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FOREWORD

This is the fourteenth Annual Report of the Center for Human Radiobiology. The past year was marked by the retirement of Dr. Robert E. Rowland, who was principally responsible for establishment of the Center at Argonne to investigate the biological effects of radium and other internal emitters in man. Dr. Rowland directed the Center and the Radiological and Environmental Research Division (RER) until he was appointed Interim Associate Laboratory Director for Biomedical and Environmental Research, the Argonne post from which he retired in February 1983. Because of related management changes, the organizational listings in this Annual Report were in effect for only a limited period during the year. Dr. John Rundo was Section Head until he was appointed Interim Associate Laboratory Director in February. The Environmental Research Division was organized in May by merger of the RER Division with the Environmental Impact Studies Division and portions of other Argonne groups.

New cases of bone cancer or carcinoma of head sinuses are still occurring at the rate of about one per year in patients who acquired radium burdens 50 or 60 years ago. The first paper describes the three most recent cases; in one of these, the radium burden was the lowest yet observed for bone cancer in this population, and in another case it was the lowest for mastoid carcinoma. A new approach to analysis of time-dose relationships for bone cancers in radium cases is indicated in the second report. This is followed by two reports on the risk of other types of cancer among female radium dial workers and a paper on skeletal effects other than cancer that have been found in radium patients. The sixth paper refers to electron-microscope measurements of cell size and shape that the late E. L. Lloyd incorporated into studies of the effects of alpha particles on cells in situ and in culture.

Papers 7 through 16 deal with the measurement and/or dosimetry of alpha-emitting radionuclides in man, in animals, or in the environment. Ongoing work to determine the dose from 5.75-yr ^{228}Ra in early radium dial workers is described in paper 8. Paper 10 is a summary of a widely disseminated report on the Center's measurements of radon in houses. Paper 7 describes a recent case of inhaled radium that was ascribed to a maintenance procedure at a uranium mill, while paper 16 is a report on lung doses that were estimated on the basis of reported levels of airborne uranium dust and radon and gamma rays

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at uranium plants in the mid-1940s. Paper 12 presents experimental evidence that the fractional transfer of uranium from the gut to blood in humans is much less than the value of 0.2 that has been proposed for use in setting safety standards for uranium in drinking water. The measurements of ^{226}Ra and ^{228}Ra in well water (paper 9) were done in collaboration with the Illinois State Geological Survey, and the work on the gastrointestinal absorption of actinide elements in animals (papers 13 and 15) was done in collaboration with Argonne's Biological and Medical Research Division. Radiochemical methods that were developed in the Center for measurement of the actinide elements are described in paper 14. Papers 11, 17, and 18 are reports of work done at Argonne by visiting scientists.

The report concludes with the usual appendices; Appendix A contains data on the exposure of 2312 persons whose radium content has been determined, while the tables in Appendix B list the classical radium-related malignancies (osteosarcomas and carcinomas of the paranasal sinuses and mastoid). The section on unmeasured cases in Appendix B contains a lengthy discussion on obtaining useful estimates of radium burdens from hard-to-interpret measurements reported in the early literature on radium cases.

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AN UNUSUAL CASE OF RADIUM EXPOSURE

R. E. Toohey, J. Y. Sha, P. W. Urnezis and E. Y. Hwang

We have determined the body content, distribution, retention, and excretion rate of ^{226}Ra for a uranium mill worker who inhaled ^{226}Ra in an unknown form. Radium was retained in the lung with a biological half-life of 120 days, and the amount initially inhaled was estimated to be 180 ± 30 Bq. These values, combined with an observed radon retention factor of 71%, implied a 50-year dose commitment of 0.16 ± 0.04 Sv to the lung. Although it is believed that this case represents an isolated incident, it is possible that some uranium mill workers may form a contemporary population that is occupationally exposed to radium.

Introduction

At the request of the U.S. Nuclear Regulatory Commission, we determined the body content of ^{226}Ra of a uranium mill worker (CHR Case No. 30-206) on four occasions over 13 months. The subject was a 47-year-old white male in generally good health who was a rubber worker at the mill. Among other tasks, his job included the removal of old rubber liners from ion-exchange tanks by use of a hand-held grinder. It is believed that he inhaled ^{226}Ra-contaminated particles generated during this operation. The midpoint of that particular work period was taken as the time of exposure.

Measurements

The body content of ^{214}Bi was determined with a 152 × 203 mm NaI(Tl) detector in the reclining chair geometry, and the emanating radium content was determined by collecting and determining exhaled ^{222}Rn by means of our standard procedures.[1] The results are listed in Table 1. The ^{214}Bi values were calculated on the assumption that the activity was in the chest only, as determined from the seven-position scan data given below.

The emanating radium values were lower in the afternoons than in the mornings of days 182 and 270. This may have been due to a postprandial effect, since the subject ate breakfast, but no lunch, on those days.[2] Consequently, we decided that the best estimates of body content could be obtained by taking the weighted mean of the morning and afternoon ^{214}Bi measurements for days 182 and 270, and adding the afternoon values for

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TABLE 1. ^{214}Bi in the lungs, emanating ^{226}Ra, and total ^{226}Ra values for case 30-206. One standard deviation is listed with each entry.

Days post-exposure	^{214}Bi, Bq	Emanating ^{226}Ra, Bq	Total ^{226}Ra, Bq
109	59 ± 16	41 ± 6	100 ± 17
182 (a.m.)	52 ± 14	51 ± 13	
182 (p.m.)	36 ± 10	31 ± 7	73 ± 11[a]
270 (a.m.)	29 ± 10	34 ± 4	
270 (p.m.)	28 ± 10	16 ± 8	45 ± 11[a]
508	30 ± 11	23 ± 3	53 ± 11

[a] Sum of the afternoon emanating radium and the weighted mean of the ^{214}Bi measurements.

emanating radium. Unfortunately, time did not permit replicate measurements on days 109 and 508. The body burdens of ^{226}Ra are listed in the final column of Table 1, and it is apparent that they did not follow a simple function of time post-exposure. It should be pointed out, however, that the ^{214}Bi values on days 270 and 508 were not statistically significant (more than 3σ above zero), and so the resulting body burden numbers may be unreliable. The mean radon retention was 71% (range 69-75%). This value was consistent with values ranging from 48-90% observed by Marinelli et al. in six subjects who had inhaled radium sulfate dust.[3]

The distribution of ^{214}Bi in the body was determined by a seven-position scan performed each day. As shown in Figure 1, the data from one of the two crystals used indicated that no detectable amounts of activity were translocated from the lung to other parts of the body as the lung content decreased. It was possible to determine the half-life for retention in the lung from these data. The net counting rates in the upper and lower crystals at position 2 (thorax) were added, and the sums are shown in Figure 2. A single exponential function of time was fitted to these data and also is shown in Figure 2. The half-life was 120 ± 20 days, and the intercept was

85 ± 16 cpm. This latter value was converted to an activity by determining the mean ratio of the summed counting rates in position 2 to the ^{214}Bi content of the chest as measured in the reclining chair. The ratio was 0.67 cpm/Bq, so the estimated initial lung content was 127 ± 23 Bq ^{214}Bi. The initial radium burden was then obtained by dividing the ^{214}Bi content by 0.71, the observed mean radon retention. The result was 180 ± 30 Bq ^{226}Ra. A fit to the first three body burden values gave results consistent with these values, equal to 170 ± 60 Bq initial burden and 140 ± 35 day half-life. Since the results from the scan geometry had greater precision, they were used in calculations of excretion rate and dose commitment presented below.

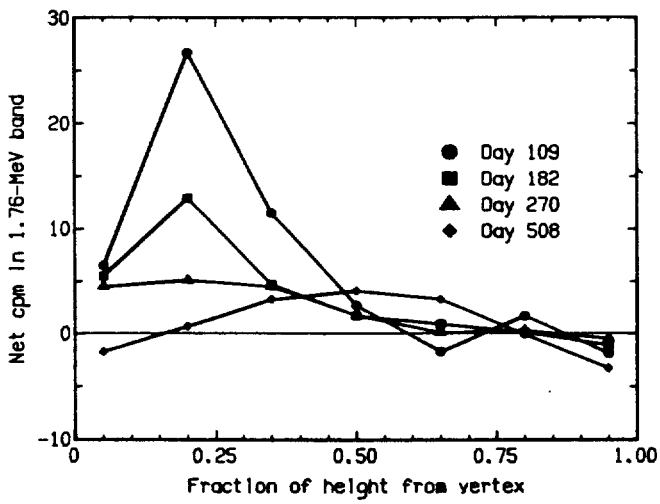

FIG. 1.--Results of seven-position scan measurements of ^{214}Bi in Case 30-206 with an uncollimated 292 × 102 mm NaI(Tl) detector below the supine subject. The fourth point of each scan is at the midpoint of the subject's supine body, and the remaining points are separated from one another by 15% of his height.

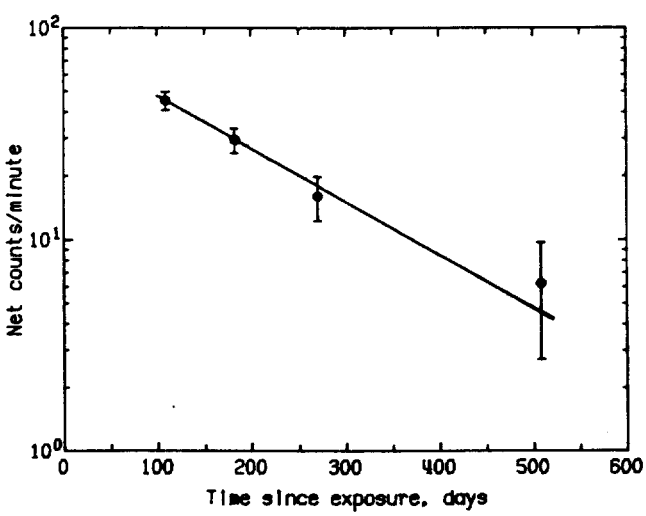

FIG. 2.--The summed net counting rates (y) of the upper and lower crystals at position 2 of the seven-position scan, as a function of time post-exposure (t). Error bars represent one standard deviation, and the smooth curve is a plot of the function $y = 85e^{-0.639t/120}$.

On each day that the subject visited CHR, 24-hour urine and feces samples were collected. The samples were later analyzed for uranium and thorium by alpha spectrometric isotope dilution, and for radium by de-emanation of radon. No uranium or thorium isotopes in excess of natural levels were observed, indicating that the subject was exposed to ^{226}Ra rather than to any of its precursors. The radium excretion rates are listed in Table 2. The mean excretion rate was (0.58 ± 0.42)% of the lung burden per day. Despite the wide range of individual values, this mean was exactly that predicted by the observed half-life of the lung content, and confirmed that the radium leaving the lungs was being excreted rather than deposited elsewhere in the body. It seemed likely that the radium was firmly attached to dust particles that were cleared from the lung, swallowed, and excreted. As a matter of interest, the excretion rates in this case were quite comparable to those observed in the only one of Marinelli's cases for whom excreta were analyzed. In that subject, the excretion rate decreased from 6% of the body burden at 2 days post-exposure, to 0.6% at 80 days, and 0.5% at 250 days.[3]

TABLE 2. Daily excretion rates of ^{226}Ra for case 30-206. For the excreta, one standard deviation is approximately 2% of the listed value.

Days post-exposure	^{226}Ra excreted, Bq day^{-1}			% of lung contents
	Urine	Feces	Total	
109	0.009	0.78	0.79	0.79 ± 0.13
182	0.008	0.17	0.18	0.25 ± 0.04
270	0.007	0.47	0.48	1.07 ± 0.26
508	0.009	0.10	0.11	0.21 ± 0.04

Discussion

The 50-year dose commitment to the lungs of this subject can be calculated according to the method of ICRP 30.[4] The data tabulated in ICRP 30 for inhalation of ^{226}Ra cannot be used directly because they assume retention in the lung has a mean halftime of 50 days (class W compound) and that 30% of the radon produced is retained in the body.

The 50-year dose commitment to a target organ, T, is given by:

$$H_{50,T} = 1.6 \times 10^{-10}\ U_T\ \Sigma SEE \quad , \tag{1}$$

where: U_t = the number of nuclear transformations in 50 years,

and $\Sigma SEE = \Sigma_i\ Y_i E_i AF\ (T \rightarrow T) Q_i / M_T$,

where: Y_i = the yield of a radiation, i, per nuclear transformation,

E_i = its energy in MeV,

$AF(T \rightarrow T)$ = 1.0 for alpha particles emitted in the target organ,

Q_i = the quality factor for alpha particles (20),

and M_T = mass of target organ in grams = 1000 for lung.

For ^{226}Ra with 71% radon retention, $\Sigma SEE = 0.372$ MeV g^{-1}.

In this case, $U_T = \int_o^{50} U(t)dt = \int_o^{50} U_o e^{-\lambda t} dt = U_o/\lambda$,

where U_o = 180 Bq, and $\lambda = 6.77 \times 10^{-8}\ sec^{-1}$ (120-day half-life). Therefore,

$H_{50,T} = 1.6 \times 10^{-10} \times [180/(6.77 \times 10^{-8})] \times 0.372 = 0.16$ Sv.

Propagation of errors results in an estimated standard deviation of 0.04 Sv. The dose limit for non-stochastic effects is 0.5 Sv, and the limit for (weighted) stochastic effects is 0.05 Sv.[4] Since the weighting factor for stochastic effects is 0.12 for the lung, 0.12 × 0.16 Sv = 0.02 Sv, and so this incident did not represent an overexposure.

Perhaps the most interesting aspect of this case, however, was that it demonstrated that uranium mill workers could be exposed to ^{226}Ra free from its precursors. In a recent report on whole-body counting of uranium mill workers, Helgeson reported that of 321 persons whose ^{214}Bi contents were measured, 77, or 24%, had amounts greater than the minimum detectable.[5] Although the minimum detectable amount was not specified, it may be presumed to be 75 or 100 Bq. Measurements at the mill site were difficult to interpret because of high background counting rates and the possibility that ^{214}Bi observed <u>in vivo</u> may have resulted from the inhalation of radon and its daughters, rather than from an intake of radium.

Body radioactivity measurements in a low background facility and analysis of exhaled breath for radon may be necessary to determine the true levels of internal radium in these workers. Furthermore, excreta samples can be

analyzed for α-emitting isotopes of the ^{238}U and ^{232}Th decay chains in an effort to determine exactly to which isotopes uranium mill workers are exposed, since whole body counting cannot detect ^{230}Th nor establish the ratio of ^{234}Th to its parent ^{238}U.[5]

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GASTROINTESTINAL ABSORPTION OF URANIUM IN MAN

R. P. Larsen and K. A. Orlandini*

A method has been established for determining the fractional absorption of uranium directly in man. Measurements are made of the urinary excretion rates of uranium for individuals whose drinking water has a high ^{234}U to ^{238}U activity ratio and is the primary source of ^{234}U in their diets. For two individuals, the values obtained for the fractional absorption of ^{234}U were 0.004 and 0.006. The values obtained for the fractional absorption of ^{238}U, using a literature value for the ^{238}U intake from food, were 0.008 and 0.015. The present ICRP value is 0.20.

Introduction

The concentration of uranium in the drinking water consumed by about 3% of the U.S. population exceeds 10 pCi/L, a value that the U.S. Environmental Protection Agency (EPA) may propose as a "health effects guidance level."[1] This value is based in part on the ICRP value of 0.2 for f_1, the fraction of uranium transferred from the gut to blood.[2] This value of f_1 is based, to a large extent, on the uranium intakes from foodstuffs as reported by Welford and Baird[3] for the United States and by Hamilton[4] for the United Kingdom, and the excretion rate of uranium for Chicago residents as reported by Welford and Baird[3] and for residents of the United Kingdom as reported by Dean.[5]

The values reported by Welford and Baird and by Hamilton for the amounts of uranium in U.S. and U.K. skeletal remains, 46 and 67 μg, respectively, are consistent with a value of 0.20 for f_1, but the values reported recently by Fisenne et al.[6] for the amounts in Nepalese and Australian remains, 10 and 3 μg, respectively, are not. The possible reasons for the differences in amounts of uranium in skeletal remains would appear to be: (1) the fractional absorption of uranium is indeed about 0.2, and the concentrations of uranium in foodstuffs consumed in Nepal and Australia are much lower than those in foodstuffs consumed in the United States and the United Kingdom, or (2) the value of f_1 for uranium is much lower than 0.2 and there were unidentified sources of uranium in the diets of the individuals from the United States and

*Environmental Effects Research Program.

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United Kingdom. In the U.K. study the concentration of uranium in drinking water was not measured; in the U.S. study it was determined in New York City water and found to be insignificant. Consequently, the concentration was considered to be insignificant in Chicago water.

A strong argument can be made on the basis of existent information that the ICRP value of f_1 for uranium, 0.20, is a factor of 4 too high. This is done by considering just the Chicago population, the only population for which all the parameters are known. The intake from food, 1.5 μg/day, is that reported by Welford and Baird for Chicago residents; the intake from water, 0.42 μg/day, is calculated from the concentration of uranium in Chicago drinking water, 0.28 μg/L; and the excretion rate, 0.09 μg/day, is based on the values for nine Chicago residents as reported by Welford and Baird. (Two of their 11 values were rejected as being statistical outliers.) Using this information, one obtains a value for f_1 of 0.05 (0.047 rounded to one significant figure).

Experimental

A direct measurement of the gastrointestinal absorption of uranium in man can be made by determining the urinary excretion rate of ^{234}U in Chicago suburban-area residents.

The value of the ratio, $R_{4,8}$, of the activities of ^{234}U and ^{238}U in some of these suburban water supplies is as high as 30 to 1, and the concentrations of ^{234}U and ^{238}U in such waters are about 3000 and 100 fCi/L, respectively. If the intakes of this water by the residents of these suburbs were 1.5 L/day, their daily intakes of ^{234}U and ^{238}U from drinking water would be 4500 and 150 fCi, respectively. The value of $R_{4,8}$ in food is known to be 1.1 or less,[6] and the daily intake each of ^{238}U from food and hence that of ^{234}U, have been estimated to be 500 fCi.[3] Thus, the primary source of ^{234}U in the diets of these individuals is drinking water, and the primary source of ^{238}U is food. If an individual has been consuming this water at a consistent and known rate for a protracted period, the excretion rate of ^{234}U will be equal to the intake rate.

The urinary excretion rates of ^{234}U of such individuals can be used to establish the fractional absorption of the uranium that is consumed in drinking water, $^{W}f_1$, and their urinary excretion rates of ^{238}U can be used

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to establish the fractional absorption of uranium consumed in food, ${}^{F}f_1$. The value of ${}^{W}f_1$ is $({}^{W}E_4 + {}^{F}E_4)/({}^{W}I_4 + {}^{F}I_4)$ and the value of ${}^{F}f_1$ is $({}^{W}E_8 + {}^{F}E_8)/({}^{W}I_8 + {}^{F}I_8)$, where E and I are rates of excretion and input, respectively; the superscripts W and F refer to the uranium from water and food, respectively; and the subscripts 4 and 8 identify ${}^{234}U$ and ${}^{238}U$, respectively.

For the individual intakes cited above, it is apparent that the errors in the values of ${}^{W}f_1$ due to the intake of ${}^{234}U$ from food will be small. Over- or underestimating the intake of ${}^{234}U$ from food by a factor of 2 would have an inconsequential effect on the value of ${}^{W}f_1$ since ${}^{W}I_4$ is a factor of about 10 higher than ${}^{F}I_4$, and ${}^{W}E_4$ is hence a factor of about 10 higher than ${}^{F}E_4$. With the value of ${}^{W}f_1$ well established and the intake of ${}^{238}U$ from drinking water known within a factor of 1.5 (due to uncertainty in the individual's estimate of his drinking water consumption), the error in the value of ${}^{F}f_1$ would be dominated by the error in the estimate of ${}^{238}U$ intake from food if the values of ${}^{W}f_1$ and ${}^{F}f_1$ do not differ markedly. The value of ${}^{F}E_8$ would be a factor of 3 larger than ${}^{W}E_8$, and hence the value of ${}^{F}I_8$ would be a factor of 3 larger than ${}^{W}I_8$. It would appear that the intake of ${}^{238}U$ from food would not vary with the individual by more than a factor of 1.5. The range in the values of the intake rates by residents of New York, Chicago, and San Francisco, which were based on the analysis of a wide variety of foodstuffs, was 462 to 523 μg/year, and the variation in concentration with type of foodstuff was not large.

Two 24-hr urine samples from one of the authors (KAO) and one from his wife (DMO) have been analyzed. The value of $R_{4,8}$ in their domestic water is 27, and the ${}^{234}U$ concentration is 1800 fCi/L. A known amount of ${}^{236}U$ was added to each sample, the sample was digested with nitric acid, the solution was evaporated to a small volume, and the uranium was separated by the method reported by Orlandini.[7] The solution is made 1.5 M in aluminum nitrate and 0.5 M in nitric acid, and then passed through a Dowex-1 anion exchange column. The column, which strongly retains the uranium, is washed first with a solution of the same composition and then with a 9 M hydrochloric acid solution, and the uranium is eluted with 1 M hydrochloric acid. The uranium is then electrodeposited and the planchets assayed in an alpha spectrometer for about one week.

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Results and Discussion

The data obtained (Table 1) are obviously quite limited, but they suggest strongly that the value of f_1 for uranium is in the order of 0.005 to 0.01 rather than 0.20 (the EPA value), or 0.05 (the value calculated above from the Welford and Baird data). Our values for the urinary excretion rate of ^{238}U range from 3 to 9 fCi/day, with an average value of 6 fCi/day, while those of Welford and Baird average 30 fCi/day (range 20 to 40 fCi/day, two outliers again being excluded). The difference between their value for f_1 and ours may be the consequence of our having a very limited data set. The number of subjects in our study should obviously be increased. It also appears that the value of f_1 should be based on the data for ^{234}U rather than that for ^{238}U, the reliability of the intake rate of ^{234}U being quite superior to that of ^{238}U.

TABLE 1. Uranium gastrointestinal absorption in two human subjects based on their dietary intakes and urinary excretions of ^{234}U and ^{238}U.

Subject	Uranium isotope	Intake, fCi/day			Excretion, fCi/day	Percent absorption
		Food[a]	Water[b]	Total		
KAO-1	234	500	1800	2300	9 ± 1	0.4
	238	500	70	570	3 ± 0.5	0.5
KAO-2	234	500	1800	2300	7 ± 0.7	0.3
	238	500	70	570	6 ± 0.6	1.1
DMO	234	500	2700	3200	18 ± 1	0.6
	238	500	100	600	9 ± 0.6	1.5

[a] From Welford and Baird.[3]

[b] Based on the subjects' estimates of their water consumptions.

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