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THE ESTIMATION AND POTENTIAL RADIOBIOLOGICAL SIGNIFICANCE OF THE INTAKE OF ^{228}Ra BY EARLY Ra DIAL WORKERS IN ILLINOIS†

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Abstract—We made radiochemical determinations of ^{226}Ra and the ^{228}Ra-decay product, ^{228}Th, in samples of bone from former Ra dial workers who belonged to a major cohort of Ra-exposed persons under study for health effects at our institution. Most of the former workers were long-term residents of two communities supplied with drinking water containing elevated natural levels of ^{228}Ra and ^{226}Ra, so determinations also were made of radioactivity in samples of bone from long-term residents not occupationally exposed to Ra. The ^{228}Th activity of the bones of the former workers, after correction for the presence of natural radioactivity, showed that some had significant occupational intakes of ^{228}Ra, contrary to published reports that ^{228} Ra was never used by the Illinois company that had employed the cohort of early workers. For 14 workers hired in the years 1920–23, the calculated ratio of the occupational intake of ^{228}Ra to ^{226}Ra activity averaged 0.15 (coefficient of variation 0.65), whereas for three workers hired in 1924, it was not significantly different from zero (mean 0.05, coefficient of variation 1.5). The risk of radiogenic cancer for the typical worker hired before 1924 may have been nearly twice that incurred in the absence of the ^{228}Ra component of the Ra intakes.

INTRODUCTION

THE FORMER employees of the Radium Dial Company, which operated dial-painting studios in Illinois from 1918 to 1936, constitute one of the major cohorts of Ra-exposed persons under study for health effects at the Center for Human Radiobiology (CHR) (Ro78b; Ru86). Both ^{228}Ra (half-life 5.75 y) and ^{226}Ra (half-life 1600 y) were used in Ra-activated luminous compounds in the early years of the dial painting industry (Au52), but earlier studies reported contradictory anecdotal data on the use of ^{228}Ra by the Radium Dial Company, i.e., ^{228}Ra was used before 1926, probably in 1920, but not after 1926 (Au52) and ^{228}Ra was never used (Fi69).

The determination of the residual activities of both isotopes of Ra is important not only because absorbed doses to the various body organs following the uptake of ^{228}Ra generally exceed those following the uptake of a comparable activity of ^{226}Ra (Ke83), but also because the effectiveness of the α-emitting decay products of β-emitting ^{228}Ra in producing bone sarcomas appears to exceed that of ^{226}Ra and its decay products for equal absorbed doses to bone (Ro78a). In this paper, we present results of measurements of radioactivity in former Radium Dial Company workers and in control subjects, estimate the relative occupational intakes of ^{228}Ra and ^{226}Ra by the workers, and examine the radiobiological significance of the ^{228}Ra intakes.

MATERIALS AND METHODS

Exposure period

Information gleaned from former workers suggested the following dates of operation of the Radium Dial Company studios in the four Illinois

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cities or towns where they were located: Chicago—from 1918 to 1920 or 1921; Peru—from 1920 or 1921 to August 1922; Ottawa—from September 1922 to December 1936; and Streator—about 9 mo in 1925. Some of the former workers transferred to a dial-painting studio founded in 1934 in Ottawa by another company. Data on the employment periods of former workers were obtained from personal interviews, sometimes corroborated by listings of employees supplied by the company and published in city directories, or by the identification of employees in dated group photographs.

By 1925, mouth-tipping of the brushes used by the workers to apply the luminous compound had been implicated as the chief mode of intake of radium (Ca25; Ho25; Mar25). This practice was largely discontinued by 1926 (USBL29), and significant residual activities of Ra are comparatively rare in former workers hired after 1925 (Ro78b; Ru86). Consequently, all Ra intakes by the workers hired before 1926 were assumed to have occurred during the interval between the date of hire and the end of employment or 31 December 1925, whichever came first.

Radioactivity measurements

Whole-body ^{226}Ra and ^{228}Ra decay-product activities were determined *in vivo* at the CHR in 113 women hired by the Radium Dial Company before 1926. The techniques used at CHR to determine whole-body ^{226}Ra and ^{212}Bi (half-life 1.0 h) activity *in vivo* have been described by Toohey *et al.* (To83). Estimates of whole-body ^{228}Ra activity *in vivo* were inferred from determinations of whole-body ^{212}Bi (Ke84).

Radium-226 and the ^{228}Ra decay product ^{228}Th (half-life 1.91 y) were determined radiochemically by CHR or its predecessors (Ev58; Ev60; Ev66; Mi69b) in a total of 48 samples of bones or teeth, acquired at surgery, autopsy or exhumation, from 17 women hired at a median age of 18 (range 15 to 30) by the Radium Dial Company before 1925. Radium-226 activities in samples of bone were determined by dissolving the ashed bone in nitric acid, passing N through the solution to remove Rn, and subsequently counting the α particles emitted by the collected ^{222}Rn (half-life 3.823 d) and its decay products (Au52; Lu61). Thorium-228 activities in samples of bone were determined either by passing N through the ashed-bone solution and subsequently counting the α particles emitted by the electrostatically collected decay products of ^{220}Rn (half-life 55 s; Au52; Ku76) or by extracting ^{228}Th into dibutyl phosphate and counting the α particles emitted by the ^{228}Th-decay product ^{224}Ra (half-life 3.62 d) stripped from the organic phase (St56; Lu74).

The estimation of the occupationally derived activities of ^{226}Ra and especially of ^{228}Ra in the bones of former Ra dial workers often is limited not by the sensitivity and specificity of the radiochemical method, but rather by the magnitude and the variability of the natural levels of these isotopes in bone. The 17 former workers whose bone samples were analyzed radiochemically could be placed in three categories with respect to levels of dietary intake of Ra. At Level I were seven former workers who had been long-term residents of Ottawa, IL, a town supplied with untreated drinking water pumped from deep wells. At Level II were three former workers who had been long-term residents of Peru, IL, a town supplied with treated drinking water pumped from deep wells. At Level III were seven former workers who had been long-term residents of communities located in surficial drinking-water regions, for whom food rather than water would have been the principal source of dietary Ra (Lu61).

Natural radioactivity in bone for persons at Level I was determined radiochemically in 2–10 g samples of ashed rib from 12 long-term Ottawa residents not occupationally exposed to Ra. Natural radioactivity in bone for persons at Level II was estimated from the values for Levels I and III and the radioactivity concentrations measured by Lucas in one well-water and one tap-water sample from Peru, and in four well-water samples from Ottawa (Lu85). Natural radioactivity in bone for persons at Level III was estimated from the skeletal ^{226}Ra content of Reference Man (ICRP75), the population distribution of the natural ^{226}Ra content of bone determined by Walton *et al.* (Wa59), and the natural ^{228}Th content of bone determined by Lucas and Markun (Lu74) and by Wrenn *et al.* (Wr81; Ib83). A portion of the natural ^{228}Ra content of bone arises from the natural ^{232}Th activity of bone (Lu70; Wr81; Ib83).

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Data analysis

Equations used in the computation of the results are presented in the Appendix. These equations express: (i) the time-dependence *in vivo* and postmortem of the ratio of the activity of ^{228}Th to the activity both of its natural and of its occupationally derived ^{228}Ra parent; (ii) the time-dependence of the ratio of occupationally derived ^{228}Ra to ^{226}Ra activity in bone; and (iii) the propagation of statistical errors. Although the variability in the natural Ra activity of skeletons is compatible with a lognormal distribution (Wa59), the parameters of the normal distribution adequately predict the upper limit of 90% confidence of the natural ^{226}Ra and ^{228}Ra activity of bone at each level of dietary intake. Therefore, for computational convenience in determining residual occupational radioactivity in the bones of the former dial workers, the arithmetic mean and standard deviation were used as parameters of the population distribution of natural radioactivity in bone.

RESULTS AND DISCUSSION

Natural radioactivity of bone

Table 1 shows the natural ^{226}Ra and ^{228}Ra activities of bone determined radiochemically in the 12 persons at Level I who had not been occupationally exposed to Ra. The specific ^{226}Ra and ^{228}Ra activities were highly correlated ($r = 0.94$, $P < 10^{-4}$). Regression of the specific ^{226}Ra activity on length of residence and on age indicated that the latter two variables would not contribute significantly as estimators of natural radium activity in the former workers. Estimates of the natural radioactivity of drinking water and bone for the three levels of dietary Ra intake are summarized in Table 2.

Radioactivity in bone samples of former workers

The results of the radiochemical analyses of the samples of bone of the 17 former Radium Dial Company workers are presented in Table 3, ordered by case number. The gross results in Table 3 for several samples that were analyzed by one of us (HFL) before the inception of CHR had been previously reported by Miller *et al.* (Mi69b). Samples analyzed by Evans *et al.* (Ev58; Ev60; Ev66) are also referenced accordingly.

The gross ^{226}Ra activities in Table 3 are sufficiently above natural levels to be ascribable virtually entirely to occupational origin. Equation (1) of the Appendix was used to calculate the expected natural ^{228}Th values. Equations (2–5) were

Table 1. Natural ^{226}Ra and ^{228}Ra in bone of residents of Ottawa, IL. In deriving the ^{228}Ra activities from the measured ^{228}Th activities, ^{228}Ra and ^{228}Th were assumed to have been at equal activities in bone at the time of sampling. Entries preceded by ± are standard errors

Age of resident (y)	Length of residence (y)	^{226}Ra (Bq*/kg ash)	^{228}Ra (Bq/kg ash)
28	28	2.0 ± 0.22	0.89 ± 0.19
44	44	5.6 ± 0.19	2.2 ± 0.11
47	34	5.2 ± 0.19	1.3 ± 0.11
50	50	4.4 ± 0.19	2.1 ± 0.37
53	53	13 ± 0.74	7.0 ± 0.44
53	53	3.2 ± 0.22	0.78 ± 0.11
54	54	5.9 ± 0.22	2.4 ± 0.26
64	33	1.6 ± 0.11	0.85 ± 0.15
65	30	3.5 ± 0.15	2.4 ± 0.15
70	18	3.7 ± 0.19	0.56 ± 0.07
75	42	3.7 ± 0.19	0.59 ± 0.07
79	55	2.2 ± 0.15	0.59 ± 0.11

*One Bq = 27.0 pCi

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Table 2. Natural ^{226}Ra *and* ^{228}Ra *in drinking water and in bone. Entries preceded by ± are standard deviations*

Level of dietary radium intake	Activity concentration in drinking water (Bq/m^3)		Specific activity of bone (Bq/kg ash)	
	^{226}Ra	^{228}Ra	^{226}Ra	^{228}Ra, ^{228}Th[c]
I	200 ± 36[a]	130 ± 44[a]	3.7 ± 1.4[d]	1.3 ± 0.78[d]
II	46[a]	25[a]	1.1 ± 0.78[e]	0.37 ± 0.24[e]
III	< 1[b]	(< 1)[b]	0.37 ± 0.18[f]	0.15 ± 0.092[g]

[a]Lu85;Lu61. Level-I water is untreated; Level-II water is treated. For Level II, the ^{226}Ra concentration was determined in 1 sample of Peru tap water (Lu85, Table 2) whereas the ^{228}Ra concentration was estimated by us using the ratio of ^{228}Ra to ^{226}Ra activity (0.54) determined in 1 sample of Peru raw water (Lu85, Table 4).
[b]NCRP75
[c]It is assumed that ^{228}Ra and ^{228}Th are at equal activities. Of these activities, it is assumed that 0.04 ± 0.03 Bq per kg ash are in secular radioactive equilibrium with ^{232}Th (Lu70;Wr81;Ib83).
[d]Table 1, outlier excluded (13 Bq ^{226}Ra per kg ash; 7.0 Bq ^{228}Ra per kg ash).
[e]Bone Ra III + $\frac{\text{Water Ra II}}{\text{Water Ra I}}$ (Bone Ra I - Bone Ra III).
[f]Wa59;ICRP75.
[g]Lu74;Wr81;Ib83.

used to calculate the ratios of occupationally derived ^{228}Ra to ^{226}Ra activity. The predicted ^{228}Th to ^{228}Ra activity ratios at the time of sampling (eqn 2 and Fig. 1, Appendix) that were used in computing the occupationally derived ^{228}Ra activities (eqns 3 and 4, Appendix) ranged from 1.96 to 1.56, depending on the time from intake to sampling. Although the standard errors of the ^{226}Ra results are not shown in Table 3 (relative errors were generally <3%), these errors were propagated with others in determining the standard errors of the occupational ratios (eqn 6, Appendix).

Intake ratios based on radioactivity in bone samples

Column 4 of Table 4 lists the weighted mean (eqns 7 and 8, Appendix) of the ratios in Column 7 of Table 3 for each worker except Case 03-455, the only former worker for whom samples of bone were obtained at two different times after intake, and Cases 03-488 and 03-649. For the latter two workers, the gross ^{228}Th and ^{226}Ra activities in Table 3 were amenable to least-squares analysis, so eqn (9) of the Appendix was used to determine both the natural ^{228}Ra activity† and the ratio of occupationally derived ^{228}Ra to ^{226}Ra activity in the bones of these two workers at the time of sampling.

Column 5 of Table 4 lists the ratio of the occupational intake of ^{228}Ra to ^{226}Ra activity for each worker, computed by use of eqns (10) and (11) of the Appendix from the mean ratio at the time of sampling in Column 4. The entries in Table 4 are ordered by the dates of hire of the

† The best-fit values of natural ^{228}Ra at the time of sampling were 2.4 ± 0.33 Bq per kg ash for Case 03-488 and 0.78 ± 1.4 Bq per kg ash for Case 03-649. These least-squares estimates are congruous with the Level-I value of 1.3 ± 0.78 Bq ^{228}Ra per kg ash (Table 2) used in eqn (1) to derive the expected natural ^{228}Th values that are shown in Column 5 of Table 3 for these two long-term Ottawa residents.

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Table 3. Gross ^{228}Th *and* ^{226}Ra *activity and expected natural* ^{228}Th *activity of bone at the time of radiochemical analysis, together with computed ratios of occupationally derived* ^{228}Ra *to* ^{226}Ra *activity at the time of sampling. Entries preceded by ± are standard errors. Roman numerals following the case numbers in column 1 designate levels of dietary* Ra *intake, as discussed in the text*

Case no. and sample description	Sample size (g ash)	Time elapsed from sampling to analysis (y)	^{228}Th at time of analysis: Gross (Bq/kg ash)	^{228}Th at time of analysis: Expected natural (Bq/kg ash)	^{226}Ra at time of analysis (Bq/kg ash)	Ratio (x100) of occupational ^{228}Ra to ^{226}Ra activity at time of sampling
01-002 (I)						
Tooth (Ev60,66)	0.44	21.31	130 ± 26	0.18 ± 0.10	81 400	1.4 ± 0.28
01-454 (III)						
Trabecular bone (cremation ash)	21.6	7.56	20 ± 1.4	0.10 ± 0.06	61 400	0.054 ± 0.0039
01-562 (III)						
Innominate bone	5.1	42.63	112 ± 12	0.04 ± 0.03	354 000	3.5 ± 0.39
Femur, prox. metaphysis	6.2	42.65	28 ± 2.0	0.04 ± 0.03	102 000	3.1 ± 0.24
Femur, dist. metaphysis	5.9	42.65	31 ± 1.3	0.04 ± 0.03	86 200	4.0 ± 0.21
Femur, distal	9.4	42.70	43 ± 1.6	0.04 ± 0.03	176 000	2.8 ± 0.14
Femur, condyle	14.1	42.72	50 ± 1.8	0.04 ± 0.03	145 000	3.9 ± 0.19
Femur, proximal	23.3	42.73	49 ± 1.6	0.04 ± 0.03	196 000	2.8 ± 0.13
Femur, midshaft	4.7	42.73	25 ± 2.3	0.04 ± 0.03	82 500	3.4 ± 0.33
Vertebra	2.0	43.42	105 ± 3.7	0.04 ± 0.03	370 000	3.5 ± 0.17
03-402 (I)						
Femur (Mi69b)	2.4	3.67	20 ± 1.1	1.1 ± 0.67	20 600	0.092 ± 0.0063
Femur (Mi69b)	3.0	4.42	22 ± 1.8	1.0 ± 0.59	23 200	0.099 ± 0.0091
Femur (Mi69b)	2.9	4.42	23 ± 1.8	1.0 ± 0.59	25 300	0.096 ± 0.0084
Femur (Ev58,60)	1.5	6.30	24 ± 4.8	0.83 ± 0.48	28 200	0.11 ± 0.023
Femur	0.052	17.20	13 ± 4.1	0.27 ± 0.15	28 900	0.23 ± 0.074
03-407 (I)						
Tibia	4.2	10.15	1.1 ± 0.11	0.59 ± 0.33	9 100	0.013 ± 0.0089

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TABLE 3 (Contd.)

Case no. and sample description	Sample size (g ash)	Time elapsed from sampling to analysis (y)	^{228}Th at time of analysis: Gross (Bq/kg ash)	^{228}Th at time of analysis: Expected natural (Bq/kg ash)	^{226}Ra at time of analysis (Bq/kg ash)	Ratio (x100) of occupational ^{228}Ra to ^{226}Ra activity at time of sampling
03-455 (III)						
Femur, head	6.85	0.86	5.55 ± 0.560	0.15 ± 0.092	7 960	0.049 ± 0.0052
Ulna	0.57	36.71	25.5 ± 0.630	0.04 ± 0.03	17 200	8.1 ± 0.32
03-479 (I)						
Toe, phalanx (Mi69b)	1.4	0.19	1.6 ± 0.22	1.3 ± 0.78	920	0.021 ± 0.057
03-487 (II)						
Vertebral body	2.1	15.78	1.7 ± 0.81	0.11 ± 0.063	5 250	0.14 ± 0.071
03-488 (I)						
Vertebra, lumbar	6.7	0.06	3.1 ± 0.52	1.3 ± 0.78	180	0.54 ± 0.31
		0.29	2.5 ± 0.41	1.3 ± 0.78		
		2.07	2.4 ± 0.70	1.2 ± 0.74		
Femur, midshaft	13.8	2.07	2.2 ± 0.22	1.2 ± 0.74	470	0.18 ± 0.14
Rib	1.0	3.60	4.4 ± 2.0	1.1 ± 0.63	2 500	0.13 ± 0.083
Iliac crest	1.4	3.65	2.7 ± 1.5	1.1 ± 0.63	600	0.27 ± 0.27
03-528 (III)						
Tibia, proximal	1.3	0.49	1.2 ± 1.1	0.15 ± 0.092	2 620	0.027 ± 0.028
03-547 (III)						
Rib	2.5	9.20	0.78 ± 0.11	0.089 ± 0.056	560	0.25 ± 0.045
03-619 (III)						
Femur	2.2	9.04	6.3 ± 0.37	0.092 ± 0.056	8 440	0.14 ± 0.0087

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TABLE 3 (Contd.)

Case no. and sample description	Sample size (g ash)	Time elapsed from sampling to analysis (y)	^{228}Th at time of analysis: Gross (Bq/kg ash)	^{228}Th at time of analysis: Expected natural (Bq/kg ash)	^{226}Ra at time of analysis (Bq/kg ash)	Ratio (x100) of occupational ^{228}Ra to ^{226}Ra activity at time of sampling
03-648 (II)						
Long bone (Ev66)	1.7	1.33	200 ± 22	0.36 ± 0.23	59 900	0.25 ± 0.028
Femur (Mi69b)	3.8	1.75	280 ± 1.8	0.35 ± 0.23	49 600	0.44 ± 0.007
Femur (Mi69b)	3.9	1.75	280 ± 1.8	0.35 ± 0.23	59 600	0.37 ± 0.006
Femur (Mi69b)	3.7	2.17	260 ± 1.9	0.34 ± 0.22	50 700	0.42 ± 0.007
Femur (Mi69b)	4.2	2.17	310 ± 2.0	0.34 ± 0.22	54 800	0.47 ± 0.008
03-649 (I)						
Femur (Mi69b)	1.4	1.81	2.2 ± 0.37	1.2 ± 0.74	26 900	0.0029 ± 0.0024
Femur (Mi69b)	1.4	1.89	0.74 ± 0.37	1.2 ± 0.74	19 200	-0.0019 ± 0.0034
Femur (Mi69b)	1.4	2.31	0.74 ± 0.37	1.2 ± 0.70	10 400	-0.0037 ± 0.0064
Vertebra (Mi69b)	1.2	3.00	3.0 ± 0.37	1.2 ± 0.70	52 400	0.0031 ± 0.0014
Femur, epiphysis (Mi69b)	3.0	4.08	4.1 ± 0.37	1.0 ± 0.63	25 100	0.013 ± 0.0031
Ulna, shaft (Ev58,60,66)	0.96	5.77	7.4 ± 3.7	0.90 ± 0.52	5 990	0.14 ± 0.080
03-666 (I)						
Femur, midshaft	11.4	49.15	80 ± 5.6	0.045 ± 0.030	259 000	7.7 ± 0.60
Vertebra	3.0	49.15	340 ± 22	0.045 ± 0.030	780 000	11 ± 0.80
Vertebra	1.0	49.18	390 ± 31	0.045 ± 0.030	1 050 000	9.1 ± 0.78
03-671 (II)						
Foot (Mi69b)	1.5	5.44	360 ± 1.6	0.27 ± 0.17	110 000	0.41 ± 0.0066
Foot (Mi69b)	1.6	5.44	360 ± 1.4	0.27 ± 0.17	99 500	0.45 ± 0.0070
Femur (Ev58,60)	1.0	7.31	170 ± 18	0.23 ± 0.14	22 900	1.2 ± 0.15
03-779 (III)						
Femur, midshaft	10.8	37.34	0.14 ± 0.12	0.042 ± 0.030	18 100	0.032 ± 0.040
Femur, midshaft	15.2	37.35	0.056 ± 0.081	0.042 ± 0.030	31 700	0.0026 ± 0.016
Femur, prox. metaphysis	8.3	38.25	0.16 ± 0.21	0.042 ± 0.030	25 100	0.031 ± 0.056
Femur, dist. metaphysis	8.7	38.25	0.29 ± 0.26	0.042 ± 0.030	27 800	0.059 ± 0.062

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Table 4. Ratios of intake of ^{228}Ra to ^{226}Ra activity for Radium Dial Company workers, based on activities of ^{228}Th and ^{226}Ra determined radiochemically in bone. Entries preceded by ± are standard errors

Case no.	Employment period (d mo y)	Average time from intake to sampling† (y)	Mean ratio (x100) of occupationally acquired ^{228}Ra to ^{226}Ra activity at sampling	Ratio (x100) of occupational intake of ^{228}Ra to ^{226}Ra activity
01-454	1Jul20-31Dec36	47.7	0.054 ± 0.0090	17 ± 2.9
01-562	1Jul21-1Jun22*	10.0	3.2 ± 0.18	11 ± 0.62
03-648	2Jan22-26Dec24	33.2	0.42 ± 0.028	23 ± 1.6
03-671	1Jul22-31Aug22	30.4	0.43 ± 0.039	17 ± 1.6
03-779	1Jul22-31Aug22	20.0	0.011 ± 0.014	0.1 ± 0.1
03-528	1Jul22-1Jul32	55.3	0.027 ± 0.028	21 ± 22
03-488	1Sep22-1Mar23	52.2	0.027 ± 0.067	14 ± 35
03-619	1Sep22-1Apr23	39.2	0.14 ± 0.023	16 ± 2.7
03-455	1Sep22-1Oct23	11.3 54.5	8.1 ± 1.3 0.049 ± 0.0090	32 ± 3.9
01-002	1Sep22-5Nov36	14.7	1.4 ± 0.35	8.2 ± 2.1
03-666	1Jan23-6Aug29	5.12	8.9 ± 0.88	16 ± 1.6
03-547	1Jul23-1Aug25	37.5	0.25 ± 0.059	23 ± 5.5
03-402	1Jul23-1Jul28	29.3	0.095 ± 0.0084	3.2 ± 0.29
03-407	1Jul23-1Jul46	36.3	0.013 ± 0.0091	1.0 ± 0.70
03-479	1Jun24-1Dec24	34.5	0.021 ± 0.057	1.3 ± 3.5
03-487	1Jul24-30Oct36	38.8	0.14 ± 0.074	15 ± 7.9
03-649	1Jul24-1Jul51	29.3	0.004 ± 0.004	0.1 ± 0.1

*Case 01-562 may have worked previously at the studio in Chicago.

†Radium intakes were assumed to have occurred in the interval between the date of hire and the end of employment or 31 December 1925, whichever came first.

workers. If only a year was reported as a date of hire, a date of 1 July of that year was assumed, except for persons hired at Ottawa in 1922, for whom a date of 1 September was assumed. The sampling times (Column 3 of Table 4) used in eqn (10) were computed from the midpoints of the corresponding intake periods.

The activity ratios in Column 7 of Table 3 for the workers with multiple samples displayed a weighted mean coefficient of excess variation of 0.15. Consequently, in determining the standard errors of the weighted mean ratios in Column 4 of Table 4 for each worker other than Cases 03-488 and 03-649 (the two former workers for whom the activity ratio in Column 4 of Table 4 was determined by the method of least squares), an excess relative standard error of $0.15\ n^{-0.5}$ was propagated with the predicted relative standard error, where n is the number of samples that were analyzed for the worker (eqn 8, Appendix). Un-

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Table 5. Ratios of intake of ^{228}Ra *to* ^{226}Ra *activity for Radium Dial Company workers, based on body activities of* ^{212}Bi *and* ^{226}Ra *determined* in vivo. *Entries preceded by* ± *are standard errors*

Year of intake (mid-interval)	No. of workers	Mean ratio (x100) of occupational intake of ^{228}Ra to ^{226}Ra activity (a)	(b)
1919	2	- 280 ± 1800	- 640 ± 1900
1922	19	- 4.9 ± 26	- 27 ± 26
1923	20	28 ± 16	21 ± 16
1924	43	- 1.0 ± 5.5	- 3.7 ± 5.5
1925	29	- 3.7 ± 6.5	- 5.5 ± 6.8

(a) All γ-ray results were corrected for γ-ray equivalents of 1.9 ± 1.4 Bq ^{212}Bi and 15 ± 2.6 Bq ^{214}Bi, determined in vivo in normal control subjects. In addition to the natural ^{226}Ra and ^{228}Ra activity, body ^{40}K and inhaled ^{222}Rn, ^{220}Rn, and short-lived decay products are the principal sources of the normal ^{214}Bi and ^{212}Bi γ-ray equivalents.

(b) All γ-ray results were corrected for expected γ-ray equivalents of 4.2 ± 2.1 Bq ^{212}Bi and 17 ± 2.7 Bq ^{214}Bi in unexposed residents of Ottawa.

identified analytical errors as well as biological variability could have contributed to the excess variance.

Comparative metabolism of isotopes

Implicit in the calculation of the intake ratios was the assumption that the biological retention of the two Ra isotopes was the same. Limited confirmation of the validity of this assumption was obtained from the results of the radiochemical analysis of the two bone samples of Case 03-455, a sample of ulna surgically removed at 11 y after intake and a sample of femur surgically removed at 54 y. Nearly identical values of the intake ratio were computed from the activities in the two bones at the time of sampling, suggesting that the ratio of the activities of the two radium isotopes in the skeleton during the 43-y interval between bone samplings was significantly altered only by radioactive decay at the differential rate $\lambda_M - \lambda_R$ (eqn 10, Appendix), which reduced the occupationally acquired ^{228}Ra to ^{226}Ra mass ratio from 3.1×10^{-4} at 11 y after intake to 1.8×10^{-6} at 54 y.†

Intake ratios based on radioactivity in vivo

Equation 10 of the Appendix was used to determine the intake ratios from the body activities of ^{228}Ra and ^{226}Ra measured *in vivo* at CHR in 113 former workers. Table 5 shows the weighted means of the ratios grouped according to the year in which the intake period was centered. We did

† The sample of ulna of Case 03-455 was analyzed for ^{228}Th by dibutyl phosphate extraction, whereas the sample of femur was analyzed by electrostatic collection of ^{220}Rn-decay products. The few other available comparisons of ^{228}Th activities determined by the two methods in different bone samples from the same worker similarly suggested that the two methods yielded comparable results. Of the two methods, dibutyl phosphate extraction had the lower detection limit, partly because of lower and less variable blank values.

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not attempt to correct for natural radioactivity in each former worker according to place of residence. Rather we derived two sets of intake ratios, one after corrections were applied for body radioactivity determined in typical unexposed subjects, and the other after corrections were applied for the greater natural radioactivity expected in Ottawa residents. Data were excluded for one person, Case 03-499, the only former worker for whom results *in vivo* at CHR were unambiguously positive for a body activity of ^{212}Bi, and for whom the intake ratio of 1.2 was greater by an order of magnitude than those based on the radiochemical determinations in colleagues.§ Miller *et al.* reported that this person had probably received ^{228}Ra medically (Mi69a).

The mean ratios in Table 5 agree statistically with the ratios derived from the radiochemical determinations. It is not surprising that none of the means is significantly different from zero. Of the 113 body ^{226}Ra activities measured *in vivo*, 47% were <0.37 kBq and 80% were <3.7 kBq. The intake ratios in Table 4 suggest that, at the times of 45 to 60 y after intake when body radioactivity in the 113 former workers was determined *in vivo* at CHR, body ^{212}Bi activity would have been of the order of 0.2% or less of body ^{226}Ra activity, i.e. <0.7 Bq in 47% and <7 Bq in 80% of the former workers. These expectations are well below the minimal detection limit (2σ) of about 20 Bq body ^{212}Bi activity when no significant ^{226}Ra decay-product activity is present.

CONCLUSIONS

Intake ratio vs. year of hire

The use of ^{228}Ra at the studio in Peru in the summer of 1922 is suggested by the data in Table 4 for Case 03-671; the use of ^{228}Ra at the studio in Ottawa after 1922 is suggested by the data in Table 4 for Case 03-666. The data in Tables 4 and 5 suggest that ^{228}Ra may not have been used after 1923. Arithmetic means and coefficients of variation were computed for the intake ratios in Table 4 for groups of workers according to date of hire. From these data, the following conclusions were drawn: (i) the ratio of the intake of ^{228}Ra to ^{226}Ra activity averaged 0.15, with a coefficient of variation of 0.65, for workers hired at Peru or Ottawa before 1924; and (ii) the intake of ^{228}Ra activity by workers hired after 1923 was probably negligible. For the intake ratios of the workers hired before 1924, the mean of 0.15 and coefficient of variation of 0.65 provide an adequate description of the observed distribution, even though a 6% occurrence of negative ratios is implied.

In the application of these results to the dosimetry of the former workers measured *in vivo*, bias would be introduced if the 17 former workers in Table 4, most of whom showed severe adverse health effects of high intakes of Ra activity, were not a random sample of the Radium Dial Company cohort with respect to the relative intakes of ^{228}Ra and ^{226}Ra by year of hire. Although the intake ratios in Table 4 for the former workers hired before 1923 showed a negative correlation with the estimated intake of ^{226}Ra activity (CHR85; the data of Case Nos. 03-488 and 03-528 were excluded because their ratios have large standard errors), this relationship was not statistically significant (Spearman $r = -0.26$, $P < 0.5$). Apart from the anomalously high intake ratio for Case 03-499 based on body radioactivity measured *in vivo*, we found no evidence to counter a suggestion that the distribution of the intake ratios of the 17 former Radium Dial Company workers by year of hire is representative.

Radiobiological significance

If ^{228}Ra decay products do not translocate *in vivo*, and if ^{226}Ra decay-product activity in soft tissue is not significant, the α-particle doses to the various tissues and organs other than bone in a 50-y period following an intake of ^{228}Ra would be 5.8 times those following the intake of an equal activity of ^{226}Ra (Ke83). For the Radium Dial Company workers hired before 1924, for whom the ^{228}Ra intake averaged 0.15 Bq per Bq ^{226}Ra intake, the dose to soft tissues from the decay of ^{228}Ra would have been (5.8)(0.15), or 0.87 times the soft-tissue dose from the decay of ^{226}Ra. If we neglected the probable ^{228}Ra intakes of those Radium Dial Company workers hired

§ Case 03-499 allegedly was hired in 1924, but she may actually have been hired earlier. The relatively high residual body ^{228}Ra activity in this former worker is especially at variance with the relatively negligible intakes of ^{228}Ra by the three workers hired in 1924 (Table 4) and by the cohort of workers whose periods of intake were centered in 1924 (Table 5).

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before 1924 whose retained body ^{212}Bi activities were below the limit of detection, we might understate their soft-tissue dose commitment by a factor of (1 + 0.87), or about 1.9.

In addition to the implication for soft-tissue health effects, the ^{228}Ra component of the intake of Ra activity by the Radium Dial Company workers hired before 1924 also carries with it a similar implication for bone-cancer risk. The relationship found by Rowland *et al.* (Ro78a; Ro83) between the incidence rate I of bone cancer among former Ra dial workers and the Ra intake D is expressed by

$$I = (C + \beta D^2)e^{-\gamma D},$$

where C is the natural incidence rate of bone cancer, β and γ are rate constants, and $D = D_{226} + 2.5D_{228}$, where D_{226} and D_{228} are the intakes of ^{226}Ra and ^{228}Ra activity, respectively.† At a combined ^{226}Ra and ^{228}Ra intake such that $e^{-\gamma D}$ is close to unity ($\gamma = 3 \times 10^{-8}$ Bq^{-1}; Ro83), the expected risk rate of radiogenic bone cancer incurred by a Radium Dial Company worker hired before 1924 ($D_{228} = 0.15D_{226}$) would be $\beta([1 + (2.5)(0.15)]D_{226})^2$, which again is 1.9 times the risk rate βD_{226}^2 incurred in the absence of the minor ^{228}Ra component.

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† The ^{228}Ra to ^{226}Ra effectiveness ratio of 2.5 for bone cancer was determined by averaging the results of one theoretical and two empirical assessments of the relative carcinogenicity of the two isotopes (Ro78a). The effect on the empirical evaluation of the relative carcinogenicity of having ignored the probable ^{228}Ra intakes of a portion of the Radium Dial Company cohort was probably small because the radiochemical analyses described in the present work had provided estimates of ^{228}Ra intake for seven of the 10 former Radium Dial Company workers developing bone cancer, as well as for several other Radium Dial Company workers at significant risk for bone cancer who were included among the former dial workers whose bone-cancer experience was evaluated.

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APPENDIX

Time dependence of natural ^{228}Th *activity in bone*

We assumed the following: (i) dietary ^{228}Ra, including ^{228}Ra in drinking water, and natural ^{232}Th (half-life 1.4×10^{10} y) in the body were the only significant sources of natural ^{228}Th in the body; (ii) ^{228}Th was not biologically cleared; (iii) ^{228}Ra arising from the activity of natural ^{232}Th in bone was in secular radioactive equilibrium with ^{232}Th at the time of analysis; and (iv) natural ^{228}Ra and its ^{228}Th daughter were at equal activities in bone at the time of sampling (i.e. at surgery or death) as a consequence of the virtual constancy of natural body ^{228}Ra activity.

Radium-228 and ^{228}Th activities in a bone sample do not remain constant with time after sampling because dietary ^{228}Ra is no longer being supplied. In estimating the ^{228}Ra activity of a bone sample from an unexposed subject, or in computing the expected natural ^{228}Th activity of a bone sample from a former worker, we must consider the dependence of natural ^{228}Th activity on the time after sampling. This dependence is expressed by eqn (1):

$$T'_a = (M'_s - T_{232}) \times (\lambda_T e^{-\lambda_M \tau} - \lambda_M e^{-\lambda_T \tau})/(\lambda_T - \lambda_M) + T_{232}, \quad (1)$$

where T'_a is the natural ^{228}Th activity of bone at the time of analysis, M'_s is the natural ^{228}Ra activity of bone at the time of sampling (Table 2), T_{232} is the natural ^{232}Th activity of bone (Table 2), λ_M and λ_T are, respectively, the radioactive decay constants of ^{228}Ra, (0.1205 ± 0.0006) y^{-1} (May62), and ^{228}Th (0.363 ± 0.0004) y^{-1} (Ki56), and τ is the elapsed time from sampling to analysis. For bones acquired postmortem, the sampling time was considered the time of death.

Time dependence of occupationally derived radioactivity

The time dependence *in vivo* of the ratio ρ of ^{228}Th activity to the activity of its parent is expressed by eqn (2):

$$\rho = \frac{\lambda_T \int_0^t M_0 R(x) e^{-\lambda_M x} \cdot e^{-\lambda_T (t-x)} dx}{M_0 R(t) e^{-\lambda_M t}} = \frac{\lambda_T e^{(\lambda_M - \lambda_T)t}}{R(t)} \int_0^t R(x) e^{(\lambda_T - \lambda_M)x} dx, \quad (2)$$

where t is the time following an uptake of ^{228}Ra of activity M_0, $R(x)$ is the fractional biological retention function for Ra, and $R(t)$ is the value of $R(x)$ at t. The three curves plotted in Fig. 1 were generated by eqn (2). The time dependence of ρ is shown by curve 1 of Fig. 1 for biological retention of Ra according to the alkaline earth model of *ICRP Publication* 20 (ICRP73; Sc82) and by curve 2 of Fig. 1 for biological retention according to the function of Norris *et al.* (No55), if ^{228}Th arising *in vivo* is not excreted. Curve 3 shows the time dependence of the ratio in an originally pure ^{228}Ra source not subject to biological elimination.

Computation of occupationally derived radioactivity in bone

Equation (3) expresses the relationship between the occupationally derived ^{228}Ra activity of bone at sampling time, M_s, and the net ^{228}Th activity of bone at the time of analysis, $T_{obs} - \hat{T}'_a$:

$$M_s = \frac{(\lambda_T - \lambda_M)(T_{obs} - \hat{T}'_a)}{\lambda_T e^{-\lambda_M \tau} - [\lambda_T - (\lambda_T - \lambda_M)\rho_s] e^{-\lambda_T \tau}}, \quad (3)$$

where T_{obs} is the gross ^{228}Th activity and $\hat{T}'_a$ the expected natural ^{228}Th activity of bone at the time of analysis (eqn 1), $T_{obs} - \hat{T}'_a$ is the ^{228}Th activity arising from the occupationally derived ^{228}Ra activity, ρ_s is the predicted ratio of ^{228}Th activity to its occupationally derived parent ^{228}Ra activity at the time of sampling (eqn 2), and τ is the elapsed time from sampling to analysis. Values of ρ_s derived from the biological retention function of Norris *et al.* (curve 2 of Fig. 1) were used in eqn (3), the Norris function providing the more accurate description of late retention in former Ra dial workings retaining >10 Bq per g bone ash (Ru85). It should be noted that errors in the values of ρ_s do not significantly affect the estimates of M_s if the time from sampling to radiochemical analysis is long compared to $1/\lambda_T$, whence the relationship expressed by eqn (3) becomes

$$M_s \simeq (1 - \lambda_M/\lambda_T)(T_{obs} - \hat{T}'_a) e^{\lambda_M \tau}. \quad (4)$$

Equation 5 expresses the relationship between the occupationally derived ^{226}Ra activity of bone at the time of sampling, R_s, and the gross ^{226}Ra activity of bone at the time of analysis, R_{obs}.

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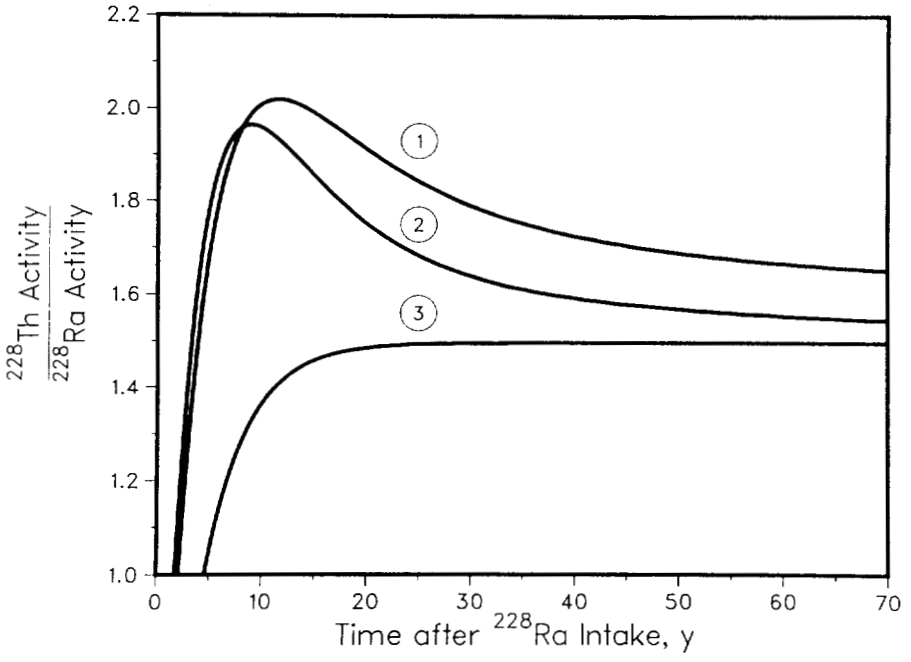

FIG. 1. Time dependence of the ratio of ^{228}Th to ^{228}Ra activity following an intake of ^{228}Ra according to the alkaline earth model of *ICRP Publication* 20 (ICRP73; Sc82; curve 1) and, according to the simple power function of Ra retention of Norris *et al.* (No55; curve 2), if ^{228}Th arising *in vivo* is not excreted. Also shown (curve 3) is the time dependence of the ratio in an originally pure ^{228}Ra source not subject to biological elimination (i.e. *in vitro*).

$$R_s = R_{obs}e^{\lambda_R \tau} - \hat{R}'_s, \tag{5}$$

where $\hat{R}'_s$ is the expected natural ^{226}Ra activity of bone at the time of sampling (Table 2) and λ_R is the radioactive decay constant of ^{226}Ra, 0.00043 y^{-1}.

The ratio of occupationally acquired ^{228}Ra to ^{226}Ra activity at sampling, $(M/R)_s$ (column 7, Table 3), was obtained by dividing M_s (eqn 3 or 4) by R_s (eqn 5). The relative standard error of $(M/R)_s$ is typically principally that of the specific net ^{228}Th activity ($T_{obs} - \hat{T}'_a$) at analysis. There are relatively small contributions to the statistical uncertainty in $(M/R)_s$ from uncertainties in the values of ρ_s, λ_M, λ_T, R_{obs}, and $\hat{R}'_s$. The relative standard errors of the values of these parameters and variables were generally $\ll 1$, so the overall standard error, $S_{(M/R)_s}$, of each value of $(M/R)_s$ in Table 3 was computed as

$$S_{(M/R)_s} \simeq (\sum S_i^2(\partial F/\partial P_i)^2)^{1/2}, \tag{6}$$

where F is the right side of eqn (3) divided by the right side of eqn (5) and the S_i are the standard errors of the parameters and variables P_i. No estimate was made of the standard errors of the values of ρ_s, but for none of the bone samples in Table 3 would probable uncertainties in ρ_s have contributed significantly to the overall uncertainty in $(M/R)_s$. The negligible contribution of the uncertainty in λ_R could be ignored.

Computation of the weighted mean, $(\overline{M/R})_s$

In determining the weighted mean, $(\overline{M/R})_s$, of the n values of $(M/R)_s$ for a given former worker (column 4, Table 4), the inverse square of the standard error of $(M/R)_s$, i.e., $S^{-2}_{(M/R)_s}$, was used for the weight, w. Thus,

$$(\overline{M/R})_s = \frac{\sum [(M/R)_s]_i w_i}{\sum w_i}. \tag{7}$$

The standard error, $S_{(\overline{M/R})_s}$, of $(\overline{M/R})_s$ was computed as

$$S_{(\overline{M/R})_s} = [n^{-1}(0.15(\overline{M/R})_s)^2 + (\sum (S^2_{(M/R)_s} + (n-1)A^2)_i^{-1})^{-1}]^{1/2}, \tag{8}$$

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where $A = S_2(M/R)_s/(T_{obs} - \hat{T}'_a)$, S_2 being the standard error of $\hat{T}'_a$. Because S_2^2 is chiefly a between-skeletons variance of the population of natural ^{228}Th activities at the time of analysis, analyses of multiple bone samples from the same skeleton would not reduce the uncertainty in $(\overline{M/R})_s$ from this source. Hence, we have added to the square of the standard error of $[(M/R)_s]_i$ the derived term $(n-1)A_i^2$ as an approximate correction. The predicted standard error of $(\overline{M/R})_s$, incorporating this correction, is $(\sum (S^2_{(M/R)_s} + (n-1)A^2)_i^{-1})^{-1/2}$. The factor 0.15 in eqn (8) is the coefficient of excess variation, $S_{exc}/(\overline{M/R})_s$, determined from the analysis of certain of the data of Table 3 as mentioned in the main text. The excess variance, S^2_{exc}, had been determined as the difference between the observed variance, S^2_{obs}, and the predicted variance, S^2_{pred}, of the population of ratios $[(M/R)_s]_i$ for a given worker, where

$$S^2_{obs} = \frac{n \sum ([(M/R)_s]_i - (\overline{M/R})_s)^2 w_i}{(n-1) \sum w_i},$$

and $S^2_{pred} = n/\sum w_i$.

Least-squares method

The precision of an estimate of $(\overline{M/R})_s$ is ultimately limited by the uncertainty in the expected value of natural ^{228}Th activity. The ^{228}Th activities determined in several samples of bone from Cases 03-488 and 03-649 were in the range of expected natural levels, and the specific ^{226}Ra content varied widely among the samples, so the specific natural ^{228}Ra activity, M'_s, and the mean ratio of the occupational ^{228}Ra to ^{226}Ra activity in bone at the time of sampling, $(\overline{M/R})_s$, were determined by combining eqns (1), (3) and (5) to form eqn (9), in the expectation that this alternative method might provide a more accurate estimate for these two workers:

$$qT_{obs} + (1-q)\hat{T}_{232} = (1 + (\rho_s - 1)qe^{-\lambda_T \tau}) \times (R_{obs}e^{\lambda_R \tau} - \hat{R}'_s)(\overline{M/R})_s + M'_s, \quad (9)$$

where $q = (\lambda_T - \lambda_M)/(\lambda_T e^{-\lambda_M \tau} - \lambda_M e^{-\lambda_T \tau})$, and $\hat{T}_{232}$ is the expected specific ^{232}Th activity of bone (Table 2).

We carried out a weighted least-squares fit of eqn (9) to the specific ^{228}Th activity, T_{obs}, and ^{226}Ra activity, R_{obs}, determined in the bone samples of each of the two workers, modified by the time-dependent factors shown in eqn (9) before fitting. Thus, the left side of eqn (9) was the dependent variable and the coefficients of $(\overline{M/R})_s$ constituted the independent variable with respect to the least-squares estimation of the slope, $(\overline{M/R})_s$, and the intercept, M'_s. The assumptions that M'_s and $(\overline{M/R})_s$ were constant among the samples are implicit. Values of the dependent variable were weighted by the factor $(q^2S_1^2 + (1-q)^2S_b^2)^{-1}$, where S_1 is the standard error of T_{obs} and S_b is the standard error of $\hat{T}_{232}$. The relative errors of the other known quantities in eqn (9) were comparatively small and were ignored.

Ratio of occupational intakes

Equation (10) expresses the relationship between the ratio of the occupational intake of ^{228}Ra to ^{226}Ra activity, $(M/R)_0$, for a worker and the mean ratio of occupationally derived ^{228}Ra to ^{226}Ra activity in bone at the time of sampling, $(\overline{M/R})_s$:

$$(M/R)_0 = (\overline{M/R})_s e^{(\lambda_M - \lambda_R)t}, \quad (10)$$

where t is the time elapsed from the intake of Ra to the sampling of bone.

The standard error, $S_{(M/R)_0}$, of $(M/R)_0$ (Table 4) was computed as

$$S_{(M/R)_0} = (M/R)_0([S_{(\overline{M/R})_s}/(\overline{M/R})_s]^2 + S_4^2t^2)^{1/2}, \quad (11)$$

where S_4 is the standard error of λ_M.

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