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Center for Human Radiobiology

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FOREWORD

The papers in this report deal with several different aspects of the Center's studies of the effects of internally deposited radionuclides in humans: experimental and theoretical research on the induction of tumors by alpha-particle irradiation (papers 1–4); medical effects (papers 5–8); metabolism and dosimetry (papers 9–17); methods for the determination of radioactivity (papers 18–25); and processing of data (papers 26–27).

The reader's attention is directed to paper 1, which reports that the mean lethal dose for mammalian cells corresponded to about 27 alpha particles per cell nucleus when cultured cells were irradiated with 5.6 MeV alpha particles. This finding casts doubt on the common supposition that a single alpha particle through the nucleus is sufficient to kill a cell in situ. Also of special interest is the report on a case of internal contamination with ^{241}Am plated on silver for use in smoke detectors (paper 16). Two disks, accidentally swallowed in the same incident, were found to have lost less than 1% of the americium in passing through the gastrointestinal tract.

Exposure data for persons whose body burdens of radium have been measured are given in Appendix A at the back of this report. As was the case in our previous Annual Report (ANL-75-60, Part II), tumor cases considered to have radium-induced malignancies are listed in Appendix B for persons whose body burdens of radium have been measured: bone sarcomas in Table 1 and carcinomas of the paranasal sinuses and mastoid air cells in Table 2. To these we have now added lists of "probable or confirmed" malignancies of the same types in persons known to have been exposed to radium, but whose body burdens have not been determined (Appendix B, Tables 3 and 4). These are mostly persons who died before population studies of radium-exposed persons were initiated in the 1950's. The cases of suspected radium-induced malignancy were identified by careful review of case histories for any indication of bone sarcoma or carcinoma of the sinuses or mastoids. This review included all persons for whom any medical data were available at the end of 1975.

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AN UNUSUAL CASE OF INTERNAL CONTAMINATION WITH ^{241}Am

J. Rundo, M. Essling, D. R. Huff, and W. D. Fairman*

A woman accidentally swallowed two sources of ^{241}Am of the type used in domestic smoke detectors; the nominal activity of each source was 2.5 μCi, and they were not voided until the 16th and 24th days after intake. A little less than 1% of the total activity of the two sources was found in the fecal material after removal of the sources and ^{241}Am was detected in the urine until the second source had been voided.

Introduction

A female employee of a local electronics firm engaged in the manufacture of domestic smoke detectors containing ^{241}Am, was referred to us by the Nuclear Regulatory Commission's Compliance Division for investigation of possible radioactive content. She was thought to have swallowed a source of ^{241}Am (nominal activity 2.5 μCi), deposited on a silver disk 2.35 mm in diameter and 0.2 mm thick, and coated with a very thin layer of gold. Radiographs of the abdominal region 11 days after the putative intake had failed to reveal the presence of such a radiopaque object, and it was thought that it had been voided in the feces. Our help was requested in order to confirm this belief. Urine and feces collected at a local hospital on day 11 were sent to ANL for assay of ^{241}Am by the Bioassay Group of OHS Division, ANL.

Body Radioactivity Measurements

The subject (CHR case number 30-063) came for body radioactivity measurement on day 14. A measurement in the tilted chair revealed the presence of a substantial amount of ^{241}Am. An attempt was made to localize this from a longitudinal profile scan taken with a 1-cm wide slit-collimated detector. Because the subject complained of having had a sensation of "something stuck in the throat," the scan covered the interval 15 to 80 cm from the vertex. Her height was 156 cm. The profile curve, shown in Figure 1, revealed no

*Occupational Health and Safety Division, ANL.

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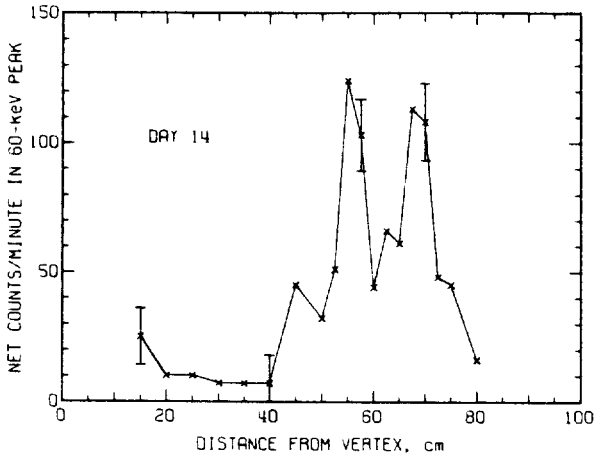

FIG. 1.--Radioactive profile curve (slit-collimated 29-cm diameter detector), showing the presence of two sources in the abdomen (case 30-063).

significant excess activity in the pharyngeal region, but two sharp peaks, centered at about 56 cm and 68 cm from the vertex, demonstrated the presence of two sources in the abdomen.

Measurements were also made in arc geometry, to establish the total content. The interpretation of the counting data posed an unusual problem because of the attenuation of the 60-keV gamma rays in the silver foil, and of the impossibility of knowing the direction(s) that the sources were facing. Thus, with a mass attenuation coefficient of 5.49 $cm^2\ g^{-1}$ for 60-keV gamma rays in silver of density 10.5 g cm^{-3}, the intensity of the photons from the gold-coated side (attenuation ignored) should be 3.16 times the intensity from the other side (silver thickness 0.02 cm). The observed ratio of the counting rates from front and back of the subject in the 1.5-m arc was 3.7 ± 0.1; this suggested that both sources were facing in the same direction, i.e., gold-coated side forward, since the higher counting rate was observed from the front of the subject. However, it is entirely possible for a ratio of 3.7 to be observed from a source of ^{241}Am in vivo if it is about 6 cm deep in a 20.4-cm thick abdomen, and is not backed by severely attenuating material. In the absence of other information, we assumed that the sources were both close to the midplane of the abdomen. In making the calculation of the content, R, we used the following equation:

$$R = \sqrt{3.16\ N_1 N_2} \times \left(\frac{D-d}{D}\right)^2 \times e^{\mu d} \times f_{fs} \times \frac{S}{N_3}\ \mu Ci\ , \qquad (1)$$

where 3.16 is the calculated counting-rate ratio for the disk sources,

N_1 is the counting-rate (min^{-1}) from the front of the subject,

N_2 is the counting-rate (min^{-1}) from the back of the subject,

D is the radius of the arc (150 cm),

d is the half-thickness of the abdomen (10.2 cm),

μ is the measured linear attenuation coefficient of 60-keV γ rays in tissue-equivalent material (0.15 cm^{-1}),

f_{fs} is a forward scatter correction (0.763),

S is the activity of the standard source, and

N_3 is the counting-rate (min^{-1}) from the standard source counted at 150 cm.

We observed values of 750 ± 9 and 203 ● 5 counts/min for N_1 and N_2 respectively, S was 9.8 μCi and N_3 was 4472 counts/min. Inserting these and the other values in Eq. 1, we obtain

$$R = 4.65\ \mu Ci\ .$$

The factor of 3.16 enters the equation for R to correct the counting rate for attenuation in the silver. The value of 4.65 μCi for R is in reasonable agreement with the expected value of 5.0 μCi for two sources.

On day 16, a fecal sample was voided and when examined by gamma-ray spectrometry, it was found to contain a substantial amount of ^{241}Am. We shall discuss the examination of this and of a subsequent sample below.

A second determination of body radioactivity was made on day 21. ^{241}Am was still present and measurements were again made in arc geometry; time did not permit localization measurements. As on day 14, the counting rate from the front (432 ± 7 min^{-1}) was much greater than that from the back (138 ± 4 min^{-1}); the ratio was 3.1 ± 0.1. From Eq. 1, we obtain

$$R = 2.72\ \mu Ci\ .$$

This second source was found in feces voided on day 24, no other samples having been voided in the interim, and a third examination of body radioactivity on day 28 revealed no detectable content of ^{241}Am.

The two fecal samples were radiographed for us by the Non-Destructive Testing Group of the Quality Assurance Division (ANL), and each was seen to

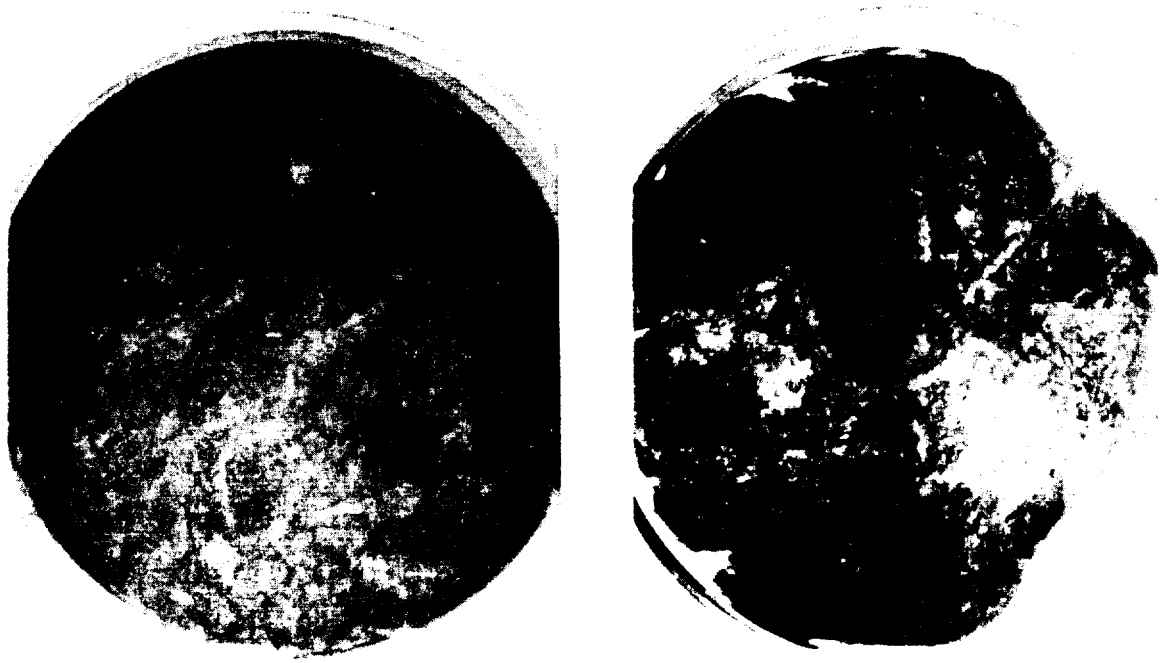

FIG. 2.--Prints made from radiographs of fecal samples (in plastic bags inside waxed cardboard cartons) voided 16 days (left) and 24 days after the incident. The silver disks are easily seen. (ANL neg. 235-76-21)

contain a small disk of a dense material. Prints from the radiographs are shown in Figure 2. In the sample voided on day 16, the disk is seen almost in plan view, and it presents as an incomplete circle with a short straight edge. The shadow of the disk in the sample voided on day 24 is seen as an eccentric ellipse. Each disk was isolated from the feces, washed in a little water, dried and mounted in a cardboard slide mount between two sheets of laminator film* 0.04 mm thick. The activities of the two sources were estimated to be 1.86 ± 0.02 μCi (day 16) and 2.36 ± 0.02 μCi (day 24). The water used to wash the sources contained no detectable activity (limit 100 pCi).

Apparently Eq. 1 gave overestimates of the contents of the subject on both occasions, and the principal reason for this is the use of the calculated ratio, 3.16, of the counting rates from the two sides of the sources. After isolation from the feces, this ratio was determined to be 2.1, indicating that the silver foil may have been thinner than 0.2 mm, or that the mass attenuation coefficient was smaller than 5.49 $cm^2\ g^{-1}$, or both. In retrospect, we can

*Ply-On, available from American Photocopy Co., Evanston, Ill.

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correct the estimates of body content by the ratio $\sqrt{2.1/3.16} = 0.815$, obtaining 3.8 μCi and 2.2 μCi for days 14 and 21, respectively. These are in slightly better agreement with the known activities of the two sources, and the validity of Eq. 1 is established.

Excretion Analysis

Following the subject's visit to ANL on day 14, she was asked to collect and submit all urine and feces voided. Most urine voided between days 14 and 31 was received and analyzed for ^{241}Am, as was the 550-ml sample collected in hospital on day 11. Samples voided from day 23 onward contained no detectable ^{241}Am (limit 0.1 pCi); the results for the other samples are set out in Table 1. Not all samples represented 24-hr excretions. The subject voided feces rather infrequently (usually about once a week). Samples were voided on days 11 (in hospital), 16, and 24. At this time, she was treated with cathartics which caused diarrhea for several days, and she did not collect these samples. Two other fecal samples voided on days 30 and 31 were received. The results of the analyses for ^{241}Am are set out in Table 2. The samples voided on days 16 and 24 were the ones which contained the active disks, and the amounts of activity reported in Table 2 were found in the feces after removal of the disks. The total activity recovered in feces, 34.7 nCi, amounted to about 0.8% of the activity on the two disks (4.22 μCi). A little more may have been voided during the period of diarrhea.

The presence of detectable ^{241}Am in the urine until day 23 proved that there was some entry into blood, but the fall below the limit of detection on day 24 showed that the systemic content must have been very small. A very approximate upper limit for the intake to the blood can be calculated if we make the pessimistic assumptions that the activity entered the blood on day 1, that the urinary excretion rate on day 24 was equal to the limit of detection (0.1 pCi/day), and that the excretion rate followed Langham's power function equation for plutonium. With these assumptions, a value of 525 pCi is calculated, and this must be an extreme upper limit. It is negligible from the point of view of radiological protection. It is obvious from the results in

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Table 1. Urine samples and their contents of ^{241}Am

Days since intake	Collection period, hr	^{241}Am content of sample, pCi ± 2σ
11	?	0.18 ± 0.04
14—15	13	0.22 ± 0.05
15—16	24	0.22 ± 0.04
16—17	24	1.3 ± 0.2
17—18	24	0.31 ± 0.06
20—21	19	<0.1
21—22	21.5	0.35 ± 0.06
22—23	24	0.23 ± 0.04

Table 2. Contents of ^{241}Am in five fecal samples

Days since intake	^{241}Am content of sample, pCi ± 2σ
11	194 ± 32
16	17,700 ± 2900
24	16,800 ± 2800
30	≤1.8
31	≤0.9

Table 1 that the urinary excretion of ^{241}Am was being supported by the continued presence of the second source in the gastrointestinal tract, and it fell below the limit of detection in the 24-hr sample excreted just before the source was voided in the feces. The actual initial systemic burden must therefore have been much lower than 525 pCi.

Finally, it may be noted that a little more ^{241}Am was lost from the body than was accounted for in the feces, because about 2 nCi were found in the

trousers of the pajamas worn by the subject when the body radioactivity was investigated on day 14. This was attributed to contamination from the anal region, and other clothing worn between days 11 and 24 may have been similarly contaminated.

Conclusions

A most unusual feature of this case was the long delay between intake and voiding of the sources. They were not in the abdomen on day 11 (radiographic evidence) but they were there three days later (Figure 1). It was suggested by a consulting physician that they may have been trapped in the vallecula or the pyriform sinus, or elsewhere in the pharynx. This would account for the holdup and for the reported discomfort in the throat. We can only speculate on whether the sources left the pharyngeal region together or separately. In any event, it seems that the first source was voided in the feces with little delay after entry into the stomach.

If the sources of ^{241}Am involved in this incident are representative of those incorporated in the domestic smoke detectors, then the most important conclusion that can be drawn is that they are remarkably secure. Apparently they lose very little (less than 1% in the present case) of their radioactive content when exposed to body fluids (although not necessarily gastric juices all the time) for 16 days or more. Furthermore, what activity is released under these circumstances is so inert that there is negligible absorption into the blood. This is an important and comforting finding in view of the growing numbers of smoke detectors available to the general public.

Another important conclusion, but in a different context, is the validation of Eq. 1 for the calculation of the body content. A slightly different version of the equation is used to calculate the body content of radium daughters, and this was the first opportunity we have had to determine directly the validity of this approach.

Acknowledgements

We thank Robert L. Irwin and Joseph W. Lucas of the Quality Assurance Division for making the radiographs of the fecal samples. Fred S. Iwami,

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Theresa L. McFall, and Jean J. Robinson of the Occupational Health and Safety Division made the measurements of ^{241}Am in the excreta.

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