated by a commercial firm using the skeleton of one of our mature control dogs. Figure 7.7 compares the phantom with a live dog. The lung area was filled with a lung equivalent material and the phantom was divided into 33 2-cm thick sections containing a grid of holes on 2-cm centers. The phantom is shown with a section removed in Figure 7.8. The section is shown in Figure 7.9. The holes are shown filled with tissue-equivalent plugs. The plugs can be replaced by tissue-equivalent capsules containing a radionuclide standard.

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(a)

Neg 39326-10

(b)

FIGURE 7.5. Scintillation Counter Used for Measurement of ^{239}Pu in Thorax of Dogs (52 Detectors in four arrays of 13 each.)

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FIGURE 7.6. Scintillation Counter for Measurement of ^{239}Pu in Dog Thorax (Two arrays of 13 detectors mounted on opposing sides of dog-holding box.)

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FIGURE 7.7. Comparison of Dog Phantom with Anesthetized Dog

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FIGURE 7.8. Dog Phantom Showing Section Removed

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FIGURE 7.9. *Section of Thorax Removed from Dog Phantom [Tissue equivalent plugs (white) can be replaced with tissue equivalent capsules containing radionuclide standard sources.]*

Such a capsule is shown in the photograph. Figure 7.10 is a radiograph of two sections removed from the phantom thorax and shows the grid of holes and the extent of the lung-equivalent material. The tissue-equivalent plugs and capsules can be seen in the lung area.

Figure 7.11 is a ventral-dorsal radiograph of the phantom. The ribs and vertebrae can be identified. Also the stomach can be seen. Figure 7.12 is a lateral radiograph showing the rib cage.

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FIGURE 7.10. Radiograph of Two Sections Removed from Thorax of Dog Phantom Showing Dog Skeleton and Spaces Where a Number of Tissue Equivalent Plugs Have Been Removed (Fourteen radionuclide-containing capsules can be seen in the section on the right.)

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FIGURE 7.11. Radiograph of Dog Phantom - Dorsal-Ventral View

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FIGURE 7.12. *Radiograph of Dog Phantom - Lateral View*

Tissue and fat equivalent pads representing the addition of 2 kg of tissue can be used to adjust the phantom weight.

Figure 7.13 shows the phantom in the counting box with the two banks of detectors attached. Figure 7.14 shows the phantom in the box used for the scanning counter. The phantom is shown in a chest counter in Figure 7.15. This counter has four 2 mm x 5 in. crystals surrounding the chest. The background for this counter was about 50 cpm in the low energy region, 13.6 to 25.6 keV, and 200 cpm in the second, 13.6 to 66.3 keV. The lower detection limit for the 17 keV X-ray region is about 8 nCi and for the 60 keV photon region, the limit is about 7 nCi.

Factors Influencing In Vivo Counting

Bone shielding is a significant problem in detecting the 17 keV X rays. The half value layer for 17 keV X rays is

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FIGURE 7.13. Dog Phantom in Dog Counting Box on Which 2-13
Detector Arrays are Mounted

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FIGURE 7.14. Dog Phantom Being Placed in Counting Box for Longitudinal Scanning in Dog Counter

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FIGURE 7.15. Dog Phantom in Dog Thorax Counter Consisting of Four 2 mm $\times$ 5 inch Crystals Arranged Around the Thorax Region

BNWL-1221

0.6 cm in tissue but only 0.03 cm in bone. Therefore, the skeleton is essentially opaque to 17 keV X rays. In the dog, the half value layer for a plutonium source in the chest was 1.3 cm. This value is higher than for tissue or skeleton because of the relatively low density of tissue in the chest.

Changing distribution of plutonium within the dog is an important factor. This occurs after inhalation of plutonium as it slowly accumulates in the tracheobronchial lymph nodes and more slowly is translocated to liver, etc. An example of how this changes the sensitivity of in vivo counting is shown in Figure 7.16. As the percent of the body burden accumulates in the lymph nodes, the sensitivity decreases. This Figure also shows that the sensitivity increases as more of the plutonium is translocated to the liver.

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FIGURE 7.16. Effect of Distribution of ^{239}Pu in Dog Phantom on Counting Sensitivity

Table 7.3 shows a comparison between the X-ray sensitivity obtained for four dogs and the value obtained using the same plutonium distribution in the dog phantom. The close agreement illustrates the value of the phantom.

TABLE 7.3. *Effect of Organ Distribution of ^{239}Pu on Calibration of Counter*

Dog Number	Percent in Tissue				Calibration Factor	
	Lung	Bronchial Lymph Nodes	Liver	Other	Measured	Predicted From Phantom
76	47	45	3	5	2.1	2.14
183	52	38	6	4	2.1	2.19
213	49	27	15	8	2.3	2.35
272	63	17	14	6	2.45	2.42

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Another factor of importance to in vivo counting was mentioned earlier. This is the isotopic composition of the plutonium. The effect of isotopic composition on sensitivity of counting is shown in Figure 7.17. Isotope A identifies a relatively old PuO_2 source in which ^{241}Am has grown. Isotope B was a relatively pure ^{239}Pu source. The sensitivity for detection of Isotope A was about twice that of Isotope B.

Whole body burden values obtained with the whole body counter have also been compared with values estimated from the analysis of excretion data. An example is shown in Figure 7.18. The whole body burden is expressed in microcuries. When the animal was sacrificed after 90 days and analyzed for plutonium, the body burden was found to be 7.5 μCi . The in vivo counter

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FIGURE 7.17. Effect of Isotopic Composition
on Counting Sensitivity

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FIGURE 7.18. Comparison of Whole Body Burden of ^{239}Pu Determined from Analyses of Excreta with that Obtained with Whole Body Counter

estimate was 7.8 μCi , an error of about 5%. In about 12 dogs which had body burdens ranging from 1 to 45 μCi , estimates obtained by counting the 60 keV region were within a few percent. Estimates obtained by counting the 17 keV region were generally higher than the actual analyzed burdens.

One of the causes for the largest errors encountered in in vivo counting of plutonium in experimental animals is external contamination. A very small amount of plutonium on the outer surface of skin is counted with a very high efficiency, relative to that within the animal because of the lack of shielding by body tissues. This is particularly troublesome in inhalation studies where it is very difficult to avoid small amounts of external contamination. Therefore, washing of the animal with detergent is generally required each time the dog is counted until levels in the excreta are so low that they constitute a negligible contaminating source.

SUMMARY

The use of in vivo counting for experimental animals has proved feasible in our laboratory. A number of factors have been identified which constitute potential problems to correct interpretation of the data. Among these are isotopic composition of the plutonium, distribution of plutonium within the animal, separation of plutonium from americium in the case of inhaled $\text{Pu}(\text{NO}_3)_4$, and external contamination of the animal.

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