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Center for Human Radiobiology

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FOREWORD

Vigorous efforts to elucidate the mechanisms and dosimetry of radium-induced malignancies continued during the past year. Paper 1 marshalls evidence that, contrary to previous assumptions, ^{228}Ra may have dosimetric significance comparable to that of ^{226}Ra for the induction of carcinomas of the sinus and mastoid epithelia. This paper also raises the possibility that ^{224}Ra and actinides that emit high-energy alpha particles may be capable of inducing sinus or mastoid carcinomas. Paper 4 points out that the cells at risk for induction of bone tumors may be at some distance from bone surfaces and suggests that the entire range of alpha particles from bone-deposited radionuclides should be included in the calculation of dose, instead of the commonly used 0 to 10 microns from bone surfaces. The growth of osteosarcoma cells in culture was found to be less inhibited by alpha-particle irradiation than was the growth of normal cells (paper 5).

Paper 12 is an abstract of a report that higher than expected rates of breast cancer have been found among women radium dial painters. The differences were correlated with radium intake, but it is not yet established whether the pertinent dosimetry involves internally-deposited radium, absorbed radon, or external radiation in the work rooms. Health findings and radioactivity measurements in our study of another occupational group, former thorium workers, are summarized in paper 14.

Radioactivity studies of individuals or small groups of persons exposed to various radionuclides are reported in papers 17, 18, and 20-25. Among these it may be noted that excess fallout radioactivity was not detected in former military personnel who participated in the "Smoky" nuclear test of 1957 (paper 22). Higher than normal amounts of ^{210}Pb in urine were found for residents of houses with high levels of radon (paper 17) and for people who work at the site of a former uranium mill (paper 18). A model to estimate exposure to radon and radon daughters from ^{210}Pb excretion rates is being developed (paper 17).

Among the papers in this Annual Report are a description of our system for coding medical information from case records (paper 13) and a massive compilation of osteometric data (paper 33).

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HEALTH STATUS AND BODY RADIOACTIVITY OF FORMER THORIUM WORKERS*

A. F. Stehney, A. P. Polednak,† J. Rundo, A. M. Brues, H. F. Lucas, Jr., B. C. Patten, and R. E. Rowland

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This is a progress report of a study of the health effects of industrial exposure to thorium. Included is the three-year period from inception (1976) to September, 1979. The study population comprises the former employees of a thorium mill located in West Chicago, Illinois. Thorium and rare earth chemicals were extracted and purified from monazite ores at this plant from 1932 to 1973, and thorium mantles, for gas lamps, were manufactured from 1936 to 1947.

The objectives of the study are: (1) to assess possible health effects of employment in the thorium milling industry by comparison of mortality and morbidity characteristics of former thorium workers with those of suitable general populations; (2) to examine disease outcomes by estimated exposure levels of thorium and thoron daughter products for possible radiation-related effects; and (3) to determine the body distribution of inhaled thorium (and daughters) and rare earths in humans by radioactivity measurements in vivo and by analysis of autopsy samples. The principal end points for investigation are respiratory disease and cancers of lung, liver, bone, and bone marrow.

The West Chicago plant was operated by the Lindsay Light Company and its corporate successors, which finally included the Kerr-McGee Chemical Corporation (1967). Company records dating back to 1925 identified about 4600 individuals (80% men) employed at plant sites in Chicago (up to 1936) and West Chicago (1932-1973) and at a mantle factory in Morris, Illinois (1947-1953). Since records before 1940 and for the Morris plant were incomplete, the study was limited to 3222 men and 714 women who worked at the West Chicago plant after 1939. Social Security numbers, job classifications, and work dates have been found for almost

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*Executive Summary from the interim report of the same title, NUREG/CR-1420, ANL-80-37 (January 1980).

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all of these former employees.

An industrial hygiene survey of the plant in 1952 showed that gamma-ray levels of 0.5 to 5 mR per hour were common in locations where thorium was processed or stored and that the levels of airborne thorium and thoron daughter products were of the order of present day maximum permissible concentrations. Estimates of radiation exposures during 1956-1973 are being compiled from inspection reports of the U.S. Atomic Energy Commission, records of personal dosimeter readings, and company records of radiation and radioactivity measurements. Filter samples that were collected while the plant was in operation are being analyzed for particle sizes and chemical composition of airborne materials.

Overall and cause-specific mortality was studied in a cohort of 3039 male thorium workers on the basis of deaths reported by the Social Security Administration and causes on death certificates. Comparisons were made with sex-, age-, time-, and cause-specific mortality rates for U.S. white males. In the total cohort, there were 511 observed deaths and 486.8 expected, and differences between observed and expected numbers were significant at the 95% confidence level only for deaths from diseases of the circulatory system (205 observed vs. 249.5 expected) and from motor vehicle accidents (38 observed vs. 23.2 expected). However, notably higher than expected numbers of deaths were observed for respiratory diseases (33 vs. 25.2) and cancers of the lung (31 vs. 21.6), pancreas (9 vs. 4.5), and rectum (6 vs. 3.2).

In general, the mortality differences were not strongly associated with job type or length of employment, but excess deaths from cancer of the pancreas were significantly greater among men employed at least one year than among shorter-term workers. Data on a small sample of the study population indicated a higher proportion of cigarette smokers than among U.S. males, and this could explain at least part of the excess mortality from lung cancer and respiratory diseases.

Medical examinations and in vivo measurements of body radioactivity are being done on a subpopulation of 592 male employess who have worked a year or more in job classifications involving probable exposure to thorium. By measurement of radioactive thoron-220 in exhaled breath, higher than background

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amounts of radium-224 were found in 131 of 194 men who have been examined (range 2 to 667 pCi). By gamma-ray spectrometry, measurable amounts of bismuth-212 in the thorax were found in 55 of the men (range 0.2 to 3 nCi). Comparisons with measured amounts of thorium-232 and thorium-228 in autopsy samples are needed for interpretation of these in vivo measurements of thorium daughter products.

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RADIOACTIVITY IN FORMER WORKERS AT A THORIUM REFINERY

J. Rundo, D. R. Brewster, M. A. Essling, and J. Y. Sha

As part of an epidemiological study of the possible late biological effects of thorium, measurements have been made of radioactivity attributable to thorium daughters in almost 200 men who had worked in a thorium refinery which closed in 1973. For external gamma-ray measurements statistically significant results ($> 2\sigma$) were obtained in 55 of these, with three showing more than 2 nCi ^{212}Bi in the thorax. For measurements of daughters of exhaled thoron, statistically significant results were obtained in almost every case, but for 63 subjects the values of < 2 pCi of freely emanating ^{224}Ra at the mouth could not be attributed unequivocally to thorium acquired occupationally; 131 men exceeded this lower limit and four had more than 200 pCi of ^{224}Ra. The mean ratio of emanating ^{224}Ra to retained ^{212}Bi was 101 pCi/nCi, with individual values ranging from 11 to 581. The problem of interpreting the data in terms of the actual amounts of thorium in the thorax is discussed briefly.

*Abstract of a paper presented at the Workshop on Measurements and Interpretation of Actinide Accumulation by Man, Snowbird, UT, October 15-17, 1979.

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EXCRETION OF ^{210}Pb AND ^{210}Po BY WORKERS IN AN AREA WITH HIGH LEVELS OF ATMOSPHERIC RADON, AND ESTIMATES OF EXPOSURE TO SHORT-LIVED RADON DAUGHTERS

R. B. Holtzman, P. W. Urnezis, and J. Rundo

The urinary excretion rates of ^{210}Pb were used to estimate the exposure to ^{222}Rn and its short-lived daughter-products for 12 persons who worked at an industrial site formerly used for the processing of uranium ores. The geometric mean excretion rates of ^{210}Pb and ^{210}Po of 0.86 and 1.93 pCi/day were significantly above normal environmental levels; the geometric standard deviations (σ_g) were 2.0 and 5.7, respectively. The mean value of the "available" ^{210}Pb (that in the soft tissue) was estimated from the excretion rates of ^{210}Po to be about 2.6 nCi. The geometric mean lung exposure derived from the ^{210}Pb excretion rates was estimated at 0.41 WL based on a conversion factor of 1.49 pCi day^{-1} WL^{-1}. This mean did not agree with the mean value of 0.14 WL that had been estimated from radon decay products in the atmosphere of the building.

Introduction

A survey of an industrial park in Canonsburg, PA, at which uranium ores had been processed and residues dumped, showed high levels of radon in the air in some buildings (up to 200 pCi/L), and the presence of high levels of uranium and ^{226}Ra in the dust and buildings.[1] Consequently, a group of people who worked at the site were measured at Argonne for possible internal contamination. Measurements of their body radioactivity in a low-background counting facility did not show levels in excess of those found in the general population.[2] Urine specimens were collected for bioassay for possible contaminants from the uranium decay series, uranium, ^{226}Ra, ^{210}Pb, and ^{210}Po. The results of the analyses for ^{210}Pb and ^{210}Po are discussed here.

Experimental Methods

Urine specimens were obtained from the subjects when they came to Argonne for the studies starting on a Monday morning. Each subject was asked to collect all urine over a known period of time (approximately 1 day) in clean plastic bottles supplied by us. The urine samples were then wet ashed in hydrogen peroxide and perchloric acid, and the samples were aliquoted for the various analyses.

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The ^{226}Ra was determined by the de-emanation method of Lucas.[3] The ^{210}Pb and ^{210}Po were determined by removal of the ^{210}Po from the solution by the spontaneous deposition of ^{210}Po, the decay product of ^{210}Pb, onto a silver disk which was then alpha counted.[3,4] This plating process was repeated after about 3 months, and the activities of both the ^{210}Pb and ^{210}Po were determined from the counting data and application of the Bateman equations for radioactive growth and decay.

Results

Data on the subjects are presented in Table 1, including their age at the time of measurement, the numbers of the buildings in which they worked, and the approximate length of time they had worked in the building. The radon concentrations and Working Levels (WL)* measured in 1977 in buildings in which the subjects worked are presented in Table 2.[1] It should be noted that the values reported for the WL were not independent measurements made simultaneously with those of the radon concentrations, but were derived from the latter by application of an average value of about 0.4 for the equilibrium factor. This is a source of uncertainty in determining the exposure to individuals in our study.

The results of the analyses of the urine from 12 subjects (10 males, 2 females) are presented in columns 2 and 3 of Table 3, and they are plotted by case number in Figure 1, which includes the results for ^{226}Ra for comparison. It is seen that there is little correlation between the values for the three nuclides. The exceptionally high value for ^{210}Pb for case 30-170 was confirmed on re-analysis. The geometric mean excretion rates of ^{210}Pb and ^{210}Po were about four and seven times normal, respectively, and the geometric standard deviation for the latter was much higher than that for the former, as expected from the very large range of values.

Five subjects (30-159, -163, -167, -170, and -171) gave values for the urinary excretion rate of ^{226}Ra that were more than five times normal

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* "Working Level" is defined as any combination of concentrations of radon daughters (^{218}Po, ^{214}Bi, and ^{214}Pb) in one liter of air that results in 1.3×10^5 MeV of potential alpha energy.

Table 1. Data on the subjects.

Subject,[a] CHR case number	Age at time of measurement, yr	Building number	Time worked in building, yr	Comment
30-159	43	18	5	
160	37	10 and 18	1 and 6	
161	46	10, 9	12	
162 F	45	16, 9	12	
163	32	10A	7	Exposed to dust.
164	65	10A	7	
165	39	7	7	
166	40	15	9	10 days since last exposure, spends much time in a concrete pit.
167 F	46	3	9	
170	32	10A	7	Works in various places.
171	68	10	12	One month since last exposure.
172	54	16	14	

[a]Males, except as noted by F.

(0.2 pCi d^{-1}). This suggested the possibility of contamination from inhaled dust, which might also have contained significant amounts of ^{210}Pb and ^{210}Po, in addition to that produced from radioactive decay from the ^{226}Ra content. Many of the subjects commented that their working environments were quite dusty.

Table 2. Average daytime concentrations of radon and its short-lived daughter products in buildings at the Canonsburg site, March 14-April 1, 1977.[a]

Building number	Concentration of ^{222}Rn, pCi L^{-1}	Working Levels
3	106	0.43
7	34	0.14
9	51	0.21
10	38	0.15
15	15	0.06
16	38	0.16
18	31 (2.6[b])	0.12 (0.01[b])
19	19	0.08

[a] From Ref. 1.

[b] In office.

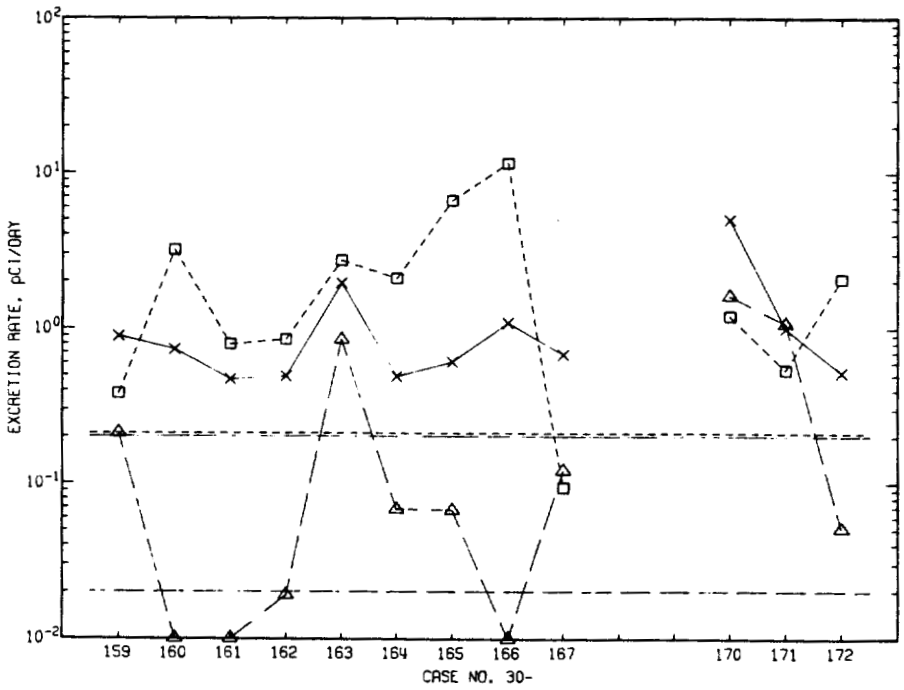

FIG. 1.--Urinary excretion rates of Canonsburg cases plotted against case number. x, ^{210}Pb; □, ^{210}Po; Δ, ^{226}Ra. The horizontal lines represent the normal urinary excretion rates for the respective nuclides.

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Table 3. Excretion rates and estimated exposure.

Subject,[a] CHR case number	^{210}Pb, pCi d^{-1}	^{210}Po, pCi d^{-1}	Estimated exposure,[b] WL	Daytime levels,[c] WL
30-159[d]	0.89 ± 0.17	0.38 ± 0.09	0.46	0.1
160	0.73 ± 0.08	3.17 ± 0.17	0.36	0.12
161	0.47 ± 0.11	0.79 ± 0.14	0.18	0.15
162 (F)	0.49 ± 0.17	0.85 ± 0.19	0.19	0.16
163[d]	1.95 ± 0.30	2.73 ± 0.36	1.17	0.15
164	0.49 ± 0.13	2.09 ± 0.30	0.19	0.15
165	0.61 ± 0.22	6.67 ± 0.29	0.28	0.14
166	1.09 ± 0.22	11.59 ± 0.62	0.60	0.06
167 (F)[d]	0.68 ± 0.10	0.094 ± 0.098	0.32	0.43
170[d]	5.0 ± 0.5	1.20 ± 0.46	3.22	0.15
171[d]	1.00 ± 0.15	0.54 ± 0.15	0.54	0.15
172	0.52 ± 0.19	2.08 ± 0.24	0.21	0.16
Geometric Mean	0.86	1.37	0.41	0.14
σ_g	2.0	3.7	2.4	1.6
Normal excretion rates (range):	0.2(0.1-0.4)	0.2(0.1-0.5)		

[a] Males, except as denoted by F.

[b] Calculated with the assumptions that an exposure of 1 WL (equilibrium factor 0.4) will produce sufficient ^{210}Pb to cause an excretion rate of 1.49 pCi day^{-1} 3 days after the end of chronic exposure for 10 years, and that the normal excretion rate of ^{210}Pb is 0.2 pCi d^{-1}.

[c] Average daytime levels in building taken from Table 2.

[d] Subjects showing high urinary excretion rates of ^{226}Ra.

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Discussion

It has been shown that the mean excretion rates of ^{210}Pb and ^{210}Po from natural sources are similar at about 0.2 pCi d^{-1} and exhibit similar ranges.[5] Under such conditions, the principal site of deposition of the ^{210}Pb is bone and the rate-controlling factor for excretion of both nuclides is the loss from bone. By contrast, exposure of the subjects in the present study was relatively short-term and the deposition in bone may supply a much smaller fraction of the total excreted. While the ^{210}Pb may be moderately tightly bound in its soft tissue pools (with a biological half-life of the order of 1 year)[6] the plutonium may be much less so, and thus more readily available for excretion. If we assume that the higher excretion rates of the polonium are not due to much greater intakes of this nuclide, we may estimate the amount of ^{210}Pb in the soft tissue pools. Thus, 200 pCi ^{210}Pb produce 1 pCi of ^{210}Po/day by radioactive decay. With a ratio of endogenous fecal-to-urinary excretion rates of ^{210}Po of 10,[7,8] a net urinary rate of 1.2 pCi d^{-1} (1.4 pCi d^{-1} observed, less 0.2 pCi d^{-1} environmental, Table 3) represents about 2.6 nCi ^{210}Pb that is "available" to produce the readily eliminated ^{210}Po. It does not necessarily represent the total ^{210}Pb.

We may use the excretion rates of ^{210}Pb (in excess of the normal level of 0.2 pCi d^{-1}) to calculate the exposure to short-lived radon daughters using the model developed in the previous report.[9] We use the conversion factor of 1.94 pCi d^{-1} WL^{-1} appropriate to the average equilibrium factor of 0.4 reported[1] for the buildings in the Canon Industrial Park, and modify it by a factor of 0.77 to allow for the decrease in the excretion rate in the 2 or 3 days between the end of the exposure and the collection of urine (as discussed in Appendix B of Ref. 8). This results in a value of 1.49 pCi d^{-1} WL^{-1}; the normal level of 0.2 pCi d^{-1} was subtracted from each value in column 2 of Table 3 and the result was divided by 1.49 to give the estimated WL in column 4 of Table 3. In column 5 the appropriate WL reported in Ref. 1 and shown in Table 2 is given for each subject. The geometric mean of the values estimated from the model is three times that for the values in column 5, but it should be noted that the ratio of the estimate from the model to the value for the appropriate building is less than the geometric standard deviation for each of six (i.e., half) of the subjects and that only one

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of these (30-167) showed a high excretion rate of ^{226}Ra. Furthermore, the geometric mean for the seven subjects who did not show high levels of ^{226}Ra in the urine, was 0.27 WL (with a geometric standard deviation of 1.5), little different from the arithmetic mean of 0.30 (standard deviation ± 0.16). If we accept that the larger differences in the subjects who showed high excretion rates of ^{226}Ra were indeed due to their having inhaled contaminated dust, we are left with only two subjects (30-160 and 30-166) where the discrepancy between estimated and "observed" WL is unreasonably high. The difference (a factor of 10) for subject 30-166 (a mechanic) might well be explained by his having spent much time in a concrete pit where the concentration of radon would be expected to be higher than in the building proper. No such explanation is apparent for subject 30-160.

The extent of the agreement between the values in columns 4 and 5 of Table 3 leaves little room for complacency, because we have not taken into account the fact that the subjects were only exposed to high levels of radon and daughters during the working day of about 8 hr, for 5 days a week. Thus, the "occupancy factor" was only about 0.25, and the estimates in column 4 should probably be multiplied by four, eliminating any agreement between the pairs of values in columns 4 and 5.

Two possible reasons for this suggest themselves. First, the model may be in error, and possible ranges of the conversion factor deduced from the model are discussed in Ref. 9. Second, the value of 0.2 pCi d^{-1} for the normal excretion rate of ^{210}Pb may not apply to the individuals of this study. Examination of the values in column 2 of Table 3 shows that five of them do not differ significantly from the upper limit (0.4 pCi d^{-1}) of the normal range. If that value is used for the seven subjects who did not show high ^{226}Ra excretion rates, the geometric mean of the estimated exposures is 0.12 WL (σ_g 2.3) but application of the "occupancy factor" again results in no agreement with the mean "observed" value for the WL.

It is clear that the model has not yet been sufficiently refined to give accurate results for the exposure, but it is also clear that subjects who have been exposed to high levels of radon and its short-lived daughter products do

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excrete significant amounts of ^{210}Pb. The problem of interpretation of those amounts must await further developments of the model.

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AN IMPROVED ANALYTICAL PROCEDURE FOR THE DETERMINATION OF ^{210}Pb AND ^{210}Po USING ALPHA-SPECTROMETRIC ISOTOPE DILUTION

P. W. Urnezis and R. B. Holtzman

An isotope dilution method has been incorporated into the ^{210}Pb-^{210}Po analysis. A known amount of ^{209}Po is added to the sample before analysis. Then both ^{209}Po and ^{210}Po are deposited on a silver planchet which is assayed in an alpha spectrometer to determine the activities of each isotope. The recoveries generally range from 70% to 90%.

Introduction

Several methods are available for the determination of ^{210}Pb.[1] The 47 keV gamma ray can be measured directly. This allows for measurements in vivo, but this method is practical only when sufficient amounts of ^{210}Pb are present, since the gamma ray occurs in only about 5% of the disintegrations. For low-level activity, other methods are necessary.

The activity of ^{210}Pb can be determined by measurements of its daughters, ^{210}Bi or ^{210}Po. Because of the short half-life of ^{210}Bi (5 days) radioactive equilibrium is rapidly established between it and ^{210}Pb, so that only a single analysis of ^{210}Bi is necessary to determine the amount of ^{210}Pb. If ^{210}Po is used to determine the activity of ^{210}Pb, two analyses are required, but the resulting data may then be used to determine the amount initially present of both ^{210}Pb and ^{210}Po. Since ^{210}Po is an alpha emitter, it can be measured with alpha-particle counters which have significantly lower backgrounds than do the beta-particle counters used to measure ^{210}Bi. If the activities of ^{210}Pb are low and time is not critical, the indirect method of measuring ^{210}Po is the method of choice. We have improved our routine method[2,3] by modifying it to use alpha-spectrometric isotope dilution.

Experimental Methods

The sample is ashed at a low temperature to destroy any organic matter present while at the same time preventing volatilization of polonium and lead. The methods used depend on the type of sample, as previously described.[2,3]

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All samples, whether bone, metabolic (e.g., food and excreta), or soft tissue samples, may be ashed by repeated additions of nitric acid while being heated. Alternatively, metabolic and soft tissue samples may be ashed with hydrogen peroxide acidified with nitric acid while being heated. Soft tissue may also be oxidized in a low temperature asher that uses an oxygen plasma. After ashing is complete, or nearly so, the sample is fumed with perchloric acid to complete the oxidation and to remove residual nitric acid or hydrogen peroxide, since both of these attack silver and would disrupt the analysis.

The sample holder (see Fig. 1) is assembled with a silver planchet in the Teflon insert and the sample is poured in, after the pH has been adjusted to 0.3. The ^{209}Po spike is added, the sample holder is placed in the heating mantle, and the stirrer is lowered into place. A watch glass with a slot for the stirrer covers the sample to reduce the evaporation rate. The sample is heated to 95°C and stirred for 6 hr. The stirrers are then raised and the sample holder is removed from the heating block. The sample is saved for a repeat analysis several months later. The silver planchet is rinsed with distilled water and 1.0 M HCl, and dried, and the activity on it is assayed in an alpha spectrometer to determine the contributions of ^{209}Po and ^{210}Po. The procedure is described in detail in Appendix A and the calculation in Appendix B.

The heating mantle is a cylindrical aluminum block into which 4 stainless steel tubes are inserted. These tubes are 6.4 cm i.d. by 15 cm deep and contain water. The stirrer assembly is supported above the mantle by a vertical rod anchored to the center of the heating block. The solutions are stirred by four Nalge polyethylene propeller-type stirring rods linked to a variable speed motor by a slip-proof belt.

The activity of the ^{209}Po standard solution was determined by stippling a small known amount onto a silver planchet which was gently heated from below with a hot plate and from above with a heat lamp. The planchet was then counted in an internal gas proportional counter of known counting efficiency and then in an alpha spectrometer. The proportional counter was used to determine the activity and the alpha spectrometer to check the purity. Another aliquot of the ^{209}Po solution was diluted to 200 mL with distilled water, and the pH was adjusted

MEASUREMENTS OF RADIOACTIVITY IN FORMER MILITARY PERSONNEL EXPOSED TO WEAPON DEBRIS

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Sixteen former military personnel who were present at the "Smoky" atmospheric nuclear weapons test have been investigated for internal deposits of radioactivity. Whole-body and thorax γ-ray measurements, thorax and skeletal actinide measurements, and urinalyses for ^{239}Pu and ^{90}Sr were performed. No evidence of radioactivity in excess of that found in the general population was observed.

Introduction

During the 1950's, an estimated 250,000 military personnel were present during nuclear weapons tests at the Nevada Test Site, either as permanent party support troops, or as participants in maneuvers designed to determine their ability to accomplish assigned missions on a nuclear battlefield. On 31 August 1957, a 44-kiloton device code-named "Smoky" was exploded. Eight cases of leukemia have been reported in 1900 of the 3200 troops present at that test. Film badge readings were available for seven of the eight cases, and ranged from 0 to 2,997 mrem with a mean of 1,178 mrem. The mean 1957 cumulative dose for the entire 3200-man cohort was 493.4 mrem.[1]

Because the film badges recorded only the external gamma dose, the Center for Human Radiobiology was asked to determine the levels of internal radioactivity in a group of men present at the "Smoky" test. Nineteen men were selected by the Center for Disease Control on the basis of high film-badge readings and/or their opportunity for inhalation or ingestion of weapon debris. None of them exhibited any clinical signs of malignancy or other radiation-induced pathology. The nineteen were assigned CHR case numbers 30-140 to 30-158. Sixteen of them visited ANL in 1979 for body radioactivity measurements; three chose not to participate.

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The men were briefly interviewed during their visits, and several reported being present at up to three tests. Their locations at the time of detonation ranged from a trench at 2.5 km from ground zero to a hillside 16 km away. After the tests, some were trucked or marched to within an estimated 200 m of ground zero where they were timed on their performance of routine tasks such as weapon cleaning. One man reported that protective masks were worn, while another reported that they were not. One man was a wireman whose job was to re-install telephone lines that were destroyed by each test, and he reported that packed lunches were eaten frequently while in the vicinity of ground zero. All the men reported that the test site was an extremely dusty environment. Thus it appears that the potential for internal deposition of radioactivity did exist.

Measurement Techniques

The whole-body contents of γ-ray emitters were measured with large NaI(Tl) detectors in both the reclining chair and flat bed geometries. This latter geometry maximized the efficiency of detection of γ-ray emitters in the thorax. The detectors were calibrated for ^{40}K and ^{137}Cs by counting water-filled phantoms made of plastic bottles arranged to simulate the shape of a human body and containing known amounts of these radionuclides. Different arrangements of bottles were used to calibrate for different body sizes. The γ-ray spectra from the subjects were analyzed by a computer method of least squares.

Measurements of actinide (^{239}Pu and ^{241}Am) content were made with a 180-mm diameter xenon-filled proportional counter. A mean subject background was subtracted from the counting rate observed over the chest in the energy range 16 to 24 keV; the remainder was assumed to be due to pure ^{239}Pu, since no evidence of the 60-keV line from ^{241}Am was observed. It must be noted, however, that this method overestimates the plutonium alpha activity, since ^{238}Pu, ^{240}Pu, and ^{242}Pu all emit more x rays per alpha than does ^{239}Pu. The counting rate was converted to a lung content by applying a calibration factor obtained from a realistic thorax phantom containing lungs loaded with a known amount of ^{239}Pu and having a variable chest wall thickness.[2] The appropriate

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phantom calibration factor was determined for each subject by measuring the subject's average chest wall thickness with an ultrasonic probe. The skeletal contents of ^{239}Pu were determined from the measurements over the skulls. (The skull has been shown to be a representative bone for the determination of skeletal burdens of bone-surface seeking radionuclides in humans.)[3,4] A mean background was determined by counting over the skulls of several controls. The net counting rate from each subject was converted to skeletal ^{239}Pu by applying a calibration factor of 23 ± 1 nCi ^{239}Pu per cpm.[5] This factor was derived from measurements made over the skull of a case from a different group of subjects[6] whose skeletal burdens of plutonium were determined radiochemically.[3] Note that because the thickness of soft tissue over the skull does not vary appreciably, the calibration factor is constant despite the range of body sizes.[4]

While they were at ANL, the subjects in this study supplied 24-hr urine specimens. Aliquots were analyzed for ^{239}Pu by isotopic dilution alpha spectrometry, and for ^{90}Sr by chemical separation and beta-counting. Estimates of an upper limit for the body content of ^{239}Pu were made (a) with the aid of Langham's power function equation,[7] which yields a value for the intial systemic content, and (b) with the application of the retention function proposed by a Task Group of the ICRP.[8] The second method required an assumption about the amount excreted in feces. We multiplied the urinary excretion rate by 1.47 to derive a value for the total excretion rate.[9] It should be mentioned that it is known that use of the Langham equation results in an over-estimate of the initial systemic content of plutonium when it is applied at late times afer intake.[9]

The possible body contents of ^{90}Sr were calculated in the following manner. The urinary excretion rates of ^{90}Sr were multiplied by 1.3 to allow for fecal excretion, and the total excretion rates were converted to current body contents with the aid of the retention equation suggested by a Task Group of the ICRP.[10] The ^{90}Sr levels of control subjects were determined in like manner.

Results

The results are presented in tabular form. Table 1 gives biometric data on the subjects, and Table 2 presents the results of the anlyses of the gamma-ray

Table 1. Biometric data on the 16 subjects.

CHR case No.	Date of birth	Height, m	Weight, kg	Chest wall thickness, mm
30-140	Feb. 22, 1931	1.78	108	29
-141	Mar. 20, 1935	1.78	68.6	33
-143	Jan. 31, 1936	1.66	64.8	13
-145	Jul. 14, 1937	1.64	62.2	17
-146	Sep. 24, 1938	1.75	79.2	20
-147	Dec. 1, 1930	1.71	58.8	15
-148	Apr. 22, 1926	1.70	83.5	28
-149	Aug. 24, 1936	1.75	75.4	36
-150	Feb. 14, 1938	1.83	93.5	36
-151	Nov. 21, 1930	1.85	76.2	20
-153	May 30, 1939	1.85	79.9	28
-154	Mar. 2, 1923	1.75	83.9	33
-155	Feb. 11, 1933	1.73	66.0	13
-156	Apr. 11, 1927	1.85	81.3	29
-157	Jul. 29, 1919	1.83	76.7	35
-158	Aug. 23, 1936	1.80	101	30

spectra, which were made with the assumption that the whole of the response was attributable to naturally occurring ^{40}K and fallout ^{137}Cs. Below the individual entries are shown the mean potassium content (as % of body mass) and the mean cesium to potassium ratio (pCi ^{137}Cs/g K), with their standard deviations and variance ratios (i.e., ratio of observed to predicted variances). The standard deviations are given rather than the standard errors of the means because the former are more indicative of the spread of the individual values. The fact that the variance ratios are greater than unity is indicative of biological variation in the potassium and ^{137}Cs contents, and is to be expected. This biological

Table 2. Potassium and ^{137}Cs contents.

Case No.	Potassium g	Potassium % body wt	^{137}Cs nCi	^{137}Cs pCi/g K
30-140	185 ± 4	0.171 ± 0.004	2.1 ± 0.3	11.1 ± 1.5
-141	158 ± 4	0.231 ± 0.006	1.8 ± 0.2	11.6 ± 1.6
-143	133 ± 4	0.206 ± 0.006	0.8 ± 0.2	6.2 ± 1.6
-145	123 ± 4	0.197 ± 0.006	0.8 ± 0.2	6.4 ± 1.8
-146	151 ± 3	0.191 ± 0.005	0.7 ± 0.2	4.5 ± 1.3
-147	112 ± 4	0.191 ± 0.006	not significant	
-148	124 ± 3	0.149 ± 0.004	1.4 ± 0.2	10.9 ± 1.9
-149	185 ± 4	0.245 ± 0.006	1.9 ± 0.3	10.2 ± 1.4
-150	175 ± 3	0.188 ± 0.004	1.7 ± 0.2	9.6 ± 1.3
-151	144 ± 3	0.189 ± 0.005	0.9 ± 0.2	6.1 ± 1.4
-153	194 ± 4	0.242 ± 0.005	1.9 ± 0.3	9.9 ± 1.3
-154	144 ± 4	0.172 ± 0.005	0.9 ± 0.2	6.6 ± 1.7
-155	170 ± 4	0.258 ± 0.007	not significant	
-156	140 ± 4	0.172 ± 0.005	1.1 ± 0.2	8.1 ± 1.6
-157	174 ± 4	0.227 ± 0.005	0.9 ± 0.2	5.3 ± 1.2
-158	176 ± 4	0.174 ± 0.004	not significant	
Mean ± S.D.		0.20 ± 0.03	1.3 ± 0.5	8.2 ± 2.5
Variance ratio		36	5.5	2.6
12 Control subjects				
Mean ± S.D.		0.19 ± 0.03	1.4 ± 0.4	8.9 ± 2.4
Variance ratio		38	2.7	2.0

variation was confirmed by the corresponding values given in Table 2 for 12 other men who were investigated during the same period and whose spectra were analyzed in the same way. The results for the mean potassium concentrations and the ^{137}Cs/potassium ratio of the 12 controls are similar to those for the 16 subjects, and the variance ratios were again high. It should be noted that the mean potassium content is exactly that given for Reference Man,[11] indicating that the calibration is correct. The ^{137}Cs contents and the $^{137}Cs/K$ ratios agree quite well with the results of others for the general population. The mean $^{137}Cs/K$ ratio for a group of 16 controls at the Atomic Energy Research Establishment, Harwell, UK, was 8.5 pCi/g in 1976,[12] and for a group of 40 workers at Los Alamos Scientific Laboratory, also in 1976, the ratio was 10.3 pCi/g, with a mean ^{137}Cs content of 1.3 nCi.[13] Thus it is evident that these subjects do not contain an excess of this fission product. All other gamma-ray emitters in the 16 subjects were below the limit of detection of $\sim$ 0.2 to 0.5 nCi (depending on the gamma-ray energy), with the exception of short-lived radon daughters. There was some evidence for the presence of ^{214}Pb and ^{214}Bi in most members of the group, as well as in the control group. However, statistically significant amounts were present in only three subjects. The results of the measurements of ^{239}Pu made with the proportional counter are set out in Table 3. Not one result is greater than zero at even the 90% (1.64 σ) confidence level, and the mean value for each set of measurements is close to zero. The fact that the variance ratio is close to unity indicates that biological variation is not playing a large role in these measurements, and that the results vary randomly about zero.

The plutonium (^{239}Pu) content of each urine sample was below the limit of detection (4.5 fCi) of our standard method. From Langham's equation relating daily urinary excretion to systemic intake, we deduce that the latter was less than 1.7 nCi in August 1957 (7900 days before the urine collections). Since use of the equation at times much longer than 5 years is known to over-estimate the systemic intake,[9] the value of 1.7 nCi must be regarded as an extreme upper limit. A limiting value for the current body content of < 200 pCi results from use of the retention equation suggested in ICRP Report 19.[8] These low values

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Table 3. External counting of low energy photon emitters.

	Measurements over chest			Measurements over skull	
Case No.	Net cpm[a]	Calibration factor, nCi/cpm	^{239}Pu in chest, nCi[b]	Net cpm[c]	^{239}Pu in skeleton, nCi[b]
30-140	0.45 ± 0.54	119	54 ± 64	0.62 ± 0.46	14 ± 10
-141	0.25 ± 0.53	154	38 ± 82	0.02 ± 0.41	0.5 ± 10
-143	-0.15 ± 0.51	22	-3 ± 11	-0.52 ± 0.37	-12 ± 9
-145	-1.08 ± 0.44	34	-37 ± 15	0.15 ± 0.42	3.5 ± 10
-146	0.12 ± 0.52	50	6 ± 26	-0.78 ± 0.34	-18 ± 8
-147	-0.48 ± 0.48	29	-14 ± 14	0.42 ± 0.45	10 ± 11
-148	-0.81 ± 0.46	110	-89 ± 51	0.55 ± 0.46	13 ± 11
-149	0.12 ± 0.52	192	23 ± 26	0.62 ± 0.46	14 ± 10
-150	-0.01 ± 0.51	192	-2 ± 98	-1.05 ± 0.37	-24 ± 9
-151	0.12 ± 0.52	50	6 ± 26	0.15 ± 0.42	3.5 ± 10
-153	0.25 ± 0.53	110	27 ± 58	-0.12 ± 0.40	-3 ± 9
-154	-0.41 ± 0.49	154	-63 ± 75	0.28 ± 0.43	6 ± 10
-155	1.05 ± 0.58	22	23 ± 13	0.55 ± 0.46	13 ± 11
-156	-0.01 ± 0.51	132	-1 ± 67	0.14 ± 0.42	3 ± 10
-157	-0.28 ± 0.50	182	-51 ± 91	-0.25 ± 0.39	-6 ± 9
-158	0.72 ± 0.56	127	91 ± 71	-0.52 ± 0.41	-12 ± 9
Mean ± S.D.	-0.01 ± 0.54		0.5 ± 45	0.02 ± 0.52	0.3 ± 12
Variance ratio	1.1		0.6	1.5	1.5

[a]After subtraction of control background of 3.68 ± 0.13 cpm (±1 S.E.)

[b]Calculated with the assumption that the whole of the net response was due to pure ^{239}Pu.

[c]After subtraction of control background of 2.05 ± 0.18 cpm,

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for plutonium content based on excretion rates confirm the negative results of the external measurements of plutonium content made with the proportional counter.

The results for the daily urinary excretion of ^{90}Sr are given in Table 4. Also included are the values for seven control samples. Three of the control values were obtained from 24-hr urine collections by individuals, and the remaining four were obtained from pooled urine samples obtained from a number of individuals during working hours. The values from the pooled samples (pCi/L) were converted to daily excretion values by multiplying them by the average daily urinary output of 1.4 L given in ICRP Report 23.[11] None of the control subjects was ever exposed to ^{90}Sr other than that from global fall-out.

The mean excretion rate and its standard deviation from the 16 test subjects are essentially identical to those from the controls. We thus infer that both groups are drawn from the same population, i.e., the ^{90}Sr excreted by the test subjects arises from global fallout rather than from their presence at weapon tests.

Summary and Conclusions

We tested 16 subjects exposed to weapon debris for internal deposition of radioactivity by a combination of external counting and urinalysis. In none of the 16 subjects were we able to detect radioactivity in excess of normal levels carried by all members of the population, and we conclude that long-lived isotopes have not contributed any internal component to the radiation doses received by these men due to their participation in the "Smoky" test.

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Table 4. Urinary excretion rates and calculated current body contents of ^{90}Sr.

Case No.	Urinary excretion, pCi/d	Systemic burden, nCi[a]
30-140	0.47 ± 0.06	1.8 ± 0.2
-141	0.92 ± 0.08	3.4 ± 0.3
-143	0.40 ± 0.05	1.5 ± 0.2
-145	0.30 ± 0.05	1.1 ± 0.2
-146	1.39 ± 0.13	5.2 ± 0.5
-147	0.19 ± 0.05	0.7 ± 0.2
-148	0.29 ± 0.07	1.1 ± 0.3
-149	0.69 ± 0.06	2.6 ± 0.2
-150	0.74 ± 0.11	2.8 ± 0.4
-151	0.66 ± 0.08	2.5 ± 0.3
-153	0.44 ± 0.05	1.6 ± 0.2
-154	0.72 ± 0.06	2.7 ± 0.2
-155	0.42 ± 0.07	1.6 ± 0.3
-156	0.50 ± 0.10	1.9 ± 0.4
-157	1.23 ± 0.09	4.6 ± 0.3
-158	0.70 ± 0.09	2.6 ± 0.3
Mean ± S.D.	0.63 ± 0.33	2.4 ± 1.2
Variance ratio	17.6	17.8
Controls		
1	1.13 ± 0.09	4.2 ± 0.3
2	0.95 ± 0.10	3.6 ± 0.4
3	0.58 ± 0.09	2.2 ± 0.3
4[b]	0.50 ± 0.07	1.9 ± 0.3
5[b]	0.21 ± 0.07	0.8 ± 0.3
6[b]	0.57 ± 0.06	2.1 ± 0.2
7[b]	0.67 ± 0.09	2.5 ± 0.3
Mean ± S.D.	0.66 ± 0.30	2.5 ± 1.1
Variance ratio	13.4	13.6

[a] Calculated from the retention equation suggested in ICRP Report 20.

[b] Pooled sample, pCi/L x 1.4 = pCi/d.

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