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COUNTING PLUTONIUM IN THE TRACHEOBRONCHIAL LYMPH NODES*

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Abstract—A series of intraesophageal probes containing avalanche diode detectors were developed for detecting plutonium translocated to the tracheobronchial lymph nodes. Experiments with dogs having body burdens of plutonium yielded an average sensitivity of 67 cpm/μCi for the middle tracheobronchial node and 124 cpm/μCi for the right tracheobronchial node. Experiments with a human thoracic phantom resulted in sensitivities of 30–60 cpm/μCi for the middle tracheobronchial node. Two standard deviations of the background for a 20 min count correspond to 8 nCi under optimum counting conditions with the phantom.

INTRODUCTION

A PRINCIPAL route of internal contamination for workers exposed to plutonium is via the respiratory tract from which the plutonium will be translocated to other portions of the body including the thoracic lymph nodes.[1] Research at Battelle conducted on dogs has shown that up to 50% of the alveolar deposited plutonium may be translocated to thoracic lymph nodes resulting in concentrations 50–100 times that in the lung.[2] Due to the radiotoxicity of plutonium it is desirable to obtain an accurate assessment of the quantity and location of *in vivo* deposited plutonium. This paper describes the application of prototype detectors in a preliminary investigation of the detection of plutonium translocated to the tracheobronchial lymph nodes. Following the development of suitable probes, experiments were performed on dogs and on a phantom representing the thoracic area of a human.

Measurement of plutonium *in vivo* is performed by counting the *L* X-rays of the uranium daughter and radiations from ^{241}Am, the daughter of ^{241}Pu. These X-rays (13.6, 17.0 and 20.2 keV) plus the 60 keV gamma of ^{241}Am are of low intensity[3,4] and rapidly attenuated by tissue.[5] Typically, fewer than 30 X-rays per min will escape from the body of a subject with alveolar deposited plutonium representing one Maximum Permissible Lung Burden (16 nCi).

Following inhalation of plutonium dioxide, alveolar deposited plutonium will translocate to other tissues including the tracheobronchial lymph nodes. After 8–10 yr in dogs, 40–50% of the plutonium has been translocated to the thoracic lymph nodes, 10–15% to the liver, 5% to the skeleton and 5% to the abdominal lymph nodes. About 15% is left in the lungs.[2]

Due to the rapid attentuation of the X-rays and 60 keV gammas associated with plutonium, the redistribution will affect the sensitivity of external measurements of the plutonium content of the body.[6,7] This is further complicated by the differential translocation of ^{241}Am and ^{239}Pu.[8] Body variations, differential translocation and low count rates make it impractical to correct for sensitivity of external counting,[9] or to determine the relative distribution of the translocated material.

DESCRIPTION OF PROBES AND COUNTING GEOMETRY

THE TRACHEOBRONCHIAL GEOMETRY

Figure 1 shows a typical geometry of the tracheobronchial area observed on one human cadaver. The nodes in the angle of the bifurcation extended 4 cm along the right bronchus and 2 cm along the left bronchus. Other nodes

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REPOSITORY PNL
COLLECTION Intraesophageal
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FOLDER HSC 73-5

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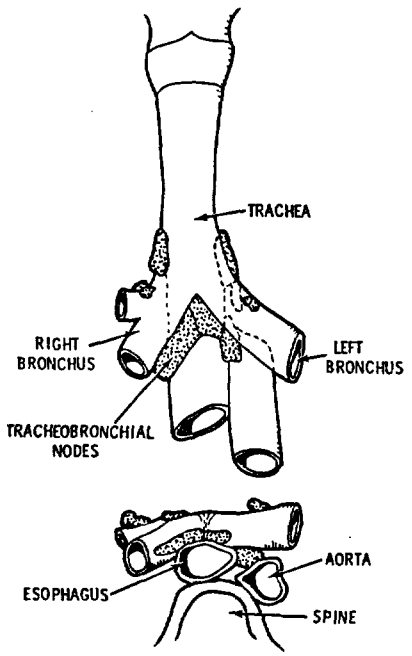

FIG. 1. Observed geometry of lymph nodes near the bifurcation of the trachea on a human cadaver. This illustrates a typical geometry, but is not meant to represent the average individual.

are observed along the sides of the trachea above the tracheal bifurcation and are shown approximately to scale. There are a number of smaller pulmonary nodes along the bronchi as they extend into the lungs and also along the trachea.

Nodes near the bifurcation receive afferent lymphatics directly from the lungs or from minor nodes generally located along the bronchi; however, the superior region of the left lung drains into the mediastinal nodes located on the forward portions of the aorta and carotid arteries.[10] The right and left laterotracheal and intertracheal–bronchial node groups form three irregularly shaped masses accessible for counting by a probe placed in the esophagus. Mediastinal and pulmonary nodes which also drain the lungs are located too far away from the esophagus to be counted efficiently.

The geometry observed in dogs is similar, but less complex.[11,12] Mediastinal and tracheobronchial lymph nodes are present but fewer "accessory" nodes such as the bronchopulmonary nodes are observed. The node groups are smaller and more uniform than one observes in humans[13] and have a mass of 0.548 ± 0.016 g.[11] Measurements by POCHIN[14] indicate an average mass of 15 g for the tracheobronchial nodes in humans with a range of 10–30 g.

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 accurately assess the distribution by mass or size of the various nodes in humans;[15] however, this data is available for the Beagle.[11,12] It is difficult to judge the relative distribution among nodes of material translocated from the lungs; however, data from dogs indicate a fairly uniform distribution of the material among the tracheobronchial nodes on the basis of concentration. Since the intertracheobronchial node is generally the largest, the greatest activity should be found at this point.

Esophageal detectors

To detect plutonium in the tracheobronchial lymph nodes at low levels (2 nCi), one must have a detector capable of meeting the following requirements:

—Detect X-rays at the end of a flexible 45–60 cm cable
—Withstand the moist environment of the esophagus
—Have uniform sensitivity over a cylindrical volume of about 1 cm diameter × 2–5 cm long
—Present no hazard to the subject
—Operate at body temperatures
—Have small overall size and easy to swallow
—Have low background count rate
—Have high sensitivity to 13–20 keV X-rays
—Have low sensitivity to or capability to discriminate against 60 keV gammas from ^{241}Am.

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 with a resulting charge gain of 20–100. At low bias

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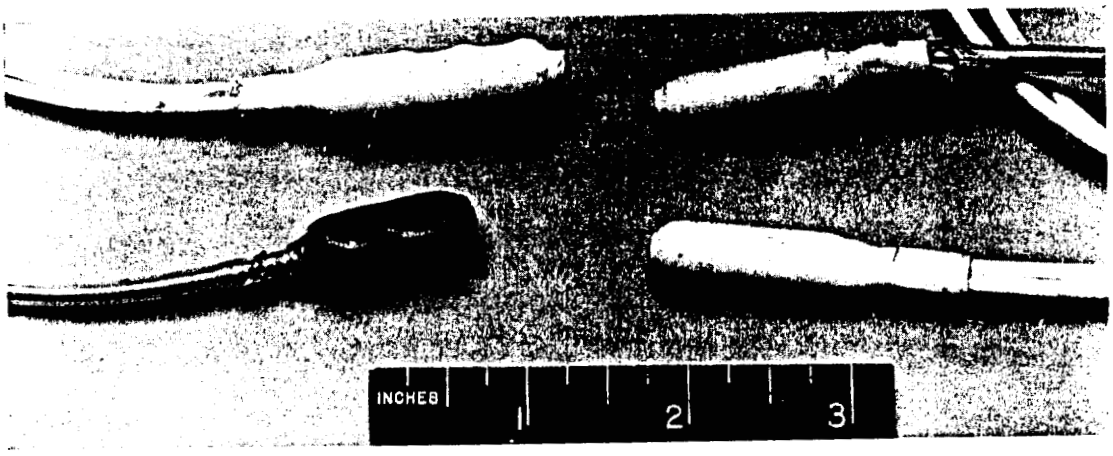

FIG. 2. Photograph showing four probes developed for the detection of plutonium in the tracheobronchial lymph nodes.

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voltages the diodes operate 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. 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 nonmultiplying 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.

Avalanche detector performance can be maintained with the diode at the end of long cables if an ordinary charge-sensitive preamplifier is employed.[16] Although the X-ray counting efficiency is lower than desired (5–10%), the diodes have low background rates (~ 1 cpm) which help to compensate for the low sensitivity. The efficiency drops rapidly at higher energies, thus effectively discriminating against the 60 keV gammas of ^{241}Am. Discrimination is important because of the possible differential translocation of ^{241}Am and plutonium[8] and to maintain good spatial discrimination.

Increasing temperatures affect the noise levels of avalanche diodes; however, the increase is not detrimental if the diode gain is large enough. Efficiency of the diodes varies with applied voltage (1.3% per V) and in turn varies with temperature which will change the avalanche point (2.8% per °C). With the constant temperature environment of the body and a stable high voltage supply, the gain can be held constant.

Encapsulation of the diode in a probe plus a rubber sheath protects the diodes from the moist esophagus and in turn protects the subject from the high voltages on the diode.

A number of probes have been fabricated for evaluation as esophageal probes, some of which are shown in Fig. 2. 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 arrays to obtain a uniform angular response.[17] The diodes are flat planes and at angles approaching 90° from a perpendicular to the plane, the sensitivity falls rapidly. Using a configuration of three 0.5 × 2 cm diodes mounted in an equilateral triangle array we obtain the angular response shown in Fig. 3. 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.

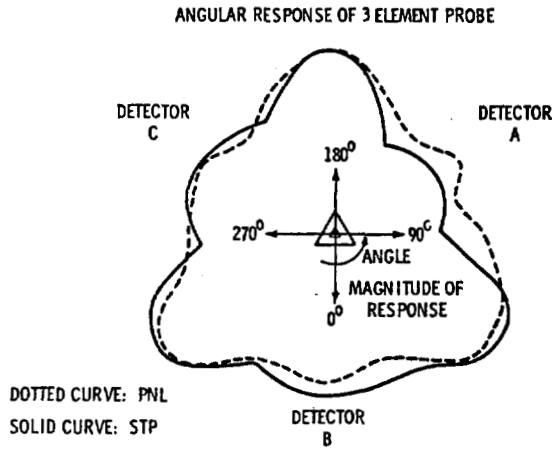

FIG. 3. Angular response of probe containing three 0.5 × 1.0 cm detectors in an equilateral triangle geometry. This illustrates the uniformity of response obtained. STP refers to measurements made by General Electric while PNL refers to independent measurements at Battelle.

Table 1 shows measured sensitivities and background rates for three of six probes that 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 a nonuniform linear response.

Table 1. Comparison of probes

Probe (detector)	Sensitivity* (cpm/μCi)	Background rate (cpm)
LA-4(A)	35.4	1.05 ± 0.25
LA-4(B)	71.0	2.05 ± 0.32
LA-4(C)	48.5	1.95 ± 0.31
D-1	94	1.7
LA-3	126.0	1.2 ± 0.5

* Measured in standard fixture using 4.233 μCi source of ^{239}Pu.

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RESULTS

Dog counting

Counting experiments have been performed with 5 dogs including counts with anesthetized dogs and with euthanized dogs. Two of 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. This meant that 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 V and detector LA-3 operated at 1800 V while in the dog, compared to room temperature operation of 2270 and 1720 V, respectively.

Figure 4 shows a series of counts taken as the probe (LA-3) was moved down the esophagus of an anesthetized dog. The exposed lung of this dog had been removed and the maximum count rate corresponds to the location of the tracheobronchial lymph nodes. The shoulder of the curve appears to be from the right tracheobronchial node and the peak from the middle node. This interpretation agrees with X-rays; however, a post-mortem experiment was not performed for verification. Experiments were repeated with consistent results and significant count rates were localized to the area of the tracheobronchial nodes.

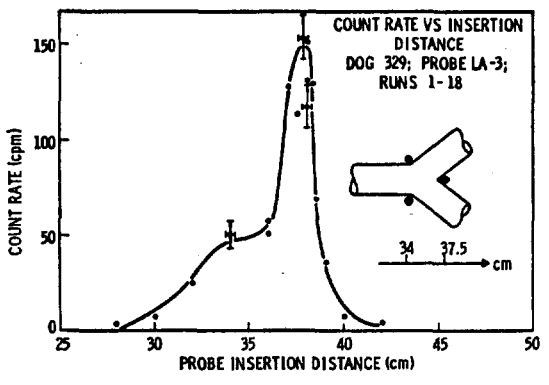

FIG. 4. Count rate vs position for probe LA-3 in a dog. The sketch shows the assumed positions.

Table 2. Counting sensitivity for tracheobronchial lymph nodes in dogs based on post-mortem analysis of tissues

Dog	Counting sensitivity (cpm/μCi) Middle T–B	Right T–B
329	48.6 ± 4.2	300‡
	47.5 ± 4.5†	
248		168 ± 10†
335*	66.3 ± 4.5	74 ± 11.5
344*	85.1 ± 5.5	129 ± 20
Unweighted average	66.7	124 (without 329)

* Post mortem experiment.

† Counts with probe D-1 normalized to probe LA-3.

‡ Count rate estimated from Fig. 4.

Table 2 summarizes data obtained on the dogs. With the extremely high concentrations of plutonium found in the nodes, one must consider the possibility of self absorption of the X-rays. A rough calculation assuming the plutonium was uniformly distributed, indicated this effect should be less than 10%. The variation appears to be due primarily to the lymph node-detector geometry. Sensitivity is consistently higher for the right tracheobronchial node. With a background of one cpm and the average sensitivities listed in Table 2, the Minimum Detectable Amounts (MDA) at the 84% confidence level (approximately 2σ) are 30 and 16 nCi for the middle and right nodes, respectively, for 1 min counts.

Human phantom results

To experimentally evaluate the results expected when using the probe on humans, a phantom was constructed using available information concerning the tracheobronchial geometry. It consists of an esophagus and tracheobronchial tree fabricated of silicon rubber and with plutonium sources of equal activity placed at the carina and on each side of the trachea to simulate the 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. The absorption of the *L* X-rays resulting from the decay of plutonium is greater in silicon rubber than in tissue

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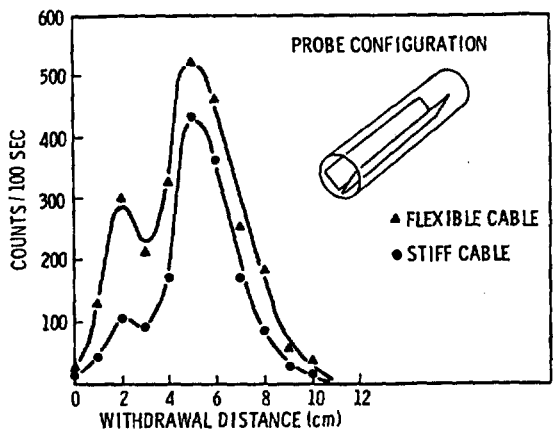

FIG. 5. Scans made in a human thorax phantom showing increase in count rate near lymph node sources with an optimum detector configuration. The inter-tracheobronchial source is at the 2 cm position and the laterotracheal sources are at 5 cm.

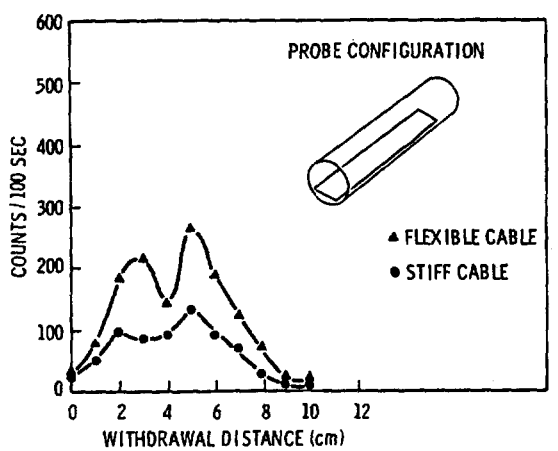

FIG. 6. Scans made in a human thorax phantom showing increase in count rate near lymph node sources for a plane detector configuration. The inter-tracheobronchial source is at the 2 cm position and the laterotracheal sources are at 5 cm.

equivalent plastic ($T_{1/2} = 0.25$ cm vs. $T_{1/2} = 0.6$ cm), but this difference was ignored because of the small thicknesses involved and the preliminary nature of the experiment.

Figure 5 shows scans made in the phantom with probe LA-4. The probe was operated in an optimum orientation taking advantage of the multiple detector array. The scans show a small peak corresponding to the position of the middle group of nodes at the carina and a larger peak for the laterotracheal 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 6 shows a similar scan taken with LA-4 using only one detector (B) in a plane configuration similar to LA-3.

Table 3 summarizes the peak sensitivities obtained with probes LA-4 and LA-3 in the phantom.

Both the highest and lowest sensitivities were obtained with probe LA-3. This probe had the poorest geometrical configuration and the highest efficiency (Table 1); therefore, the results are not unexpected. Probe LA-4 in the optimum orientation had uniform sensitivity for the nodes at the carina and at the laterotracheo position as desired. With this probe the MDA for a 20 min count is about 15 nCi at the 84% confidence level. Under optimum conditions

Table 3. Peak sensitivities for probes LA-3 *and* LA-4 *for counts taken in the thoracic phantom*

Probe	Sensitivity (cpm/μCi) Middle T–B (Carina)	Laterotracheal nodes
LA-4 Optimum orientation (Fig. 5)	42.6	40
LA-4 Plane orientation (Fig. 6)	31.2	18.9
LA-3	61.1	3.9

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with probe LA-3, the MDA is approximately 8 nCi.

For *in vivo* applications where the count rate cannot be used for probe positioning, it would be necessary to take appropriate radiographs to verify the position of the probe. Development of a longer more uniform probe should produce a uniform sensitivity. This would reduce the need for extremely accurate positioning.

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. Although swallowing a probe is not a comfortable task, such procedures are performed routinely in medicine. 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 plutonium in the lymph nodes are also under investigation such as fiber optic coupled scintillators.

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