RECEIVED BY DTIE JUL 30 1971

BNWL-SA-3759

MASTER

CONF-710710--6

COUNTING PLUTONIUM IN THE TRACHEOBRONCHIAL
LYMPH NODES^(a)

K. L. Swinth, J. F. Park
Battelle Memorial Institute
Pacific Northwest Laboratories
Richland, Washington 99352

and

P. J. Moldofsky
General Electric Company
Philadelphia, Penn. 19101

June 24, 1971

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

(a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Fig

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

COUNTING PLUTONIUM IN THE TRACHEOBRONCHIAL
LYMPH NODES*

K. L. Swinth, J. F. Park
Battelle Memorial Institute
Pacific Northwest Laboratories
Richland, Washington

and

P. J. Moldofsky
General Electric Co.
Philadelphia, Pennsylvania

ABSTRACT

Following inhalation, plutonium will translocate to the thoracic lymph nodes. On a long term basis, up to 50% of the plutonium may be translocated to lymph nodes with concentrations of 50 to 100 times that in the lung.

In order to differentially detect the plutonium in the tracheo-bronchial lymph nodes an esophageal probe was built using avalanche detectors. To date, five probes have been built for evaluation. The typical probe is 2.5 cm long by 1 cm in diameter and located at the end of a 60 cm cable. The latest probe contains 3 detectors in an equilateral triangle arrangement which increases the sensitivity and reduces the angular response of earlier probes.

Experiments with five dogs, both when anesthetized and later when they were euthanized, have shown the usefulness of the probes. A peak in count rate occurs as the probe, located in the esophagus, passes the lymph nodes at the bifurcation of the trachea. Data from one dog indicates a Minimum Detectable Amount (MDA) of 26 nCi using a one minute count.

Evaluation of the probe in a human phantom indicates an MDA of 7 nCi to 22 nCi for a 20 minute count. The variation results in interpretation of the counting data due to geometrical considerations. The unique ability of the avalanche diode to detect plutonium X-rays while at the end of a long cable makes the detection of plutonium in the bronchial lymph nodes possible. Although the avalanche diodes have a low efficiency, (5-10%) they do have extremely low backgrounds (~ 1 cpm).

*This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830.

INTRODUCTION

Due to the great radiotoxicity of plutonium, accurate assessment of the quantity and location of plutonium material present In Vivo must be made in order to determine a course of action. Plutonium decays by alpha emission with insignificant accompanying gamma radiation; however, L X rays of the uranium daughter are emitted due to internal conversion. The X rays, with energies of 13.6, 17.0 and 20.2 keV, provide the means of detecting plutonium when the alpha radiation is absorbed by surrounding material (i.e. tissue). The range of the 5.15 MeV alpha of ^{239}Pu is about 48μ in water and about 100μ in lung tissue.

For pure ^{239}Pu about 4.8 X rays are emitted for every 100 alpha particles; however, small amounts of other isotopes are generally present and their effect is to increase the ratio of X rays to alpha particles. ^{241}Am , a daughter of ^{241}Pu , with its high specific activity and yield of X rays (37.6 per 100 alphas) has a major role in increasing the ratio. Typically a ratio of 7.4 X rays per 100 alpha particles is observed. ^(1,2) This indicates a total emission rate of 6.6×10^3 X rays per minute for one Maximum Permissible Body Burden (40 nCi) of plutonium (2.6×10^3 X rays per minute for one Maximum Permissible Lung Burden). When the plutonium is located in the lungs, less than one percent of these X rays will escape from the body. ⁽³⁾

A further factor complicating the detection of inhaled plutonium is the translocation of alveolar deposited plutonium. Figure 1 shows the change in distribution of inhaled plutonium with time for a group of experimental dogs. After 8 to 9 years, 40-50% of the material has been translocated to the thoracic lymph nodes, 10 to 15% to the liver, 5% to the skeleton and 5% to the abdominal lymph nodes. About 10% is left in the lungs.

This redistribution of plutonium is important for a number of reasons. First, the distribution will affect the sensitivity of any external measurements of the plutonium body burden as shown in Reference 4. With the low count rates available it would be impossible to externally determine the portion of the material located in the thoracic lymph nodes. For example, if a suspected contamination case is counted and later found to have a previous history of inhalation exposure, it would be difficult to ascertain the exact calibration to be employed. In fact, with the increase in sensitivity of whole body counters, the majority of the material could be previously undetected plutonium which has translocated to the thoracic lymph nodes. Second, the mean concentrations of plutonium in the lymph nodes can be 50 to 100 times that found in the lungs. In the dogs described previously, the tracheobronchial and mediastinal lymph nodes had the highest concentrations followed by the hepatic lymph nodes and then the lungs. For dosimetric purposes it is desirable to know how much material might be located within the lymph nodes.

The tracheobronchial lymph nodes are located near the bifurcation of the trachea with the largest mass at the carina. Most of these lymph nodes are in close proximity to the esophagus thus making them accessible for counting by a probe placed in the esophagus. This circumvents the problem with tissue absorption in the chest wall; however, biological variations and control of the geometry, become important for absolute determinations. Although operation of a probe within the trachea would be possible, the esophagus is the most desirable mode of entry due to biological considerations. This paper describes the application of avalanche diodes to the counting of plutonium concentrated in the tracheobronchial lymph nodes.

The Tracheobronchial Geometry

Figure 2 shows a typical geometry of the tracheobronchial area. This is the geometry observed on one preparation. There is a considerable amount of variation; however, one generally observes a group of nodes (3-5) extending along the right bronchus.⁽⁵⁾ The nodes in the figure extended 4 cm along the right bronchus and 2 cm along the left bronchus. Other nodes are observed along the sides of the esophagus above the tracheal bifurcation. In another preparation the node(s) along the right bronchus was of a single elliptical configuration 2 cm long and about 1 cm in diameter. There are a number of smaller pulmonary nodes along the bronchi as they extend into the lungs and also along the trachea. With the exception of the superior region of the left lung and a portion of the middle region of the same lung all lung areas are tributaries of the right latero-tracheal chain (not shown on the figure). The right latero-tracheal chain is located above the bronchus on the trachea and extends onto the forward surface more than the left latero-tracheal chain which is shown on Figure 2. The lymph vessels of the superior region and a portion of the middle region of the left lung end at the left latero-tracheal chain and at the mediastinal lymph nodes located on the forward portions of the aorta and carotid arteries.

The large mass of nodes within the bifurcation receives efferent vessels directly from the lungs or from minor nodes generally located along the bronchi; however, it has efferent vessels leading to the right latero-tracheal chain (right paratracheal). The nodes of greatest interest for counting are the right and left latero-tracheal nodes and the intertracheal bronchial nodes. The mediastinal nodes are located too far away to be counted efficiently. These groups of nodes constitute three irregular shaped groups located in the "arches" of the tracheal bifurcation. A very similar geometry exists in the dog; however, the masses of the node groupings are not as extensive.

A feeling for the physical sizes and geometries is best obtained from the sizes of surrounding structures. The trachea from the bifurcation (carina) to the larynx is about 11 cm long.⁽⁸⁾ In adult males the trachea has a mean diameter of 18.2 mm with a range of 15 to 20 mm. The right bronchus has a diameter of 17.0 mm (range 14-20 mm) and the left bronchus a diameter of 13 mm (range 10-16 mm)⁽⁹⁾ with a mean angle of 56.4° between the major bronchi.⁽¹⁰⁾ The mass of the tracheobronchial glands had a mean value of 16 gm and a range of 10 to 30 gm in nine adult cadavers and showed a positive correlation with age.⁽¹¹⁾

Although it is safe to say that the major mass of the lymph nodes draining the lungs can be found in the region of the bifurcation of the trachea, it is presently impossible to give an accurate assessment of the distribution by mass or size of the various nodes. It is also impossible to obtain a realistic appraisal of the distribution of material from the lungs within these nodes.

Biologic variability and lack of knowledge in this area limit the accuracy of data collected by counting the lymph nodes; however, it will be possible to identify "old" exposures and with reasonable certainty give a general categorization to the amount of exposure.

Esophageal Detectors

In order to count material deposited in the tracheobronchial lymph nodes one must have an X-ray sensitive probe capable of operation at the end of long cables (18-24 inches). The probe must be able to withstand the moist environment of the esophagus with a minimum of risk to the subject. Positioning of the probe is difficult so it must have a uniform response over a fairly long active length (2-5 cm). Since it would be desirable to detect as low as 2 nCi (~ 300 x/m) within the nodes, one must have the highest sensitivity and lowest background possible.

Solid-state avalanche diodes have most of the characteristics desired and are the most promising candidate for this application. An avalanche detector is similar to a conventional diode detector except that surface treatment allows operation of the diode at high voltages. At low bias voltages the diodes operates like a standard semiconductor detector (surface barrier), but with an improved signal-to-noise ratio. As the voltage is increased carrier multiplication occurs resulting in operation similar to a proportional counter. Operation at high voltages (2000 volts) with the resulting charge gain (20 to 100) is possible only because of the surface treatment given the diode. Due to the charge multiplication it is possible to detect photons of a few keV without interference from bulk current generated noise. Typically, the bulk generated noise will be equivalent to 100 keV or less deposited in a non-multiplying diode. With a charge multiplication of 30, a 10 keV photon will have an equivalent energy of 300 keV which is well above the noise.

These diodes are generally low volume detectors. The sensitive depth ranges from 10 to 60 microns with a typical area of 0.07 cm^2 ; however, larger areas (1 cm^2) are currently being produced. The small size limits the photon sensitivity at high energies, but yields a detector suitable for In Vivo studies. Low energy photons (1 keV to 20 keV) are detected with enhanced sensitivity.

The avalanche detector is generally coupled to a voltage sensitive circuit containing one transistor and a tunnel diode.^(12,13) This produces a simple and small detector unit, but one that is sensitive to added capacitance and one that does not put out a signal proportional to deposited energy. If one wants to use large area detectors or long cables, the extreme capacitance sensitivity of the voltage sensitive tunnel diode unit is extremely limiting. Commercial charge-sensitive preamplifiers can be used and this results in

enhanced sensitivity. Measurements show a factor of 2 increase in count rate over the standard tunnel diode unit. The charge-sensitive preamplifier reduces the sensitivity to added capacitance dramatically. (14)

The efficiency of the diodes is limited by the thin depletion depth. With a charge-sensitive preamplifier a detector efficiency of 10% has been measured in agreement with calculations; however, due to non-uniformities in the gain of the detectors across the surface an efficiency of 5 to 10% is typical. Background rates are normally about 1 cpm and in one case an average background rate of 0.255 cpm was observed over a large series of counts.

Detection efficiency near avalanche varies with the voltage on the detector (1.3%/volt) and in turn varies with the temperature of the detector since this changes the avalanche point (2.8%/°C). These are not problems for our application since the detectors are operated in a constant temperature environment and stable high voltage supplies are readily available.

A number of probes have been fabricated for evaluation as esophageal probes some of which are shown in figure 3. We have progressed from small area (0.07 cm^2) multiple diodes to large area (1 cm^2) multiple diodes. It has been necessary to use multiple diode arrangements to obtain a uniform angular response. Figure 4 shows the angular response for a single large area diode ($0.4 \text{ cm} \times 1.0 \text{ cm}$) mounted in a probe. The reduced response at the back of the diode is due to the relatively thick insensitive region at the back of the diode. Using a configuration of three $0.5 \text{ cm} \times 2 \text{ cm}$ diodes mounted in an equilateral triangle array we obtain the angular response shown in figure 5. The sensitivity varies from 0.68 to 1.43 with a normalized value of 1.05. Unfortunately the larger surface area of these diodes is compromised by a lack of uniformity in sensitivity along the 2 cm length. Figure 6 shows scans taken along the length of the three diodes.

Table 1 shows measured sensitivities and background rates for three of six probes which have been evaluated to date. Probe LA-4 has three detectors which are operated separately. These measurements were made in a standard fixture with the source at approximately equal distances from the probe surface. Detector D-1 contains two small area detectors connected within the probe. The loss of sensitivity in detector LA-4 is probably due to the non-uniform response (Figure 6) and it should be noted that the source was located above the dip in sensitivity at the middle.

Table 1. Comparison of Detectors

<u>Detector</u>	<u>Sensitivity*</u> <u>(cpm/μCi)</u>	<u>Background</u> <u>Rate (cpm)</u>
LA-4(A)	35.4	1.05 $\pm$ 0.25
LA-4(B)	71.0	2.05 $\pm$ 0.32
LA-4(C)	48.5	1.95 $\pm$ 0.31
LA-3	126	1.2 $\pm$ 0.5
D-1	94	1.7

*Measured in standard fixture using 4.233 μ Ci source of ^{239}Pu

RESULTS

Dog Counting

Counting experiments have been performed with five dogs. The dogs were anesthetized during the counting and postmortem experiments have been performed on four of the dogs. These dogs were particularly suited for this experiment since they had received large inhalation exposures to only one lung with subsequent removal of the exposed lung in some animals. This meant that in selected animals the primary body burden consisted of material translocated to the lymph

nodes from the exposed lung.

Two probes were used in these experiments, D-1 and LA-3, (Table 1). A commercial charge-sensitive preamplifier and commercial amplifiers and counters were used during the experiments. Detector D-1 operated at 2390 volts and detector LA-3 operated at 1800 volts while in the dog compared to room temperature operation of 2270 volts and 1720 volts, respectively.

Figure 7 shows a lateral X-ray of a dog with detector D-1 positioned at the point of maximum count rate. Figure 8 shows a series of counts taken as the probe (LA-3) was moved down the esophagus of the same dog. Figure 9 shows results from a series of counts taken around the area of maximum count rate. The exposed lung of this dog had been removed and it is felt that the maximum count rate corresponds to lymph nodes at the carina.

A definite variation in count rate with position was noted for each dog counted with count rates as high as 1570 counts per minute. Three of the dogs counted show a relatively broad peak in the count rate. Postmortem observation and counting experiments on these dogs indicated that we were counting not only the material in the tracheobronchial lymph nodes, but also material in the lungs. The exposed lung of these dogs, which had not been removed, had become non-functional and each lobe had shrunk to approximately the size of a golf ball and was resting near or against the esophagus. Counting experiments with the thorax of the dog opened indicated that we could pick out the counts due to the tracheobronchial lymph nodes. These experiments also helped in delineating the geometries involved and in interpretation of X-rays.

Analysis of data on the tissue distribution and amount of plutonium in one dog (248), indicate an MDA of 26 nCi for a one minute count. This dog was counted with probe D-1 with a sensitivity of about 76 counts/minute per microcurie. Our MDA is based on counts from the right tracheobronchial (latero-tracheal) nodes since X-rays and counting data indicate that this was the area from which counts were received at the maximum count rate. This probe is not as sensitive as later models so even lower MDA's are now possible. Experiments

after euthanization of the dog yielded count rates of 753 counts/minute per microcurie for the right tracheobronchial node with the node pressed against the most sensitive detector. This gives an indication of count rate loss due to geometry.

Phantom Results

In order to evaluate experimentally the results expected with the probe on humans, a phantom was constructed. The phantom was constructed using available information concerning the geometry and consists of an esophagus and tracheobronchial tree fabricated of silicon rubber. Plutonium sources of equal activity are placed at the carina and on each side of the trachea to simulate masses of lymph nodes. This entire structure is suspended in a temperature controlled water bath to simulate the operating conditions expected during evaluation of lymph node burdens. Although absorption of the L X-rays resulting from the decay of plutonium is greater in silicon rubber than in tissue equivalent plastic ($T_{1/2} = 0.25$ cm vs $T_{1/2} = 0.8$ cm.), this difference was ignored because of the small thicknesses involved. For an accurate calibration this could not be ignored; however, during technique evaluation this results in conservative estimates of detectable limits.

Scans in the phantom with detector LA-3 yielded variations similar to our dog counting results. For the peak count rate at the carina a sensitivity of 60 counts/minute per microcurie was obtained. This would yield an MDA of about 7 nCi for a 20 minute count. If the total activity in the phantom were pooled and used with the peak count rate, a sensitivity of 20 counts/minute/microcurie would result with an MDA of 22 nCi.

Figure 10 shows scans made in the phantom with probe LA-4. In this case only two of the probes were used in an optimum orientation resulting in a sensitivity of 74 counts/minute per microcurie for

an MDA of 8.5 nCi. This is at an 84% confidence level with a counting time of 20 minutes.

The scans show peaks corresponding to the position of "carina" source and the "latero-tracheo" sources. When a stiff cable is used, the probe pushes away from the tracheal side of the esophagus causing a reduction in count rate. In a biological system this effect would not be as important since the esophagus is not as free to distend away from the trachea. Figure 11 shows a similar scan taken with LA-4 using only one detector (B) in a plane configuration similar to LA-3.

CONCLUSIONS

Experimental results have shown that it is possible to detect plutonium translocated to the tracheobronchial lymph nodes. This is accomplished with common modular counting equipment and a detector probe. Extensive radiation shielding is not needed and counts can be performed on unanesthetized cooperative subjects. Selective counting of the lymph node burdens is obtained because of the geometry involved.

The avalanche probe is not meant to replace whole body counting techniques for plutonium. However, it is expected that it can be used as a complementary technique thus allowing screening of individuals for lymph node burdens and for a rough estimate of the amount deposited in the lymph nodes.

Developmental work is continuing on the probes and it is expected that enhanced sensitivity along with more uniform response will be obtained with future probes. Complementary techniques for detection of lymph node burdens are also under investigation such as fiber optic coupled scintillators.

REFERENCES

1. K. L. Swinth, "Measurement of the Photon to Alpha Ratio for Plutonium Isotopes," Nucl. in Aerospace, edited by Paul Polishuk, Plenum Press, New York, 1968, p. 279.
2. K. L. Swinth, "The Measurement and Use of X/ α Ratios For Transuranic Alpha Emitters," Presented at Annual Meeting of the Health Physics Society, Chicago, Ill., June 30, 1970.
3. T. Rundo, K. Rudram and B. T. Taylor, Health Physics, 17, 155 (1969).
4. K. L. Swinth and B. I. Griffin, Health Physics, 19, 543(1970).
5. H. Rouvière, Anatomy of the Human Lymphatic System translated from Anatomie des Lymphatiques de L'Homme by M.J. Tobias Edwards Brothers, Inc. Ann Arbor, 1438, p. 288.
6. See pp. 113-118 in Reference 5 above.
7. Henry Gray, "Anatomy of the Human Body" edited by C. M. Goss. Lea & Febiger, Philadelphia, 1966, pp. 754-759.
8. See p. 1137 in Reference 7 above.
9. M. Lippman and R. E. Albert, Amer. Indust. Hyg. Assoc. J. (May-June, 1969) p. 257.
10. M. Alavi, T.E. Keats and W. M. O'brien. Radiology, 108, p. 546 (1970).

11. E.E. Pochin, Health Physics, 12, p. 563, (1966).
12. R. J. Locker, IEEE Trans. Nucl. Sci. NS-13, 382 (1966).
13. R. J. Locker and G. C. Huth, Appl. Physics Letters 9, 227 (1966).
14. P. V. Hewka, G. C. Huth, and K. L. Swinth, IEEE Trans. Nucl. Sci. NS-17, 265, (1970).

Figure Captions

Figure 1: Retention and translocation of alveolar - deposited $^{239}\text{Pu O}_2$ in dogs.

Figure 2: Observed geometry of lymph nodes near the bifurcation of the trachea.

Figure 3: Photograph showing four probes developed for the detection of plutonium in the tracheobronchial lymph nodes.

Figure 4: Angular response of single large area avalanche diode in probe.

Figure 5: Angular response of probe containing three 0.5 cm x 1.0 cm detectors in an equilateral triangle geometry.

Figure 6: Response along the length of the three detectors in probe LA-4.

Figure 7: X-ray showing probe D-1 in the position of maximum count rate.

Figure 8: Count rate vs position for probe LA-3 in a dog.

Figure 9: Variation in count rate near position of maximum count rate in a dog.

Figure 10: Scans made in a human thorax phantom showing increase in count rate near lymph node sources.

Figure 11: Scans made in a human thorax phantom showing increase in count rate near lymph node sources.

Figure 1 - Retention and translocation of alveolar-deposited $^{239}\text{PuO}_2$ in dogs.

ANGULAR SENSITIVITY OF 4 BY 10 mm AVALANCHE DETECTOR

ANGULAR RESPONSE OF 3 ELEMENT PROBE

3

0

1

2

9