ANL-6398
Biology and Medicine
(TID-4500, 16th Ed.,
Amended)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

RADIOLOGICAL PHYSICS DIVISION
SEMIANNUAL REPORT

January through June 1961

J. E. Rose, Division Director
L. D. Marinelli, Associate Division Director

September 1961

Preceding Report:
ANL-6297 July through December 1960

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
United States Atomic Energy Commission

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

	<u>Page</u>
A study of the mechanism of energy transfer from naphthalene to α -NPO Isadore B. Berlman	3
Detailed autoradiographic and microradiographic studies of bone from human radium cases R. E. Rowland	5
An evaluation of the incomplete gamma function over a range of biologically significant values R. E. Rowland and Constance J. Leuer	24
Microradiographic appearance of bones subject to chronic vascular insufficiency R. E. Rowland	36
Skeletal retention of the alkaline earth radioisotopes and bone dosimetry R. E. Rowland	40
<u>In vivo</u> measurement of Cs^{137} : summary of observations up to June 1961 Charles E. Miller and John B. Corcoran	41
Evaluation of crystal body configurations used to measure the gamma-ray activity of the human body Charles E. Miller	43
Retention of radium in humans from 21 to 29 years after intravenous administration Charles E. Miller and L. D. Marinelli	44
The measurement of total body potassium in man and its relation to gross body composition Alexander P. Remenchik and Charles E. Miller	45
Liquid scintillator solvent study for the purpose of improving the "twin" scintillation fast neutron detector. Part II Roger Grismore	46
Translocation of thorium daughters to bone L. D. Marinelli and Henry F. Lucas, Jr.	50

TABLE OF CONTENTS

	<u>Page</u>
Radioactivity in bones of patients injected with Thorotrast.	
II. Variations in distribution in the cortical, trabecular and marrow portions	
Dorothy E. Wallace, Henry F. Lucas Jr., Douglas P. Krause, and Frank H. Ilcewicz	51
Further observations on Th^{228} and Ra^{226} in bones from dial painters and medical patients	
Dorothy E. Wallace.	63
Th^{228} and Ra^{226} analyses of bones from Kerala, India	
Dorothy E. Wallace.	67
Further studies on natural radioelements in bovine bones and teeth	
Elvira R. Di Ferrante.	71
Stable lead in human bone	
Douglas P. Krause	77
Fission and natural radioactivity in ground level air	
Philip F. Gustafson, Michael A. Kerrigan and S. S. Brar . . .	81
Multiple regression equations relating meteorological variables with eight-hour average radon concentrations	
Harry Moses	86
The Argonne meteorological model towing tank; two-dimensional jets in a stratified fluid	
Paul Frenzen.	96
Analysis of soil- and air-temperature data by Fourier techniques	
James E. Carson	110
Publications	130
Papers to be published	132
Patents.	133
Staff Activities.	133

A STUDY OF THE MECHANISM OF ENERGY TRANSFER FROM NAPHTHALENE TO α -NPO

I. B. Berlman

Ensuing an earlier report⁽¹⁾ on the fluorescence properties of an organic solution containing a single solute, naphthalene, this study was directed specifically toward the determination of the nonradiative energy-transfer efficiency from naphthalene to a second solute, as the concentration of the latter was varied. Although more than one method has been reported⁽²⁾ for achieving the desired studies, this report deals only with the method of uv excitation. A coordinated experiment involving excitation by a pulsed beam of electrons is in progress, and results will be published later.

The solutions being investigated are xylene plus 10 g/liter of naphthalene plus several low values of 2-[1-naphthyl]-5-phenyloxazole, (α -NPO), in the concentration range of 0 to 1 g/liter. The equipment for exciting and recording of emission spectra, as well as the conditions imposed on the preparation and handling of the solutions and the technique of computing the energy-transfer efficiency from changes in the fluorescence yield of the transferring medium, has been described previously.⁽²⁾

The donor molecules (naphthalene) were excited directly by 2890 Å radiation from the monochromator, whereas the acceptor molecules (α -NPO) were excited through the mechanism of nonradiative energy transfer; emission spectrograms were recorded for several concentration values of α -NPO. From the measured hypochromic change in the fluorescence spectrum of naphthalene, the energy-transfer parameter, ϵ , as a function of the α -NPO concentration in g/liter, C , was computed.

The curve of C/ϵ versus C , as depicted in Figure 1, departs from linearity at low values of C . Since linearity is indicative of just one transferring level,^(2,3) the deviation from linearity, engendered by the value of ϵ becoming too large, may be related to the observation that naphthalene has more than one component in its emission decay curve when excited with pulsed electrons,⁽⁴⁾ and thus, naphthalene may have several transferring levels of different degrees of metastability. It is tacitly assumed that the same decay time components are produced under uv excitation.

Since transfer quenching may be inversely proportional to the lifetime of the excited state, the longer-lived states will be preferentially depleted. Considering the long lethargic lifetimes of the slower components (≥ 0.5 μ sec) total depletion is probably achieved at acceptor molecule

concentrations of ≤ 0.1 g/liter. At larger values of C , energy transfer from the shorter-lived state then takes place in a linear fashion. Further clarifying evidence is necessary before the validity of these assumptions is established.

Figure 1

The ratio of α -NPO concentration (C) to the energy-transfer parameter (ϵ) is plotted against C .

References

1. I. B. Berlman. A study of transient dimers in a solution containing naphthalene. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1960. ANL-6297, pp. 3-7.
2. I. B. Berlman. J. Chem. Phys. 33 1124 (1960).
3. S. G. Cohen and A. Weinreb. Proc. Phys. Soc. (London) B69 593 (1956).
4. I. B. Berlman and A. Weinreb. Molecular Physics, to be published.

DETAILED AUTORADIOGRAPHIC AND MICRORADIOGRAPHIC STUDIES OF BONE FROM HUMAN RADIUM CASES

R. E. Rowland

Introduction

The observations made in this Laboratory on bone samples from a number of radium cases have been summarized in general terms in several publications. The first of these described the microradiographic appearance of these bones,⁽¹⁾ and included observations made on twelve cases. This was followed by a general description of the microscopic radium concentrations found in these cases,⁽²⁾ from which the local dose rate to bone was deduced. Subsequently the number of cases described in this fashion was increased to 19.⁽³⁾

While summaries of the observations on a group of cases are of interest, for in this manner the similarities between the cases are emphasized, much more information is routinely developed about each case than can be included in such a summary. In an effort to show the nature of the information that is developed from each sample studied, this report describes in detail the observations on five new cases.

This report covers: 1) another Elgin State Hospital patient; 2) a dial painter who died of a carcinoma of the mastoid; 3) an amputation sample obtained in 1934 from a dial painter who is still living; 4) and 5) two early radium cases (dial painters) from which bone samples were obtained from museum preparations. One of the latter samples had been stored dry for over 35 years and was found to be in excellent condition for our studies. In contrast, both the amputation specimen and the other museum sample had been stored for nearly 30 years in formalin, and considerable leaching and redeposition of the bone minerals, including radium, was found to have occurred. Consequently the autoradiographic studies of such samples were of little value. It does appear, however, that some information about the total body radium content can be determined by analysis of the radium-to-calcium ratio present in the formalin. For these two cases the body content estimated by extrapolation of this ratio to the entire body (~1000 g calcium) gave results which are in remarkably good agreement with those obtained by other methods.

Case Studies

Case 03-119.

Previous references:	Elgin State Hospital Case S, References 4, 5, 6 and 7
Date of birth:	March 7, 1880
Radium:	70 μ c Ra ²²⁶ given as 7 weekly 10 μ c I.V. injections during 1931, age 51 ^(4,5)
Physical data:	109 lbs, 5'1" tall on 1-20-1931, when 50 years old
Date of death:	April 14, 1960 (at 80 years of age)
Body burden:	0.54 μ c in 1951; ⁽⁵⁾ 0.62 μ c in 1953. ⁽⁵⁾

A portion of the femur, removed at autopsy, was available for study. Microradiographs, quantitative autoradiographs, and strip film autoradiographs have been made of a series of bone sections cut from this material.

The microradiographs of the portion of the cortical femur examined indicate little evidence of radium damage. There are a few plugged canals, and the bone is somewhat osteoporotic but, considering the patient's age, this is not surprising. On the whole, the bone looks good.

The strip film autoradiographs show that most of the hotspots are in unresorbed remnants of osteons. A few complete osteons containing radium are still present, and in each of these the alpha tracks are in a band about 50 μ wide, indicative that the radium was given over a relatively short period of time.

The quantitative alpha-track counts have yielded some rather interesting results. The specific activity in the maximum hotspots was found to be 14.2 μ μ c Ra²²⁶/mg of bone. This value is in excellent agreement with the corresponding values that have been found in other Elgin State Hospital cases, indicating that the uniform mode of administration of the radium has resulted in uniform labeling of the bone that was being formed at the time of administration. For comparison, the maximum hotspot values found in the Elgin State Hospital patients⁽³⁾ studied to date are listed in Table 1.

It is apparent that the hotspot-to-diffuse ratio in these cases decreases as the number of injections of radium was increased. This is to be expected, for the diffuse level should increase with the number of injections, whereas the hotspot intensity should be independent of this number, as long as the injections are of equal magnitude and equally spaced in time.

Table 1

Case	Number of injections of Ra ²²⁶	μc Ra ²²⁶ retained	Maximum hotspot, $\mu\mu\text{c}/\text{mg}$	Average diffuse, $\mu\mu\text{c}/\text{mg}$	Calculated uniform label, $\mu\mu\text{c}/\text{mg}$	Ratio: hotspot to diffuse	Ratio: diffuse to uniform label
03117 (Q)	45	1.2	12	0.080	0.17	150	0.47
03118 (R)	41	3.6	16	0.15	0.66	107	0.23
03109 (I)	18	0.8	13	0.060	0.15	218	0.40
03119 (S)	7	0.6	14	0.054	0.12	263	0.45

From the portion of the cortical femur available for study it was found that few radium-labeled osteons remained intact. Apparently many osteons were labeled when radium was administered, but only portions of them remained at death. Thus it is concluded that, at least in this piece of bone, considerable bone turnover has taken place during the thirty-year interval between administration and death.

Case 03-998.

Previous references: Case 1, in Reference 8; Reference 9;
Case 9 of Table I in Reference 10

Radium: Patient worked as a dial painter for
6 years, from October 1917 until 1923⁽⁸⁾

Physical data: 140 lbs, 5'5" tall (shortly before death?)⁽⁸⁾

Date of death: June 18, 1925,⁽¹⁰⁾ at age 35⁽⁸⁾ or 33⁽¹⁰⁾

Body burden: 180 μg of radioactive substances, about
70% being from mesothorium and its
decay products.⁽⁹⁾

A museum jar had been acquired which contained bone specimens from an early dial painter. The following notations were pasted on the outside of the jar:

"1925. First reported Death Due to Radium Poisoning. Thrombocytopenic Macrocytic Anemia with Red Embryonal Marrow. No Sarcoma. Dial painter for six years, licked brushes. Died two years after quitting dial painting. Age 35.

"Autopsy: Severe anemia with purpura, petechiae, buccal sepsis, early jaw necrosis and red bone marrow. Equivalent of 90 micrograms of radium in body, found during life and after death by electroscopic methods."

Within the jar there was no liquid. Instead, dry, was a mandible, cut into two pieces so that it would fit into the jar, and also three small bottles. These items are similar to the items shown in Figure 1 of Reference 8. The small jars contained the following: a portion of a femur, dry; bone ash; and spleen ash. Ten teeth were also found, eight still loosely attached to the mandible.

Studies have been completed on the materials found in this jar to determine the following:

1. The activity in the teeth
2. The total Ra^{226} present in the mandible
3. The Ra^{228} and Ra^{226} present in the bone ash
4. The autoradiographic distribution in bone sections from the femur
5. The microradiograph appearance of the bone sections from the femur.

1. The Activity in the Teeth. Each of the ten teeth was sealed, individually, within a glass vial, and the total gamma-ray spectrum of each recorded immediately and again a month later. From these two measurements the total Ra^{226} content and the radon retention of each tooth was determined. These results are listed in Table 2.

Table 2

Ra^{226} content of teeth from Case 03-998

Tooth	Weight, g	Total activity, μmc	$\frac{\mu\text{mc}}{\text{g}}$	Radon retention, %
Right canine	1.022	859 ± 8	840	76.3
Left canine	1.013	1318 ± 12	1300	71.3
Right lateral incisor	0.477	570 ± 7	1190	75.1
Left lateral incisor	0.317	595 ± 6	1870	59.1
Right first premolar	0.799	659 ± 8	830	69.6
Left first premolar	0.715	519 ± 7	730	54.4
Right second premolar	0.398	485 ± 4	1220	61.6
Left second premolar	0.932	711 ± 8	760	66.0
Right second molar	1.939	1623 ± 11	840	68.4
Left second molar	1.970	1614 ± 12	820	59.8
Total	9.582	8953	930	

The differences in weight between pairs of teeth are due to the fact that some of these teeth were broken; note in particular the low weight of the right second premolar. This may account for some of the difference

in activity per gram of tooth that will be noted. On the other hand, the significant difference in activity per gram between the two canine teeth is real, for both were intact.

These data indicate that differences as great as a factor of two in the radium content per gram of tooth may be expected between teeth of a single individual.

If the body burden is estimated from the average content of the 10 teeth, namely $930 \mu\mu\text{c Ra}^{226}$ per gram of tooth (by assuming first, that teeth are 28% by weight calcium, and second, that the body contains 1000 g Ca) a value of $3.3 \mu\text{c Ra}^{226}$ is obtained.

The radon retention of these teeth is seen to vary between 54 and 76%. These values are in agreement with a value of 75% obtained from a single fresh tooth,⁽¹¹⁾ but somewhat larger than most of the determinations on teeth made by Miller and Corcoran.⁽¹²⁾ Evidently sizeable variations in the radon retention of teeth are to be expected.

2. The Ra^{226} Content of the Mandible. The mandible was sealed in a glass vial and two gamma-ray spectra obtained, the first immediately after sealing and the second a month later. The total radium content was calculated to be $1.4 \times 10^{-7} \text{c}$; since the total weight was 43.0 g, the specific activity of these two pieces is $3300 \mu\mu\text{c/g bone}$. Assuming a total skeletal mass of 6000 g, this measurement yields a total content of $20 \mu\text{c Ra}^{226}$.

The radon retention of the mandible was found to be 58%. This value appears to be relatively high, considering the fact that only eight years elapsed between the first contact with radium and death. However, it has been observed that when bone is stored dry, the retention of radon increases, and thus the observed retention after storage bears no relation to the retention that existed in vivo at time of death.

3. The Ra^{228} and Ra^{226} Content of the Bone Ash. Through the cooperation of H. F. Lucas and D. E. Wallace of this Division, the Ra^{228} and Ra^{226} content of the bone ash (found in one of the small bottles in the museum jar) has been measured. The results are tabulated below:

On 12/21/60 $\text{Ra}^{226} = 4800 \pm 90 \mu\mu\text{c/g ash}$

On 1/15/61 $\text{Th}^{228} = 73 \pm 6 \mu\mu\text{c/g ash.}$

Employing as a half-life of Ra^{228} the value 5.8 years,⁽¹⁴⁾ the Ra^{228} content of the bone at the time of measurement is obtained by the transient equilibrium relation,

$$\text{Th}^{228} = \text{Ra}^{228} \times \frac{T_{1/2} \text{ Ra}^{228}}{T_{1/2} \text{ Ra}^{228} - T_{1/2} \text{ Th}^{228}} = \text{Ra}^{228} \times 1.48;$$

so on 1/15/61 $\text{Ra}^{228} = 49 \pm 4 \mu\mu\text{c/g ash}$.

Extending these measurements back to time of death, 6/18/25, we obtain

$4800 \pm 90 \mu\mu\text{c/g ash}$ Ra^{226}

$3300 \pm 270 \mu\mu\text{c/g ash}$ Ra^{228} .

Assuming that the total skeleton contains 2600 g ash, this yields the following total content, at death:

$12 \mu\text{c}$ Ra^{226}

$9 \mu\text{c}$ Ra^{228} .

At the termination of her employment, sometime during 1923, the Ra^{228} content would have been nearly equal in magnitude to the Ra^{226} content.

4. Autoradiographic Analysis. From the piece of femur found in one of the three small bottles within the museum jar, bone sections were cut and subsequently autoradiographed and microradiographed. The alpha-track counts from autoradiographs made on NTA plates were evaluated for the two conventional radium distributions in bone, that is, the diffuse distribution and the hotspots. In addition, these bone sections exhibited an unusual periosteal proliferation, over which track counts were also made.

The number of tracks counted per unit area was converted to Ra^{226} concentration by means of the analysis described in Reference 2. However, the radon retention in the embedded and sectioned bone was assumed to be 60%, on the basis of the measured retention of radon in the mandible. Since the alphas emitted from the daughters of Ra^{228} account for only 2% of the total recorded in the emulsion, this contribution was ignored in the analysis.

The results obtained were:

Hotspots, maximum:	$21.2 \mu\mu\text{c Ra}^{226}/\text{mg bone}$
Average diffuse:	$0.46 \mu\mu\text{c Ra}^{226}/\text{mg bone}$
Periosteal proliferation:	$2.25 \mu\mu\text{c Ra}^{226}/\text{mg bone}$
Hotspot-to-diffuse ratio:	46

It has been observed^(2,3) that the magnitude of the diffuse distribution is, on the average, one half of the hypothetical uniform distribution, i.e., the body content divided by the bone mass. This relationship provides a unique method of determining the body content from an analysis of a diffuse distribution, with an uncertainty of about a factor of two. Applied to this case, employing again a value of 6000 grams of bone, we obtain a total body content of $5.5 \mu\text{c Ra}^{226}$.

The hotspot concentration can be used to estimate the rate of intake of radium. Recalling that the Elgin State Hospital patients, who had received $10 \mu\text{c Ra}^{226}$ per week, contained hotspot concentrations ranging from 12 to $16 \mu\mu\text{c/mg}$ bone, it would appear that about $15 \mu\text{c}$ per week would have been required to produce the concentrations found in this case. However, the Elgin cases received radium intravenously, whereas this was a case of ingestion, a route in which considerable discrimination against radium exists. Therefore it appears that at least 20 to $30 \mu\text{c Ra}^{226}$ per week would have to have been ingested to produce these concentrations.

5. Microradiographic Analysis. The microradiographs of the sections cut from the femur show a rather interesting phenomenon: a periosteal proliferation (Figure 2a). This growth has taken place outward from the original periosteum, and consists of a less ordered bone structure.

The autoradiograph of this section (Figure 2b) shows that this proliferation is markedly radioactive, much more so than the diffuse reaction visible in the original compact bone, but not as radioactive as some hotspots or as the original periosteal line.

These observations suggest the following sequence of events:

- 1) The intense hotspots occurred as a consequence of a high radium intake during the early exposure to radium. The high uptake along the original subperiosteal bone indicates that lamellar bone was still being laid down in this region at this time, i.e., age 27.

- 2) The periosteal proliferation took place sometime after the early exposure, but still during the exposure to radium. At this time there was less ingestion of radium, for the specific activity of this new bone is considerably less than in the intense hotspots found in the original cortical bone. Whether this proliferation took place as a consequence of the radiation dose, or as a consequence of some other stimulation cannot be determined from these observations.

- 3) At subsequent times increased intake of radium occurred, for a few more intense deposits of radium are visible autoradiographically in this new proliferation. (see Figure 2b).

Figure 2

A microradiograph (a) and an autoradiograph (b) of a 100- μ thick femur section from case 03-998. Magnification 7X. A band of disorganized bone which contains considerable radium, has been laid down over the original periosteal surface.

This sequence of events has been subsequently supported by the microradiographs of the sections cut from a second small piece of bone from this patient. The outstanding feature here is the crack which is evident in the series of microradiographs shown in Figure 3. While at first glance this appears to be simply a crack made in cutting the section, closer inspection reveals that it actually was a break that occurred in this femur during the lifetime of the patient. Indeed, careful study of the microradiographs and autoradiographs indicates that it could only have occurred during the patient's employment as a dial painter.

Figure 3

A series of microradiographs of 100- μ thick sections cut from a femur block from case 03-998. The full extent of a microfracture is shown in (a). The subsequent microradiographs show the new bone formation within the crack, all of which contains radium, even the hypercalcified areas. Sections a and b are separated by 1400 μ ; b and c by 400 μ ; c and d by 900 μ ; d and e by 2400 μ . The total separation between a and e is 5400 μ .

The following observations serve to substantiate this observation:

- 1) The break extends only to the original periosteum, not through the new proliferation; this implies that the break occurred before the proliferation of new bone started.
- 2) The series of illustrations in Figure 3 shows that new bone formation has occurred within the break, and that this formation starts at the periosteal proliferation.
- 3) The new bone within the break is highly radioactive, indicating that it formed while radium was being ingested.
- 4) Several of the osteons split by the break were entirely labeled with radium. This finding indicates that the break did not occur until after the initial exposure to radium.
- 5) In one location a hypercalcified osteon was split by this break. Although such hypercalcifications are typical signs of radium in bone, no information is yet available to indicate how long radium must be present before such changes take place. The fact that such a system was present before the break occurred would suggest, however, that the fracture probably occurred in the latter half of the 5-year exposure period.

The typical bone changes visible microradiographically in bone in which radium has been present 20 or 30 years, such as the hypercalcified plugs, hypercalcified osteons, large regions of resorption, etc.,⁽¹⁾ are all visible in this bone, which represents the situation eight years after the start of a large intake of this isotope. Previously, the hypercalcifications found in bone from radium cases never contained significant quantities of radium, indicating that they were formed sometime after the exposure to radium. This is not the case here; many of these plugs and other hypercalcifications are highly radioactive. Indeed, within the break illustrated in Figure 3c is a hypercalcified region which is highly radioactive. It may be surmised that this hypercalcification formed after a portion of the "repair" bone within the break has been resorbed. This implies that, with high internal doses, these bone changes can take place within a relatively short time in human bone.

Case 03-999.

Previous references: Case 6 (K.S.) in Reference 10; Case 6 (K.S.) in Reference 13; Case 3 in Reference 8; KS in Reference 3

Date of birth: Unknown

Radium: Worked as a dial painter for $4\frac{1}{2}$ years during a $5\frac{1}{4}$ -year period⁽¹⁰⁾

Physical data: At age 26 (1925) 113 lbs, 63 inches in height⁽⁸⁾

Date of death: February 18, 1933, at age 31, eleven years after employment as a dial painter⁽¹³⁾

Body burden: Equivalent of 18 μ g of radium.⁽¹³⁾

A museum jar has been acquired which contained two pieces of femur and portions of an osteogenic sarcoma from an early dial painter. The following notation was on the outside of the jar:

"Femur with Sarcoma - Original Radium Dial Painter. Case No. 6. Dial painter for 5 years, licked brushes. Died in 1933, 11 years after quitting dial painting. Autopsy - Large osteogenic sarcoma of lower end of left femur with extensive necrosis and suppuration; sepsis; no regional or visceral metastases; radiation osteitis over skeleton.

"Equivalent of 19 micrograms of radium in body. CME 13, 082."

From the description of this case given in the reference it appears that this patient painted watch dials between 1917 and 1921, when she was 15 to 19 years old.

Since the contents of this jar were in formalin, the liquid was removed, and the interior surface of the jar was cleaned with acid. Analysis of these, by H. F. Lucas and D. E. Wallace, indicated that both radium and calcium were present in the formalin, but that little radium had plated out on the surface of the jar. The total amount in the formalin was:

Ra²²⁶ $3.5 \pm 0.2 \times 10^{-9}$ c

Calcium 0.40 ± 0.02 g

Ratio 8.8×10^{-9} c Ra²²⁶/g Ca

Assuming that both radium and calcium were leached from bone at the same rate, and that there are 1000 g Ca in the body, the Ra²²⁶ content of the formalin would yield a total body content of 8.8 μ c.

Radiochemical analysis of a 5-g (wet) sample of femur gave the following values (again by Lucas and Wallace):

on 4/22/59

Ra ²²⁶	2920 ± 60 μμc/g ash
Th ²²⁸	5.53 ± 0.3 μμc/g ash

therefore

Ra ²²⁸	3.74 ± 0.2 μμc/g ash
-------------------	----------------------

Extending these measurements back to time of death, 2/18/1933, yields:

Ra ²²⁶	2920 ± 60 μμc/g ash
Ra ²²⁸	81 ± 4 μμc/g ash

Assuming that the total skeleton contains 2600 g ash, this gives the following content at time of death:

Ra ²²⁶	7.6 μc
Ra ²²⁸	0.21 μc

At termination of painting, sometime during 1922, the ratio of Ra²²⁸ to Ra²²⁶ would have been approximately 0.1.

The pieces of femur from the museum jar contained intact marrow. Since there were some trabeculations present in the marrow cavity, the first attempts at embedding the bone were made with an effort to save these fragile structures. To do this, and at the same time to remove completely the contents of the marrow cavity, a $\frac{1}{4}$ -inch thick cross section was first soaked in acetone, then gently boiled in a dilute solution of ethylene diamine for a few minutes. Normally such treatment will remove the soft tissues from the surface of the bone without visibly disturbing the matrix of the bone. In this case, however, the sections fractured excessively during the sectioning process.

Subsequently other slabs of bone were cleaned and embedded without resorting to the ethylene diamine treatment. These sectioned satisfactorily, and their microradiographs showed the cause of previous difficulties. Decalcification had taken place, particularly where the bone surface was bare (see Figure 4), undoubtedly the consequence of long storage in formalin. Deposition of mineral along the outermost bone surface has also occurred, as is indicated by the hypercalcification shown in the microradiograph. An actual decrease in mineralization can be seen only along those portions of the bone that were in intimate contact with the formalin; apparently the contents of the marrow cavity prevented this gross demineralization from taking place along the endosteal surfaces. The

autoradiographs made from these sections (Figure 5) show a remarkable uniform diffuse distribution, which indicates that there has been a redistribution of radium throughout all of the bone.

Figure 4

A microradiograph of a 100- μ thick section of femur from case 03-999. Magnification 34X. Decalcification is evident under the periosteal surface, and a hypermineralized line is present at the surface, the consequence of formalin decalcification and redeposition.

Figure 5

An autoradiograph of a complete femur cross section from case 03-999. Magnification 3.2X. The very uniform diffuse distribution and the line of more intense activity around the outer edge of the bone illustrate the effect of formalin leaching.

In addition, the microradiographs show the typical hypercalcifications of radium bone, plugged canals, hypercalcified osteons, etc., as well as widespread regions in which there appear to be no lacunae. This filling of lacunae with mineral has been observed previously in radium-damaged bone, but in this case might have occurred as a consequence of the formalin leaching.

Evidence for a redistribution of radium throughout the bone is also found in the data obtained by the quantitative autoradiographic studies. The measured quantities are:

Maximum hotspots	8.9 $\mu\mu\text{c Ra}^{226}/\text{mg bone}$
Average diffuse	1.4 $\mu\mu\text{c Ra}^{226}/\text{mg bone}$
Hotspot-to-diffuse ratio	6.4
Predicted body burden	1.4 $\mu\mu\text{c}/\text{mg} \times 2 \times 5.1 \text{ kg bone}/\text{total body}$ = 14 $\mu\text{c Ra}^{226}$.

This total content, predicted on the basis that the diffuse is one half the hypothetical uniform label, is considerably greater than the values obtained by radiochemical analysis. The hotspot-to-diffuse ratio of 6.4 is a factor of ten lower than is usually observed in bones from dial painters, and thus, also indicates that significant redistribution of the radium has taken place in this bone.

Case 03-407.

Previous references:	None
Date of birth:	October 28, 1905
Radium:	Painted watch dials from 1923 to 1945
Physical data:	143 lbs, 5'3" tall on 9/18/57
Date of death:	February 2, 1961
Body burden:	1.45 $\mu\text{c Ra}^{226}$.(14)

This case is of considerable interest, for this patient had a carcinoma of the mastoid and specimens from both the mastoid and femur were available for study and comparison. A basic question is thus presented: Is there any difference between the radium distributions in these two regions that would indicate why the mastoid should be particularly vulnerable to neoplasms⁽¹⁵⁾ after long exposure to radium?

The femur sections presented a typical radium distribution. A diffuse level of 0.16 $\mu\mu\text{c Ra}^{226}/\text{mg bone}$ was measured, with a maximum hotspot level of 5.0 $\mu\mu\text{c Ra}^{226}/\text{mg bone}$. The hypothetical uniform label is

calculated to be $0.22 \mu\mu\text{c}/\text{mg}$, from the weight of 65 kg and the total body content of $1.45 \mu\text{c}$. The hotspot-to-diffuse ratio is 31, and the ratio of the diffuse to the uniform label is 0.73.

These ratios are within the expected range for exposure as a dial painter. The hotspot-to-diffuse ratio is somewhat low, undoubtedly because of the very long period of exposure. Similarly, the long exposure to radium probably accounts for the relatively high diffuse level.

The microradiographs of the femur sections show a typical radium-damaged bone. Many hypercalcifications are present, such as plugged canals and hypercalcified osteons, and much evidence of active resorption is visible.

The findings from the mastoid are not greatly different: The diffuse level is lower, while the hotspots are somewhat more intense. A diffuse level of $0.08 \mu\mu\text{c}/\text{mg}$ was measured in a small sample removed at an operation about two years before death, and a value of $0.06 \mu\mu\text{c}/\text{mg}$ was measured in an autopsy sample from the left mastoid. These values are only 32% of the uniform label. Since this is very porous bone, the track counts probably give a low value for the diffuse distribution, for the tracks counted originate from a smaller area than that actually surveyed. The hotspot levels are higher than those found in the femur, the most intense level measured being $5.9 \mu\mu\text{c}/\text{mg}$. The greatest difference between the two areas studied was that there were many more hotspots in the mastoid than in the cortex of the femur. Complete bands of buried radium were present, undoubtedly corresponding to regions of bone growth that took place at the time the radium was acquired. In this respect, although the exposure to radium took place over a 22-year period, it is likely that most of the mastoid activity was acquired during the first few years, when pointing the brush with the lips was practiced.

The microradiographs of the mastoid sections show this to be very porous bone; the small, irregular spaces in this bone are the mastoid air cells. In addition to this natural porosity, there is evidence of extensive resorption. The latter observation is of interest, for if this bone had been subject to accelerated turnover for any extensive time interval there would be little radium-containing bone left.

Even with the natural porosity of the mastoid bone and the large number of radium hotspots, the sections studied show few regions where the radium alpha particles could irradiate the soft tissues lying on the bone surfaces. The apparently recent resorption has brought to the surface some of the buried radium deposits, but this may represent a consequence of the presence of the neoplasm rather than its cause. Thus this study cannot give any clues as to apparently high radiosensitivity of this region.

Case 03-455.

Previous references:	Code 309 in References 6 and 16
Date of birth:	January 27, 1906
Radium:	Dial painter for one year at age 16(6)
Physical data:	119 $\frac{1}{2}$ lbs., 5'3 $\frac{1}{2}$ " tall on 8/21/58
Date of sample:	Amputation of left arm, 1934
Body burden:	0.52 μc Ra ²²⁶ in 1952;(6) 0.22 μc RaB + RaC and 0.36 μc radon in 1958.(14,17)

The amputated arm had been preserved in a formalin-filled museum jar from 1935 until 1961. A longitudinal incision had been made through a portion of the arm to show the sarcoma near the elbow. A two-inch-long piece of this radius and the formalin from the jar were obtained for analysis.

It is evident from the microradiographs (Figure 6) that leaching of the minerals has taken place and that some redeposition has occurred along the outermost edge of the bone surface. In regions where the bone surface was covered, as in the medullary cavity, this redeposition is least evident. Along the cut surface of the bone it should be noted that the line of greatest density is somewhat inside of the outermost edge, as is shown in the higher power magnification of the microradiograph (Figure 7).

Figure 6

A microradiograph of a 100- μ thick section cut from the radius of case 03-445. Magnification 8X. Decalcification is evident near the periosteal surface, with a hypermineralized line along the surface.

Figure 7

An enlarged portion of Figure 6 to illustrate the position of the hypermineralized line inside of the bone surface. Magnification 60X.

The quantitative autoradiographs show a diffuse concentration of $0.20 \mu\mu\text{c Ra}^{226}/\text{mg}$ bone, but there were not any recognizable "true" hotspots in any of the bone sections. This bone had been in formalin for 26 years, and, considering the results from the previous formalin-stored bone, 03-999, this diffuse level cannot be considered to be representative of the actual concentration that existed in 1934. If we assume that the radium distribution has been altered to the extent that it is now uniformly distributed throughout the bone mineral, the total body content would be

$$0.20 \mu\mu\text{c}/\text{mg} \times 5.5 \text{ kg} = 1.1 \mu\text{c Ra}^{226}.$$

This value, for the content in 1934, may be compared with the later measured values by means of the retention equation⁽⁵⁾

$$R_t = 0.54 t^{-0.52}.$$

Date	Measured body content, μc	Predicted body content, μc
1934	-	1.1
1952	0.52	0.68
1958	0.58	0.62

It is evident that a value of $1.1 \mu\text{c}$ in 1934 is compatible with the measured values in 1952 and 1958, particularly when we recall that one arm was missing when these latter measurements were made.

The radium and calcium content of the formalin were determined⁽¹⁷⁾ to be:

Ra ²²⁶	0.62 $\mu\mu\text{c}/\text{ml}$
Ca	0.46 mg/ml

or 1.36 $\mu\mu\text{c}$ Ra²²⁶/mg Ca. On the basis of 1000 grams of calcium in the body, if this ratio in the formalin were the same as the average in the bone, a total content of 1.36 μc Ra²²⁶ is obtained. This result is in good agreement with the values obtained both by autoradiography and from the present measurements.

It thus appears that a measurement of the radium-to-calcium ratio of the formalin in which radium bones have been stored can be used to estimate the magnitude of the total body content of the subject. Thus, even though the leaching action of the formalin has irrevocably altered the original radium distribution, an estimate of the body content can be made which is well within a factor of two of the actual value.

References

1. R. E. Rowland, J. H. Marshall, and J. Jowsey. Radium in human bone: The microradiographic appearance. *Radiation Research* 10 323-334 (1959).
2. R. E. Rowland and J. H. Marshall. Radium in human bone: The dose in microscopic volumes of bone. *Radiation Research* 11 299-313 (1959).
3. R. E. Rowland. Late observations of the distribution of radium in the human skeleton. *Radioisotopes in the biosphere*, Ed. R. S. Caldecott and L. S. Snyder. Minneapolis: University of Minnesota, 1960.
4. H. Schlundt, J. T. Nerancy, and J. P. Morris. The detection and estimation of radium in living persons. IV. The retention of soluble radium salts administered intravenously. *Am. J. Roentgenol.* 30 515-522 (1933).
5. W. P. Norris, T. W. Speckman, and P. F. Gustafson. *Am. J. Roentgenol.* 73 785-802 (1955).
6. W. B. Looney, R. J. Hasterlik, A. M. Brues, and E. Skirmont. A clinical investigation of the chronic effects of radium salts administered therapeutically (1915-1931). *Am. J. Roentgenol.* 73 1006-1037 (1955).
7. C. E. Miller. Retention of radium in humans from 21 to 29 years after intravenous administration. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1959. ANL-6104, pp. 85-86.

8. H. S. Martland, P. Conlon, and J. Knef. Some unrecognized dangers in the use and handling of radioactive substances. *J. Am. Med. Assoc.* 85 1769 (1925).
9. H. S. Martland. Occupational poisoning in manufacture of luminous watch dials. *J. Am. Med. Assoc.* 92 466-552 (1929).
10. H. S. Martland. The occurrence of malignancy in radioactive persons. *Am. J. Cancer* 15 2435-2516 (1931).
11. R. E. Rowland. The radium distribution in the tooth of a dial painter. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1959. ANL-6104, pp. 30-33.
12. C. E. Miller and J. B. Corcoran. Estimation of radium body content of exposed humans from the activity of extracted teeth. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June 1960. ANL-6199, pp. 68-70.
13. H. S. Martland. Occupational Tumors: (6) Bones. In International Labor Office, Geneva, Occupation and Health. Supplement. October, 1939, Brochure 13, pp. 1-23.
14. C. E. Miller. Unpublished measurements.
15. R. J. Hasterlik. Radiation neoplasia. *Proc. Inst. Med. Chicago* 23 (2) (March 15, 1960).
16. R. J. Hasterlik. The delayed toxicity of radium deposited in the skeleton of human beings. Proc. Intern. Conf. Peaceful Uses Atomic Energy, Geneva, 1955. New York: United Nations, 1956. Vol. 11, pp. 149-155.
17. H. F. Lucas. Personal communication.

AN EVALUATION OF THE INCOMPLETE GAMMA FUNCTION OVER A RANGE OF BIOLOGICALLY SIGNIFICANT VALUES

R. E. Rowland and Constance J. Leuer

In many problems of radiobiology an integral of the form

$$\int e^{-\lambda t} t^{-b} dt$$

appears; for example, λ might be the decay constant of a bone-deposited radioisotope, t the time the isotope had been carried, and the exponent b a pure number which describes the retention of the isotope in bone in the form of the power function. This is similar in form to the gamma function, which is defined as

$$\Gamma(p+1) = \int_0^{\infty} e^{-x} x^p dx \quad .$$

Our equation can be transformed into this form by a change of variable, namely by letting

$$x = \lambda t$$

so that $t = x/\lambda$

and $dt = \frac{1}{\lambda} dx \quad .$

When the limits of integration are 0 and ∞ , this integral may be evaluated by tables of the complete gamma function, several of which are listed in References 1, 2, and 3. These tables give the value of the function for values of p between 1 and 2. To obtain function values for values of p outside of this range the following recursion formula is used to extend the tables:

$$\Gamma(p+1) = p \Gamma(p) \quad .$$

When the limits of integration are between 0 and a finite value of t , we have the incomplete gamma function, which is defined as

$$\Gamma_x(p+1) = \int_0^x e^{-x} x^p dx \quad .$$

This function may be evaluated by means of Pearson's "Tables of the Incomplete Γ -Function".⁽⁴⁾ These tables, however, do not give actual values

of this function, but rather tabulate a function $I(x, p)$, which is defined as

$$I(x, p) = \Gamma_x(p+1) / \Gamma(p+1) \quad .$$

From this function and a table of the complete gamma function the desired values of the incomplete gamma function can be obtained.

In practice, however, this evaluation from the tables is a laborious task. Pearson's tables are in terms, not of x , but rather of $u = x/\sqrt{p+1}$, in order to reduce the range of this variable. In addition, the tabulated values of u are distressingly far apart, particularly when p is negative and u is close to zero, a range which is of interest to us. Fortunately, a linear interpolation is justified between values of u when an auxiliary function

$$I'(u, p) = I(u, p) / u^{p+1}$$

is used; Pearson's tables include a table of $\log I'(u, p)$, and it is from this table that values of $I(u, p)$ can be determined for small values of u .

It is evident that the evaluation of any particular incomplete gamma function is a time-consuming operation. Since for many of the problems of radiobiology great accuracy is not required, an approximate value of this function would be useful if it were readily available. With this in mind we have evaluated this function for a range of variables appropriate to many biological problems. Although the function is expressed to four-figure accuracy at each of the points at which it was calculated, we visualize that interpolation between the calculated points will be required for most of the specific integrals that one might wish to evaluate.

In Table 3 is a listing of the three variables, t , λ , and b , that are considered. For each combination of the three variables the integral was evaluated; the tabulated values follow in Tables 4 through 11.*

*The following example will serve to demonstrate the use of the tables: Evaluate the integral

$$\int_0^t e^{-\lambda t} t^{-b} dt$$

over the period from 0 to 1000 days when the half-life of a deposited isotope is 30 years and the exponent describing the retention is -0.2. Table 5, for $b = 0.2$, gives a value for the integral of 305.4 at $t = 1000$ days and $T_{1/2} = 30$ years.

In many problems the power law description of the data over the first day is not valid. In these cases a correction must be made to evaluate the integral over this period. It should be noted that the inapplicable portion of the integral can be removed by subtraction. For example, considering the above example, if the data are not valid over the first day, we obtain the desired integral as follows:

$$\int_1^{1000} e^{-\lambda t} t^{-b} dt = \int_0^{1000} e^{-\lambda t} t^{-b} dt - \int_0^1 e^{-\lambda t} t^{-b} dt \quad .$$

so

$$\int_1^{1000} e^{-\lambda t} t^{-b} dt = 305.4 - 1.250 = 304.2 \quad .$$

In these tables, one for each value of the exponent b , only the value of the integral is listed. For those who may wish the actual value of the incomplete gamma function, note that it may be obtained as the product of the integral and λ^{p+1} , for

$$\int_0^t e^{-\lambda t} t^{-b} dt = \frac{1}{\lambda^{p+1}} \cdot \Gamma_x(p+1) \quad .$$

Table 3

Values of the three variables t , λ , and b for which the incomplete gamma function is evaluated

t (time in days)	λ , where $\lambda = \frac{0.693}{T_{1/2}}$		b (exponent)
	(days ⁻¹)	$T_{1/2}$ (half-life)	
1	6.929×10^{-1}	1 day	-0.1
2	2.310×10^{-1}	3 days	-0.2
5	9.900×10^{-2}	7 days	-0.3
10	4.620×10^{-2}	15 days	-0.4
20	1.980×10^{-2}	35 days	-0.5
50	9.240×10^{-3}	75 days	-0.6
100	4.620×10^{-3}	150 days	-0.7
200	1.899×10^{-3}	1 year	-0.8
500	9.493×10^{-4}	2 years	
1,000	3.797×10^{-4}	5 years	
2,000	1.899×10^{-4}	10 years	
5,000	6.329×10^{-5}	30 years	
10,000			

Table 4

Value of the integral when the exponent $-b = -0.1$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	0.8159	0.9984	1.061	1.087	1.101	1.105	1.109	1.110	1.111	1.111	1.111	1.111
2	1.165	1.680	1.891	1.983	2.035	2.056	2.062	2.070	2.071	2.073	2.073	2.073
5	1.449	2.893	3.776	4.250	4.515	4.628	4.679	4.709	4.719	4.725	4.727	4.728
10	1.485	3.665	5.752	7.149	8.049	8.442	8.637	8.747	8.786	8.812	8.817	8.822
20	1.486	3.964	7.568	11.02	13.72	15.11	15.75	16.18	16.32	16.41	16.44	16.46
50	1.486	3.996	8.518	15.60	24.48	30.43	33.76	35.93	36.74	37.23	37.40	37.51
100	1.486	3.996	8.565	16.88	32.21	46.90	56.79	64.17	67.05	68.88	69.48	69.89
200	1.486	3.996	8.565	17.01	35.90	62.72	87.51	110.0	119.7	126.2	128.5	130.0
500	1.486	3.996	8.565	17.01	36.46	71.84	123.9	197.7	240.6	273.2	285.4	294.0
1,000	1.486	3.996	8.565	17.01	36.46	72.39	134.0	262.7	369.0	468.1	509.7	540.5
2,000	1.486	3.996	8.565	17.01	36.46	72.40	135.1	295.4	490.1	743.2	873.5	979.3
5,000	1.486	3.996	8.565	17.01	36.46	72.40	135.1	300.8	557.4	1118.0	1571.0	2043.0
10,000	1.486	3.996	8.565	17.01	36.46	72.40	135.1	300.8	561.2	1257.0	2086.0	3330.0

Table 5

Value of the integral when the exponent $-b = -0.2$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	0.9372	1.131	1.197	1.225	1.239	1.245	1.247	1.249	1.250	1.250	1.250	1.250
2	1.275	1.787	1.997	2.090	2.139	2.159	2.167	2.173	2.175	2.176	2.176	2.176
5	1.530	2.869	3.668	4.098	4.337	4.439	4.484	4.511	4.521	4.527	4.528	4.530
10	1.560	3.505	5.297	6.476	7.236	7.572	7.728	7.821	7.855	7.875	7.881	7.884
20	1.561	3.737	6.691	9.442	11.57	12.67	13.18	13.50	13.62	13.69	13.71	13.72
50	1.561	3.760	7.374	12.70	19.20	23.47	25.86	27.41	27.99	28.35	28.46	28.54
100	1.561	3.760	7.405	13.54	24.25	34.20	40.86	45.81	47.73	48.95	49.35	49.62
200	1.561	3.760	7.405	13.62	26.51	43.88	59.58	73.51	79.77	83.80	85.21	86.16
500	1.561	3.760	7.405	13.62	26.83	49.08	80.14	122.9	147.3	166.0	173.0	177.9
1,000	1.561	3.760	7.405	13.62	26.83	49.37	85.45	156.7	213.9	266.5	289.1	305.4
2,000	1.561	3.760	7.405	13.62	26.83	49.37	85.96	172.6	272.8	399.6	463.9	517.2
5,000	1.561	3.760	7.405	13.62	26.83	49.37	85.96	175.1	303.3	567.8	775.2	989.5
10,000	1.561	3.760	7.405	13.62	26.83	49.37	85.96	175.1	304.9	625.6	988.5	1521.0

Table 6

Value of the integral when the exponent $-b = -0.3$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	1.096	1.302	1.369	1.402	1.417	1.423	1.426	1.427	1.428	1.428	1.428	1.428
2	1.422	1.934	2.143	2.235	2.284	2.303	2.312	2.317	2.319	2.320	2.320	2.231
5	1.652	2.899	3.632	4.017	4.223	4.325	4.366	4.390	4.399	4.404	4.405	4.407
10	1.677	3.424	4.967	5.966	6.610	6.895	7.026	7.104	7.132	7.148	7.153	7.157
20	1.678	3.604	6.040	8.249	9.932	10.80	11.20	11.45	11.54	11.59	11.61	11.63
50	1.678	3.621	6.531	10.56	15.32	18.41	20.13	21.25	21.67	21.92	22.00	22.06
100	1.678	3.621	6.552	11.12	18.63	25.45	29.90	33.24	34.52	35.33	35.60	35.79
200	1.678	3.621	6.552	11.17	20.03	31.35	41.34	50.06	54.00	56.51	57.39	58.00
500	1.678	3.621	6.552	11.17	20.21	34.31	52.95	77.83	91.83	102.5	106.5	109.3
1,000	1.678	3.621	6.552	11.17	20.21	34.46	55.73	95.41	126.4	154.4	166.6	175.2
2,000	1.678	3.621	6.552	11.17	20.21	34.46	55.98	103.2	155.0	219.1	250.9	277.6
5,000	1.678	3.621	6.552	11.17	20.21	34.46	55.98	104.3	168.9	294.4	390.1	488.1
10,000	1.678	3.621	6.552	11.17	20.21	34.46	55.98	104.3	169.5	318.4	478.2	706.1

Table 7

Value of the integral when the exponent $-b = -0.4$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	1.312	1.532	1.607	1.638	1.654	1.661	1.664	1.665	1.666	1.666	1.666	1.666
2	1.627	2.144	2.350	2.441	2.489	2.509	2.517	2.523	2.524	2.525	2.526	2.526
5	1.834	3.003	3.671	4.024	4.220	4.303	4.340	4.362	4.369	4.374	4.376	4.377
10	1.855	3.436	4.772	5.631	6.171	6.412	6.522	6.588	6.611	6.626	6.630	6.633
20	1.856	3.575	5.598	7.377	8.723	9.399	9.718	9.916	9.984	10.03	10.04	10.05
50	1.856	3.587	5.590	9.026	12.53	14.79	16.02	16.83	17.12	17.30	17.37	17.41
100	1.856	3.587	5.964	9.391	14.70	19.37	22.42	24.64	25.50	26.05	26.23	26.35
200	1.856	3.587	5.964	9.423	15.56	22.98	29.37	34.92	37.34	38.92	39.48	39.85
500	1.856	3.587	5.964	9.423	15.67	24.67	35.93	50.50	58.61	64.72	66.98	68.57
1,000	1.856	3.587	5.964	9.423	15.67	24.75	37.39	59.64	76.53	91.72	98.09	102.7
2,000	1.856	3.587	5.964	9.423	15.67	24.75	37.51	63.44	90.39	122.9	139.0	152.1
5,000	1.856	3.587	5.964	9.423	15.67	24.75	37.51	63.96	96.64	156.7	201.0	246.2
10,000	1.856	3.587	5.964	9.423	15.67	24.75	37.51	63.96	96.93	166.6	237.4	335.8

Table 8

Value of the integral when the exponent $-b = -0.5$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	1.620	1.851	1.936	1.970	1.987	1.994	1.997	1.999	1.999	2.000	2.000	2.000
2	1.925	2.445	2.654	2.744	2.792	2.811	2.820	2.825	2.827	2.828	2.828	2.828
5	2.111	3.213	3.832	4.139	4.329	4.404	4.438	4.458	4.465	4.469	4.471	4.471
10	2.129	3.571	4.735	5.467	5.931	6.135	6.229	6.285	6.305	6.317	6.320	6.323
20	2.129	3.679	5.371	6.811	7.881	8.422	8.677	8.833	8.888	8.922	8.933	8.940
50	2.129	3.688	5.624	7.985	10.59	12.22	13.09	13.71	13.92	14.05	14.10	14.13
100	2.129	3.688	5.633	8.227	12.01	15.23	17.29	18.80	19.39	19.75	19.87	19.96
200	2.129	3.688	5.633	8.246	12.54	17.43	21.54	25.04	26.59	27.58	27.93	28.16
500	2.129	3.688	5.633	8.246	12.60	18.40	25.25	33.83	38.52	42.05	43.35	44.25
1,000	2.129	3.688	5.633	8.246	12.60	18.44	26.02	38.59	47.84	56.00	59.46	61.93
2,000	2.129	3.688	5.633	8.246	12.60	18.44	26.08	40.44	54.57	71.13	79.19	85.82
5,000	2.129	3.688	5.633	8.246	12.60	18.44	26.08	40.68	57.41	86.29	107.0	127.6
10,000	2.129	3.688	5.633	8.246	12.60	18.44	26.08	40.68	57.53	90.43	122.0	164.7

Table 9

Value of the integral when the exponent $-b = -0.6$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	2.091	2.340	2.431	2.467	2.486	2.493	2.496	2.499	2.499	2.500	2.500	2.500
2	2.386	2.914	3.118	3.214	3.262	3.282	3.290	3.295	3.297	3.298	3.298	3.298
5	2.554	3.600	4.171	4.454	4.628	4.697	4.728	4.746	4.752	4.757	4.758	4.758
10	2.568	3.896	4.916	5.548	5.935	6.118	6.198	6.246	6.263	6.273	6.276	6.278
20	2.569	3.980	5.406	6.578	7.442	7.871	8.073	8.197	8.241	8.268	8.277	8.282
50	2.569	3.986	5.588	7.417	9.358	10.56	11.19	11.64	11.79	11.89	11.92	11.94
100	2.569	3.986	5.594	7.576	10.29	12.52	13.94	14.96	15.36	15.60	15.69	15.74
200	2.569	3.986	5.594	7.588	10.61	13.87	16.52	18.78	19.74	20.37	20.59	20.74
500	2.569	3.986	5.594	7.588	10.65	14.42	18.63	23.71	26.44	28.49	29.24	29.76
1,000	2.569	3.986	5.594	7.588	10.65	14.44	19.03	26.19	31.29	35.76	37.59	38.91
2,000	2.569	3.986	5.594	7.588	10.65	14.44	19.06	27.10	34.56	43.10	47.18	50.46
5,000	2.569	3.986	5.594	7.588	10.65	14.44	19.06	27.20	35.85	49.86	59.57	69.19
10,000	2.569	3.986	5.594	7.588	10.65	14.44	19.06	27.20	35.89	51.58	65.79	84.41

Table 10

Value of integral when the exponent $-b = -0.7$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	2.889	3.163	3.259	3.298	3.318	3.326	3.330	3.332	3.333	3.333	3.333	3.333
2	3.176	3.716	3.918	4.019	4.067	4.086	4.095	4.100	4.102	4.103	4.103	4.104
5	3.327	4.330	4.862	5.127	5.282	5.346	5.374	5.391	5.396	5.400	5.401	5.402
10	3.339	4.574	5.475	6.023	6.349	6.513	6.581	6.622	6.637	6.646	6.648	6.650
20	3.339	4.639	5.852	6.815	7.514	7.839	8.019	8.118	8.152	8.175	8.181	8.186
50	3.339	4.643	5.983	7.412	8.873	9.761	10.23	10.55	10.66	10.73	10.76	10.77
100	3.339	4.643	5.987	7.518	9.484	11.04	12.02	12.69	12.99	13.16	13.21	13.25
200	3.339	4.643	5.987	7.525	9.682	11.87	13.60	15.04	15.62	16.06	16.20	16.29
500	3.339	4.643	5.987	7.525	9.703	12.18	14.79	17.82	19.43	20.56	21.05	21.35
1,000	3.339	4.643	5.987	7.525	9.703	12.20	15.00	19.11	21.94	24.38	25.32	26.10
2,000	3.339	4.643	5.987	7.525	9.703	12.20	15.01	19.55	23.53	27.92	30.02	31.68
5,000	3.339	4.643	5.987	7.525	9.703	12.20	15.01	19.61	24.12	30.98	35.56	40.01
10,000	3.339	4.643	5.987	7.525	9.703	12.20	15.01	19.61	24.14	31.69	38.14	46.30

Table 11

Value of the integral when the exponent $-b = -0.8$

t, days	Half-life ($T_{1/2}$)											
	1 day	3 days	7 days	15 days	35 days	75 days	150 days	1 year	2 years	5 years	10 years	30 years
1	4.516	4.817	4.920	4.962	4.984	4.993	4.996	4.998	4.999	5.000	5.000	5.000
2	4.793	5.351	5.558	5.657	5.706	5.726	5.735	5.740	5.742	5.743	5.743	5.743
5	4.930	5.899	6.398	6.646	6.788	6.846	6.872	6.888	6.893	6.896	6.898	6.898
10	4.940	6.100	6.903	7.382	7.669	7.806	7.864	7.899	7.912	7.919	7.922	7.924
20	4.940	6.151	7.193	7.991	8.560	8.831	8.966	9.046	9.074	9.092	9.097	9.101
50	4.940	6.154	7.288	8.417	9.524	10.19	10.53	10.77	10.85	10.90	10.92	10.93
100	4.940	6.154	7.290	8.486	9.525	11.02	11.70	12.17	12.37	12.48	12.52	12.55
200	4.940	6.154	7.290	8.491	10.05	11.53	12.66	13.60	13.98	14.25	14.34	14.40
500	4.940	6.154	7.290	8.491	10.06	11.71	13.34	15.17	16.11	16.79	17.06	17.24
1,000	4.940	6.154	7.290	8.491	10.06	11.72	13.45	15.84	17.42	18.76	19.29	19.70
2,000	4.940	6.154	7.290	8.491	10.06	11.72	13.46	16.05	18.19	20.48	21.55	22.40
5,000	4.940	6.154	7.290	8.491	10.06	11.72	13.46	16.08	18.46	21.85	24.04	26.13
10,000	4.940	6.154	7.290	8.491	10.06	11.72	13.46	16.08	18.47	22.15	25.10	28.72

References

1. B. O. Peirce. A Short Table of Integrals. Boston: Ginn and Co., 1929.
2. H. B. Dwight. Tables of Integrals and Other Mathematical Data. New York: Macmillan and Co., 1947.
3. E. S. Pearson and H. O. Hartley. Biometricka Tables for Statisticians, Vol. I. London: Cambridge University Press, 1954.
4. K. Pearson. Tables of the Incomplete Γ - Function. London: His Majesty's Stationery Office, 1922; also London: Cambridge University Press, 1946.

MICRORADIOGRAPHIC APPEARANCE OF BONES SUBJECT TO CHRONIC VASCULAR INSUFFICIENCY

R. E. Rowland

In a paper published in 1934,⁽¹⁾ Jaffe and Pomeranz described the histological appearance of bone sections from eight subjects who had extremities amputated as a consequence of arteriovascular disease. They reported that widespread aseptic necrosis was present, and that revascularization of the dead bone was occurring. Some of the haversian canals, particularly near the periosteal surface of the cortex, contained pink-staining plugs. Roentgenograms of the bones revealed extensive changes: "there were areas of rarefaction disseminated throughout and exhibited in round or somewhat elongated shadows, everywhere less dense than normal."

This description could be applied remarkably well to the bones of many of the radium cases. The reference to plugged canals is of particular interest, for this symptom is always present in the radium cases. While plugged canals are known to occur in apparently normal bone (indeed, Jaffe and Pomeranz mention this in regard to one of their control cases), it would be of considerable interest to determine what conditions, other than internal alpha irradiation, could lead to this particular phenomenon.

In view of the apparent similarities between radium bone and chronic avascular bone, an effort was made to obtain bone samples from amputations of extremities following atherosclerotic gangrene. Six samples have now been examined, and in each case a significant number of mineral changes have been observed. Each of the cases is briefly described below.

Case 284. Male, Age 75 Years.

The microradiographs show many plugged canals, enlarged lacunae, mineral-filled lacunae, many active regions of resorption, and hypercalcified osteons (see Figure 8). In brief, this case was indistinguishable, microradiographically, from a relatively high-level (1 to 2 μc) radium case.⁽²⁾ A few traces of tetracycline were found, which indicates that bone remodeling was still taking place.

Interpretation of these observations is complicated by the fact that 27 years before the amputation this man had received Thorotrast. Thus both Th^{228} (0.6 $\mu\mu\text{c/g}$ ash) and Ra^{228} (0.4 $\mu\mu\text{c/g}$ ash) were present in bone,* so the bone changes might be the consequence of alpha irradiation, not vascular insufficiency. While one cannot be positive on this point, there are two reasons which indicate that it is the vascular disorder that is responsible for the bone changes. First, these measured activity levels

*Measurements made by H. F. Lucas of this Division.

are low compared to those found in radium cases with equivalent damage ($\sim 300 \mu\mu\text{c Ra}^{226}/\text{g ash}$). Second, other Thorotrast patients containing larger ThO_2 burdens but free of vascular disease do not show similar bone changes.

Figure 8

A microradiograph of a 100-micron thick bone section from the femur of Case 284. Several plugged haversian canals are visible, and many canals with hypercalcified borders or deposits. Magnification 42.5X.

Case 340. Male, Age 59 Years.

A few plugged canals are present, but essentially the bone appears normal. However, a most unusual finding is shown in Figure 9a. There is a calcified deposit in the wall of the nutrient artery as it enters the upper tibia. A soft x-ray (3 kv) image of the identical area is shown in Figure 9b, which demonstrates that the calcification is in the medial coat of the artery. Thus it is apparent that the arterial vasculature of bone can be affected by atherosclerosis.

Case 285. Female, Age 72 Years.

A significant number of plugged canals and many areas in which the lacunae are filled with mineral are found in the microradiographs.

Case 341. Male, Age 85 Years.

Very few haversian canals are plugged. However, there are many osteons in which all or a portion of the cross-sectional area is seen to be hypermineralized. Tetracycline is present, so bone turnover has been occurring within recent years.

a. A 10-kv radiograph showing the hypercalification in the artery.

b. A 3-kv radiograph which indicates that the calcification is taking place in the tunica media, or middle layer of the arterial wall.

Figure 9

A pair of microradiographs of the nutrient artery as it enters the upper tibia; Case 340. Magnification 40 X.

Case 343. Male, Age 44 Years.

Many plugged canals and hypercalcified osteons are present. Bone turnover is indicated by the presence of tetracycline.

Case 120. Male, Age 69 Years.

A few plugged canals are present, accompanied by many hypercalcified regions. A significant amount of resorption appears to be taking place, particularly under the periosteal surface.

Considered as a group, the most striking feature of these six cases is the amount of hypercalcification present in all of them. Variable numbers of plugged canals and mineral-filled lacunae were found, but always many hypercalcified "resting lines," sclerotic edges around the haversian canals, and hypercalcified osteons were present.

A correlation may exist between the number of such bone changes and the severity or duration of the vascular insufficiency, but since this latter information is not available, it can only be concluded that radium-like changes are present in the bone of individuals suffering from atherosclerotic gangrene. This conclusion then lends support to the hypothesis that much of the damage to bone from deposited radium is actually vascular damage.

References

1. H. L. Jaffe and M. M. Pomeranz. Changes in the bones of extremities amputated because of arteriovascular disease. Arch. Surg. 29 566-588 (1934).
2. R. E. Rowland, J. H. Marshall and J. Jowsey. Radium in human bone: The microradiographic appearance. Radiation Research 10 323-334 (1959).

SKELETAL RETENTION OF THE ALKALINE EARTH RADIOISOTOPES AND BONE DOSIMETRY*

R. E. Rowland

Abstract

It is suggested that, following the deposition of any of the alkaline earth radioisotopes in bone, the dose to a bone tissue volume cannot be determined from a terminal observation of the distribution of the isotope unless the mechanisms which remove these isotopes from bone are known. On the basis of a study which demonstrated the magnitude of removal by exchange processes, it is suggested that all of the microscopic concentrations of the isotope in bone continually lose activity, and that this loss may be described by a power function of the form $q_t = q_0 t^b$. The concept of accumulated dose is discussed, and equations for the calculation of accumulated dose are derived on the basis of this hypothesis.

*Presented at Univ. Utah, College of Medicine,
Symposium on Some Aspects of Internal Irradiation,
Heber, Utah, May 8-11, 1961. Pergamon Press
(to be published)

IN VIVO MEASUREMENT OF Cs^{137} : SUMMARY OF OBSERVATIONS UP TO JUNE 1961

Charles E. Miller and John B. Corcoran

The amount of Cs^{137} present in a group of control subjects has been measured intermittently at our Laboratory since it was first detected in humans in 1955.(1)

A group of about 14 employees unexposed to radioactive materials was selected as representative residents of the Chicago area and measured every three or four months during 1956 through 1959 and every six months during 1960 and 1961. Unfortunately, complete data were not obtained on all subjects over this six-year period because some of them have left the Laboratory or have been away from it for a period of time.

The data on eight control subjects for the dates on which all were measured (except for RER on 1-20-58) are given in Table 12. These data cover the time from 4-56 through 6-61. The data on eight subjects from early 1955 until 1958 are plotted in Figure 10. The Cs contents are expressed as $\mu\mu\text{c Cs}^{137}$ per gram of body K to provide a direct comparison between subjects, regardless of their body size.

The curves of Figure 10 show that the Cs content increased quite rapidly from March of 1955 to April of 1956 and then remained rather constant until 1957. The averages of the values given in the table demonstrate that the Cs^{137} content increased more or less uniformly from then on until it reached a maximum of $65 \mu\mu\text{c Cs}^{137}/\text{g K}$ during June of 1959. The Cs concentration has decreased ever since that date, and is now lower than at any time since April of 1956.

The last three values of 5/24/60, 12/15/60 and 6/24/61 fall on a straight line on a semilog plot and yield a half-time of 17 months. This half-time in the human during the later half of 1960 and the first half of 1961 is consistent with the decrease in average atmospheric concentration observed since the fall of 1958 by Gustafson.(2)

Table 12
Cesium-137 potassium-40 ratios in eight control subjects over a span of 5 years

Subject	4/31/56	6/20/57	10/30/57	1/20/58	4/18/58	7/31/58	11/20/58	3/19/59	6/17/59	9/14/59	12/8/59	5/24/60	12/15/60	6/24/61
C. L.	42	36	37	41	51	57	49	59	69	60	56	54	38	26
I. S.	24	29	48	33	43	52	52	59	55	55	47	49	45	25
H. M.	45	41	50	51	65	71	69	75	83	68	70	65	56	40
C. M.	39	41	39	39	44	43	58	76	81	64	62	51	34	32
B. O.	31	31	39	44	42	47	48	55	57	46	42	45	30	31
B. P.	20	28	29	33	35	44	46	54	59	55	54	48	32	25
R. R.	21	25	32	-	31	39	40	47	46	46	45	38	28	23
O. S.	30	37	34	44	43	48	51	62	66	64	63	47	37	30
Average	31	33	38	41	44	50	52	61	65	57	55	50	37	29

Figure 10. Cs^{137} trend in eight humans over a four-year period.

A delay should be expected in the appearance of a maximum in the curve tracing of the Cs^{137} human contents compared to that of atmospheric concentration (fallout rate) because there are several mean lives involved in the pauses that the Cs^{137} atom experiences in its journey from deposition on the surface of the soil to the uptake by man. Without much greater effort, however, it is impossible to delineate with any precision the components of the holdup, especially the delay present in the food chain, including processing and merchandising.

References

1. C. E. Miller and L. D. Marinelli. The Gamma-Ray Activity of Contemporary Man. *Science* 124 122-123 (July 20, 1956).
2. P. F. Gustafson, M. A. Kerrigan and S. S. Brar. This report, page 81. .

EVALUATION OF CRYSTAL BODY CONFIGURATIONS USED TO MEASURE THE GAMMA-RAY ACTIVITY OF THE HUMAN BODY*

Charles E. Miller

This study was undertaken to ascertain the sensitivity of various geometries in human body counters and to evaluate the uncertainties arising in each case from the use of an average calibration factor in people of the same height and weight, but differing in other anatomical characteristics.

The following measuring techniques were used with a single crystal: a) the 175-cm arc technique, b) the "Tilting Chair" technique,⁽¹⁾ c) the crystal at the small of the back,⁽²⁾ and d) a crystal located under the chair of a seated person. In addition, various multiple-crystal arrays were evaluated by placing, in turn, a single crystal at 7 equally spaced locations 10" apart along the body, at each of two heights above and two distances below the supine subject.

The gamma-ray spectra of a group of 20 patients containing known quantities of K^{42} and a group of 12 patients containing Ra^{226} were obtained. The counting rate from each subject, corrected for excretion and decay in the case of K^{42} , was compared to the average for the group to determine the magnitude of the variations that existed between subjects of different body builds. To evaluate further the calibration problems involved in each array the subject's spectra were compared to the spectra obtained with known amounts of the same isotope in various phantoms.

Although with a multiple crystal array greater sensitivities are possible than with the tilting chair, the uncertainty involved is about twice that obtained with the tilting chair or the 175-cm arc. This uncertainty can be eliminated by administering to each person a known tracer dose; however, this may not always be feasible and should be avoided since it increases considerably the technical difficulties.

References

1. C. E. Miller. Low-Intensity Spectrometry of the Gamma Radiation Emitted by Human Beings. Proc. Second Intern. Conf. Peaceful Uses of Atomic Energy, Geneva, September 1958. Geneva: United Nations, 1958. Vol. 23, pp. 113-122.
2. L. D. Marinelli, C. E. Miller, P. F. Gustafson and R. E. Rowland. The Quantitative Determination of Gamma-Ray Emitting Elements in Living Persons. Am. J. Roentgenol. 73 661-671 (1955).

*Paper presented at the International Atomic Energy Agency Symposium on Whole Body Counting, Vienna, Austria, June 1961.

RETENTION OF RADIUM IN HUMANS FROM 21 TO 29 YEARS AFTER INTRAVENOUS ADMINISTRATION*

Charles E. Miller and L. D. Marinelli

The retention of radium in humans from 21 years to 29 years after intravenous injection has been determined by measuring the radium body content of a group of mental patients who received from 70 to 410 μc at the rate of 10 μc per week in 1931.⁽¹⁾ Their body contents were measured in 1931, in 1951-1953,^(2,3) and 1959-1960⁽⁴⁾ with whole-body gamma-ray measuring techniques that ranged from the use of an insensitive electro-scope in ordinary rooms, to a suitably calibrated and shielded, low-level crystal spectrometer.

Within experimental error, the data obtained show that the retention of intravenously injected radium between the 21st and the 29th year can be described by the power law enunciated by Norris, *et al.*⁽³⁾ with the exponent calculated for the first 21 years after administration.

References

1. H. Schlundt, *et al.* The Detection and Estimation of Radium in Living Persons. *Am. J. Roentgenol.* 30 515-522 (1953).
2. L. D. Marinelli, C. E. Miller, P. F. Gustafson and R. E. Rowland. The Quantitative Determination of Gamma-Ray Emitting Elements in Living Persons, *Am. J. Roentgenol.* 73 661-671 (1955).
3. W. P. Norris, T. W. Speckman and P. F. Gustafson. Studies of the Metabolism of Radium in Man. *Am. J. Roentgenol.* 73 785-802 (1955).
4. C. E. Miller. Retention of Radium in Humans from 21 to 29 Years After Intravenous Administration. Argonne National Laboratory, Radiological Physics Division Semiannual Report, July through December, 1959. ANL-6104, pp. 85-86.

*Paper presented at the International Atomic Energy Agency Symposium on Whole Body Counting, Vienna, Austria, June 1961.

THE MEASUREMENT OF TOTAL BODY POTASSIUM IN MAN AND ITS RELATION TO GROSS BODY COMPOSITION*

Alexander P. Remenchik** and Charles E. Miller

This study was designed to delineate the degree of precision of the estimation of total body potassium in man and to relate the estimation to the lean body mass of man. Total body potassium was estimated by the measurement of K^{40} activity in the whole body scintillation counter and by the measurement of total exchangeable potassium (K_e) using K^{42} . Lean body mass was calculated from the measurement of tritiated water space (TBW). The subjects were clinically well ambulatory hospital patients. The mean difference in 5 subjects between measurements of K_e , one week apart, is $2.4 \pm 0.7\%$. The mean difference between measurements of TBW in 6 subjects, done on the same day, is $1.3 \pm 0.3\%$. The mean K_e/K^{40} ratio is $89 \pm 1.3\%$ in 15 subjects. The lean body mass content of potassium is 2.12 ± 0.06 g/kg in 12 subjects (from K_e) and is 2.39 ± 0.04 g/kg (from K^{40}) in 14 subjects. The gross body content of potassium is 1.69 ± 0.05 g/kg (from K_e) in 20 subjects and is 2.05 ± 0.07 g/kg (from K^{40}) in 18 subjects. The mean body fat content is $17.5 \pm 2.3\%$ in 17 subjects. These data suggest that all of the potassium in man is not exchangeable. These estimates of the lean body mass content of potassium are higher than the values reported by others⁽¹⁾ but agree with their data expressed in terms of gross body weight. Our estimates of body fat agree with values reported by other investigators.⁽²⁾

References

1. T. H. Allen, E. C. Anderson and W. H. Laugham. Total Body Potassium and Gross Body Composition in Relation to Age. Submitted for publication.
2. A. Keys and J. Brozek. Body Fat in Adult Man. *Physiol. Revs.* 33 245-325 (1953).

*Paper presented at the International Atomic Energy Agency Symposium on Whole Body Counting, Vienna, Austria, June 1961.

**Clinical Investigator, Veterans Administration Hospital, Hines, Illinois. This investigation was supported (in part) by a grant from the Psychiatric Training and Research Fund of the Illinois Department of Public Welfare.

LIQUID SCINTILLATOR SOLVENT STUDY FOR THE PURPOSE OF IMPROVING THE "TWIN" SCINTILLATION FAST NEUTRON DETECTOR. PART II

Roger Grismore

The program of measurement of the relative light yields of various liquid scintillator solutions for the purpose of improving the "twin" scintillation fast neutron detector ^(1,2) has been described in previous reports. ^(3,4) Results are presented in this paper for solutions of 4 g/liter diphenyloxazole in various additional organic solvents, most of which have relatively high proton-electron ratios; all of the solutions were nitrogen-bubbled to prevent oxygen quenching of the scintillations.

The results of the first set of measurements in the current series are shown in Table 13. These measurements were made with a Co⁶⁰ gamma-ray source in exactly the manner described in the last report ⁽³⁾ with the exception that pulse heights were measured to the points of inflection in the Compton edges rather than to the centers of the Compton peaks. This was necessary because the Compton peaks were not well defined for these solutions. The ambiguity arising in the determination of the location of the point of inflection accounts for the large probable errors attached to the pulse heights shown in Table 13. These pulse heights and those following have been normalized to a value of 163 for paraxylene to permit direct comparison with the results of the previous report. ⁽³⁾

Table 13

Relative pulse heights from solutions of 4 g/liter diphenyloxazole in various organic solvents under Co⁶⁰ gamma irradiation

Solvent	Grade	Chemical formula	Pulse height at Compton edge, arbitrary units*
2,5-Difluorotoluene	High purity commercial	C ₇ H ₆ F ₂	66 ± 6
Mineral oil	Commercial	C ₁₂ H ₂₆	44 ± 4

*Pulse heights have been normalized to a value of 163 for paraxylene.

When it was found that most of the solutions in the current study were not giving well-defined Compton peaks with the Co⁶⁰ source it was decided to switch to a Cs¹³⁷ internal conversion electron source for greater precision. A cylindrical Teflon scintillation cell with inside diameter

$1\frac{3}{16}$ in. and inside length $1\frac{7}{32}$ in. made according to the design described by Berlman and Marinelli⁽⁵⁾ was used with this source. A volume of 7 cc of the solution to be measured was used in the cell in each case, as in the previous series of measurements. This meant that during each run the liquid lay on the quartz light-exit window in a layer approximately 1 cm thick. The Cs^{137} source was fixed to the end of a $\frac{1}{4}$ -in. diameter stainless steel rod and was mounted inside the cell facing the quartz window at a distance of 5 mm from it. This distance is a little over a millimeter greater than the maximum range of the 0.63-Mev internal conversion electrons from Cs^{137} in any of the solutions tested.

The results of the set of measurements made with the Cs^{137} source are shown in Table 14. They were made in the same manner as those of Table 13 except for the changes in cell and source. Pulse height was measured to the center of the internal conversion electron peak in each case, the peak being sharp and well resolved for all the solutions tried except diphenyloxazole in mono-isopropylbiphenyl. However, even with the latter solution it was possible to make a fairly accurate determination of the peak center. One innovation in the handling of the solutions prior to the measurements which was used for all the solvents of Table 14 (except for mono-isopropylbiphenyl) was that the bubbling with nitrogen in the dry box was carried out before the solvent was measured out and mixed with diphenyloxazole, rather than after. This is particularly important in the case of the very volatile solvents, such as hexane and heptane, to prevent an increase in solute concentration through evaporation of the solvent. All of the operations incident to filling the cell subsequent to the bubbling were carried out in the dry box, of course.

Table 14

Relative pulse heights from solutions of 4 g/liter diphenyloxazole in various organic solvents under Cs^{137} internal conversion electron irradiation

Solvent	Grade	Chemical formula	Pulse height at electron peak, arbitrary units*
Mono-isopropylbiphenyl	Doubly-distilled	$\text{C}_{15}\text{H}_{16}$	157 ± 7
Heptane	High purity commercial	$\text{CH}_3(\text{CH}_2)_5\text{CH}_3$	76 ± 5
Hexane	High purity commercial	$\text{CH}_3(\text{CH}_2)_4\text{CH}_3$	70 ± 5
Decahydronaphthalene ("Decalin")	High purity commercial	$\text{C}_{10}\text{H}_{18}$	91.1 ± 1.3
Hexadecane	Practical	$\text{CH}_3(\text{CH}_2)_{14}\text{CH}_3$	68.2 ± 1.0

*Pulse heights have been normalized to a value of 163 for paraxylene.

The large percentage uncertainties of the determinations for hexane and heptane in Table 14 result from the fact that it was found to be very difficult to hold these solvents in the Teflon cell, and a small part of the 7 cc leaked out in each case before or during the measurements. The effect of having a lesser amount of solution in the cell was investigated with a diphenyloxazole in xylene solution, and it was found that an increase in pulse height resulted. The experience gained from this experiment is reflected in the probable errors attached to the hexane and heptane pulse heights. One further consideration which might slightly affect the results of the hexane and heptane measurements and that of mono-isopropylbiphenyl is that the geometry of the light collection process changes with a change in the maximum range of the electrons. In these solvents the range is somewhat different from that in paraxylene. The effect of a change in light collection geometry was investigated in the heptane solution by measuring the pulse height both with the source in its usual position 5 mm from the quartz window and with a source-window distance of ~ 3.5 mm (i.e., about equal to the maximum electron range). The result of moving the source closer to the window was an increase in pulse height. However, the magnitude of the increase was small enough so that it is felt that any variations in the results for hexane, heptane, and mono-isopropylbiphenyl in Table 14 due to geometry changes must be well within the probable errors quoted. Nevertheless, due to this effect and that of solution leakage, the results for hexane and heptane must be considered as maximum values.

From the results contained in this and the earlier reports^(3,4) it is seen that a combination of 1,4-difluorobenzene or 2,5-difluorotoluene and hexane, heptane, or hexadecane would be a decided improvement over the combination of fluorinated xylene and paraxylene presently used for the solvents in the "twin" scintillation fast neutron detector. The members of such a combination would have not only greater individual light yields but also a greater proton-electron ratio difference. In addition, they would have the advantage of having initially nearly equal light yields, hence avoiding any loss of resolution ensuing from severe quenching of one of the solutions to obtain a pulse height match.

References

1. I. B. Berlman and L. D. Marinelli. Rev. Sci. Instr. 27 858 (1956).
2. I. B. Berlman, R. Grismore, and B. G. Oltman. Rev. Sci. Instr. 31 1198 (1960).
3. R. Grismore and B. G. Oltman. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1960. ANL-6297, pp. 45-48.

4. L. D. Marinelli. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1960. ANL-6297, pp. 49-54.
5. I. B. Berlman and L. D. Marinelli. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1956. ANL-5679, pp. 8-9.

TRANSLOCATION OF THORIUM DAUGHTERS TO BONE*

L. D. Marinelli and Henry F. Lucas, Jr.

Abstract

An attempt has been made to correlate the Th^{232} - Ra^{228} and Th^{228} values found in skeletal specimens of patients injected with known amounts of Thorotrast with the radioactive equilibrium of the chain in the reticulo-endothelial system (R.E.S.).

The data for Ra^{228} are in accord with the predictions of Reynolds, *et al.*⁽¹⁾ for values of the retention parameter $0.2 < b < 0.5$. No excess of skeletal Th^{228} is found to justify the assumption that "washout" Th^{228} translocates from the R.E.S. to bone mineral.

No data for actual Ra^{224} in bone are available, but whole body and excreta activity measurements are consistent with the assumption that Ra^{224} is retained in the skeleton to a greater extent than Ra^{228} . Reliable direct measurements of this isotope are needed to establish with greater precision the chronic absorbed dose to the skeleton.

Reference

1. J. C. Reynolds, P. F. Gustafson and L. D. Marinelli. Retention and elimination of radium isotopes produced by the decay of thorium parents within the body - calculations and comparison with experimental findings. Argonne National Laboratory Report ANL-5689, (1957) pp. 1-43.

*Presented at Univ. Utah, College of Medicine, Symposium on Some Aspects of Internal Irradiation, Heber, Utah, May 8-11, 1961. Pergamon Press (to be published).

RADIOACTIVITY IN BONES OF PATIENTS INJECTED WITH THOROTRAST

II. VARIATIONS IN DISTRIBUTION IN THE CORTICAL, TRABECULAR AND MARROW PORTIONS

Dorothy E. Wallace, Henry F. Lucas, Douglas P. Krause
and Frank H. Ilcewicz

Thorotrast patients, code numbers 04-101* and 04-102* have been discussed in previous reports and publications.⁽¹⁻³⁾ Further unpublished data are presented herein.

A more complete picture of the distribution of Th^{232} and its daughters in bone fractions has been made possible by analyses for Ra^{228} , Th^{228} and Th^{232} in bones from a Thorotrast patient. These specimens were obtained through the courtesy of Dr. David C. Dahlin, Mayo Clinic, Rochester, Minn. (Code number 04-104,* our number 284B).

Routine analysis for Th^{228} on the first specimen of cortical femur received April 1960 showed a much greater content than is usually found in normal human bone. Investigation confirmed the probability that the source might have been a Thorotrast patient. Dr. Dahlin subsequently provided more samples, the tibia, fibula and the head of the femur.

Patient 284B had received, it was believed, 10 ml of Thorotrast (about 2 g or $0.22 \mu\text{c Th}^{232}$) by injection in the brachial artery on 8/26/33 at 49 years of age. His left leg was amputated on 2/15/60 when he was 76 years old and atherosclerotic.

A. Cortical Bone

1. Treatment of Samples

The first cortical femur sample (284B-1a) was cleaned by scraping, washing in ether to remove marrow, dried at 110°C and ashed at 620°C . The nitric acid (2M) solution of the ashed bone contained about 0.2 g/ml. Analyses for Ra^{228} , Th^{228} and Ra^{226} were made on this solution. The Ra^{226} was determined by the radon emanation technique of Lucas and Ilcewicz.^(4,5) Th^{228} and Th^{232} were removed from the solution by dibutyl phosphate (DBP) and the Th^{228} determined from the Ra^{224} which grew into the DBP from the Th^{228} ; this procedure has been reported previously by Stehney, et al.⁽⁶⁾

*Atomic Energy Commission, Punched Card Code Numbers.

Ac^{228} was extracted from the nitric acid solution with dibutyl phosphate, and Ra^{228} was determined as described by Krause.⁽⁷⁾ The RaD (Pb^{210}) method and the analysis for this specimen has been reported by Holtzman.⁽⁸⁾

A second portion of the first cortical femur sample (284B-6) was extracted (by soxhlet) with ethylenediamine (ED) before ashing, in February 1961 to assure the removal of all of the organic matter, by the Losee method.⁽⁹⁾ A marrow-free portion of cortical tibia (284B-2) was prepared by sawing out a longitudinal diaphyseal section outside the marrow.

Th^{232} was determined by α -pulse analyses with a Frisch grid chamber and 100 channel analyzer. The DBP extract, free of Ra^{224} , was plated on platinum. Corrections were made for background, Ra^{224} growth and low energy tails from Th^{228} and Th^{230} .

2. Analyses on Cortical Femur

Table 15 presents a summary of the analyses for Th^{228} , Ra^{228} and Th^{232} on cortical femur. The Th^{232} measurement was made on 284B-6 (inorganic, ED-extracted) because the dibutyl phosphate extraction had been made more recently, and the sample was marrow-free. The Th^{228} values for the two femur samples, both by α -pulse analyses, and the two DBP-extractions for 284B-1a and 284B-6, were in good agreement.

Table 15
Analysis on cortical femur bone for Th^{228} , Ra^{228} and Th^{232}

Sample number	284B-1a	284B-1a	284B-6 Inorganic	284B-6 Inorganic	284B-6-0 Organic	284B-6 and 284B-6-0
Nuclide	Th^{228}	Ra^{228}	Th^{232}	Th^{228}	Th^{228}	Th^{228}
Treatment of sample	Ether cleaned, dried 110°C, ashed 620°C, HNO_3 solution DBP-x	As before; HNO_3 solution	ED-X, ashed; HNO_3 solution DBP-x	As before; DBP-x	Ashed (ED) and H_2O extracts DBP-x	
Fresh weight, g	-	-	5.20	5.20	-	5.20
Ashed weight, g	4.44	4.44	3.046	3.046	0.1095	3.155
$\mu\mu\text{c} \pm 0.9\text{E}^\dagger$ in sample	2.64 ± 0.030	-	0.66 ± 0.10	1.80 ± 0.10	0.031 ± 0.007	1.83 ± 0.10
$\mu\mu\text{c} \pm 0.9\text{E}^\dagger$ per g ash	0.595 ± 0.007	0.45 ± 0.05	0.22 ± 0.033	0.590 ± 0.033	-	0.580 ± 0.032
Date of analysis	5/13/60	1/12/61	7 12 61	2/10/61	3/8/61	-
$\mu\mu\text{c}$ of nuclide per g ash on 2/15/60	0.600 ± 0.008	0.496 ± 0.050	0.22 ± 0.033	0.640 ± 0.04	-	0.625 ± 0.04
$\text{Th}^{228}/\text{Ra}^{228}$	1.21			1.30		1.26

Ra^{226} content of this sample was $0.034 \pm 0.004 \mu\mu\text{g/g}$ ash on 6/28/60.

Pb^{210} content of another portion, (284B-1b) 4.114 g ash, was $0.054 \pm 0.006 \mu\mu\text{c/g}$ ash on 6/10/60.

$\mu\mu\text{c}$ Ra^{228} for 284B-1a used for correction $\mu\mu\text{c}$ Th^{228} to 2/15/60.

[†] 90% confidence interval.

The Ra^{228} analysis on 284B-1a was made 245 days after the Th^{232} and Th^{228} were removed by DBP (5/13/60), and the $0.45 \mu\mu\text{c}$ Ra^{228} value obtained corrected for decay back to that date, was $0.49 \mu\mu\text{c/g}$ ash.

Corrections for the Ra^{228} and Th^{228} content of the bone at the date of amputation (2/15/60) were made on the basis of the Th^{232} found in the sample and the 90-day period between the amputation and thorium removal by DBP (see Appendix 1).

The second portion of the same cortical bone was separated into the inorganic fraction, 284B-6, and the organic fraction, 284B-6-0. The difference in time between the two DBP-extractions was 26 days, causing an insignificant difference in the Th^{228} measurement, from 0.031 to 0.032 $\mu\mu\text{c}$ for the total organic ash. The percent of the total Th^{228} activity in organic and inorganic portions found in the organic ash was 1.7%, while the organic ash was 3.5% of the total ashed weight. The distribution of Th^{228} between the organic and inorganic portions of bone has been reported previously by Wallace, Stehney and Ilcewicz,⁽¹⁰⁾ showing the same small percentage present in the organic phase when ED-extraction was used for the separation.

The Ra^{228} value for 284B-1a was used to calculate the Th^{228} present in the inorganic sample, 284B-6, at amputation. The Th^{228} was $0.640 \pm 0.04 \mu\mu\text{c/g}$ ash, while the Th^{228} value for the combined organic and inorganic fractions does not differ statistically, $0.625 \pm 0.04 \mu\mu\text{c/g}$ ash. The Th^{232} determination in the inorganic portion, 284B-6, on 7/12/61, showed that 0.22 $\mu\mu\text{c/g}$ ash was present, the same as that present originally. The ratio of $\text{Th}^{232}/\text{Ra}^{228}/\text{Th}^{228}$ at amputation was 1.0:2.3:2.7.

At amputation, then, the analyses on 284B-1a showed this relationship.

Nuclide	$\mu\mu\text{c/g}$ ash	$\mu\mu\text{c}$ in skeleton*	% of Th^{232} injected (0.22 μc)
Th^{232}	0.22	616	0.28
Ra^{228}	0.496	1400	0.63
Th^{228}	0.60	1680	0.76

* $\mu\mu\text{c/g}$ ash x 2,800 g skeletal ash.

3. Cortical Tibia. Marrow-free

The marrow-free portion of cortical tibia (284B-2), prepared by sawing a longitudinal diaphyseal section from outside of the marrow cavity, was muffled at 620°C without any treatment. The Th^{228} content at time of DBP separation was $0.46 \pm 0.033 \mu\mu\text{c/g}$ ash. The Ra^{228} determination on the nitric acid solution was made 297 days after DBP extraction. The corrected value to 9/23/60 is $0.435 \pm 0.040 \mu\mu\text{c/g}$ ash. No Th^{232} analysis was made, but when the Th^{232} content found in cortical femur (284B-6), 0.22 $\pm 0.033 \mu\mu\text{c/g}$ ash was used, the calculation of the Ra^{228} and Th^{228} content was made as in Appendix I. Table 16 summarizes these results.

Table 16

Th²²⁸ and Ra²²⁸ in cortical tibia, marrow-free

Sample No.	284B-2	284B-2
Nuclide	Th ²²⁸	Ra ²²⁸
Fresh weight, g	6.555	6.555
Ashed weight, g	3.940	3.940
$\mu\mu\text{c} \pm 0.9 \text{ E}$ in sample	1.81 ± 0.13	-
$\mu\mu\text{c} \pm 0.9 \text{ E}$ per g ash	0.46 ± 0.033	0.39 ± 0.04
Date of analysis	9/23/60	7/17/61
$\mu\mu\text{c}$ of nuclide on 2/15/60	0.46 ± 0.033	0.45 ± 0.040
Th ²²⁸ /Ra ²²⁸	1.02	

The ratio of Th²³²/Ra²²⁸/Th²²⁸ = 1.0:2.0:2.0.

In this cortical sample, the relationship between the three nuclides at amputation is shown below:

Nuclide	$\mu\mu\text{c/g}$ ash	$\mu\mu\text{c}$ in skeleton*	% of Th ²³² injected (0.22 μc)
Th ²³²	0.22	616	0.28
Ra ²²⁸	0.45	1260	0.57
Th ²²⁸	0.46	1288	0.58

* $\mu\mu\text{c/g}$ ash x 2,800 g skeletal ash

B. Marrow from Tibia

Cross sections of diaphyseal tibia were carefully cleaned of any adhering bone particles before the marrow was removed with a small curved dissecting scalpel. The marrow was gritty and obviously calcified; the white granular ash from this marrow also showed evidence of bone ash; it dissolved readily in nitric acid.

The sample, 284B-M was small; only 4.22 g of marrow could be obtained, and the ashed weight was 0.383 g. The total Th^{228} from the ash was $0.425 \pm 0.025 \mu\mu\text{c}$ at the time of DBP separation on 9/23/60.

An estimation of the probable amount of bone present in the fresh calcified marrow was made from the ashed weight. Both from our experience and from analyses reported by Hursh et al.,⁽¹¹⁾ it is known that the average ash from cortical bone is about 60%, while the ashed yield from marrow and soft tissues is about 1.0%. The method for estimation follows:

Fresh calcified marrow, 4.22 g = X (fresh bone) + Y (fresh marrow)

Ashed calcified marrow, 0.383 g = 0.60 X + 0.01 Y

Solving for X, we find that the fresh bone weight equals 0.577 g, and the ashed weight equals 0.346 g.

Using the $\mu\mu\text{c Th}^{228}/\text{g}$ ash in the marrow-free cortical tibia bone, 0.460 $\mu\mu\text{c}$, this bone ash would represent 0.159 $\mu\mu\text{c}$ of the total 0.425 $\mu\mu\text{c}$ obtained. The remainder, 0.226 $\mu\mu\text{c Th}^{228}$, would be from the marrow, free of bony particles. The estimation is not too far removed from the actual weight since total ash weight, 0.383 g less 0.346 g for bone ash, leaves 0.037 g as the weight of ashed uncalcified marrow. On the percentage basis assumed, the uncalcified marrow in the fresh marrow should be 3.70 g, and adding the calculated weight of bone, 0.557, we find the total fresh weight is 4.28 g instead of 4.22 g.

On the basis of fresh marrow (bone free) the Th^{228} content of 3.7 g of marrow would be 0.072 $\mu\mu\text{c}/\text{g}$ fresh marrow. The total $\mu\mu\text{c Th}^{228}$ in the yellow marrow for this subject (1500 g) would then be about $1.1 \times 10^{-10} \text{ c}$ (order of 5×10^{-4} of the entire Th^{232} content) at the time of DBP separation, 0.6 years after amputation.

C. Trabecular Bone

Trabecular epiphyseal bone from the head of the femur and from the tibia was cross sectioned to give large samples in order to have as much specific activity as possible. One section of each, femur sample 284B-5 and tibia sample 284B-3, was extracted with ED. A second tibia section, sample 284B-4, was extracted with acetone in an endeavor to determine whether the highly alkaline ED might cause insoluble thorium to remain with the inorganic bone fraction (acetone is good extractant for lipids and is not alkaline).

The modified soxhlets used in the Losee extraction method had to be changed to the standard type, using Whatman fat-free extraction thimbles because all three samples were so large they prevented proper

emptying of the Losee soxhlets. The extractants were removed completely from the bone samples by extended water extractions; the residues from the organic and water extractions were HNO_3 -treated, combined, and partially oxidized before muffling at 620°C .

Table 17 presents a summary of the analyses for Th^{228} , Ra^{228} and Th^{232} in parts A and B; in part C, the acetone-extracted trabecular tibia was assayed for Th^{228} only, as Ra^{228} and Th^{232} assays were not warranted.

Table 17
Trabecular bone analyses for Th^{228} , Ra^{228} and Th^{232}

A. Femur, ED extracted					
Sample No.	284B-5 Inorganic	284B-5 Inorganic	284B-5 Inorganic	284B-5-0 Organic	284B-5 and 284B-5-0
Nuclide	Th^{228}	Ra^{228}	Th^{232} *	Th^{228}	Th^{228}
Fresh weight, g	18.0	18.0		-	-
Ashed weight, g	6.069	6.069		0.097	6.166
$\mu\mu\text{c} \pm 0.9 \text{ E}$ in sample	4.64 ± 0.25	-		0.185 ± 0.014	4.825 ± 0.25
$\mu\mu\text{c} \pm 0.9 \text{ E}$ per g ash	0.765 ± 0.040	0.55 ± 0.05	0.55 ± 0.006	-	0.782 ± 0.04
Date of analysis	2/10/61	7/17/61		3/8/61	
$\mu\mu\text{c}$ of nuclide on 2/15/60	0.840 ± 0.044	0.60 ± 0.06	0.55 ± 0.006		0.85 ± 0.040
$\text{Th}^{228}/\text{Ra}^{228}$	1.40				1.41
B. Tibia, ED extracted					
Sample No.	284B-3 Inorganic	284B-3 Inorganic	284B-3 Inorganic	284B-3-0 Organic	284B-3 and 284B-3-0
Nuclide	Th^{228}	Ra^{228}	Th^{232}	Th^{228}	Th^{228}
Fresh weight, g	17.5	17.5	17.5	-	-
Ashed weight, g	4.92	4.92	4.92	0.081	5.00
$\mu\mu\text{c} \pm 0.9 \text{ E}$ in sample	4.32 ± 0.36	-	2.71 ± 0.03	0.130 ± 0.02	4.45 ± 0.36
$\mu\mu\text{c} \pm 0.9 \text{ E}$ per g ash	0.88 ± 0.07	0.80 ± 0.08	0.55 ± 0.006	-	0.89 ± 0.07
Date of analysis	9/29/60	6/22/61	7/10/61	10/5/60	
$\mu\mu\text{c}$ of nuclide on 2/15/60	0.878 ± 0.070	0.91 ± 0.09	0.55 ± 0.006		0.888 ± 0.09
$\text{Th}^{228}/\text{Ra}^{228}$	0.965				0.975
C. Tibia, acetone extracted. Th^{228} analyses.					
Sample No.	284B-4 Inorganic	284B-4-0 Organic	284B-4 and 284B-4-0		
Fresh weight, g	16.5	-	-		
Ashed weight, g	5.51	0.012	5.52		
$\mu\mu\text{c} \pm 0.9 \text{ E}$ in sample	4.01 ± 0.35	0.00 ± 0.008	4.01 ± 0.35		
$\mu\mu\text{c} \pm 0.9 \text{ E}$ per g ash	0.73 ± 0.06	-	0.73 ± 0.06		
Date of analysis	9/29/60	10/5/60			

* Assume Th^{232} content the same as in trabecular tibia.

1. Trabecular Femur

The Th^{232} in the cortical femur was $0.22 \mu\mu\text{c/g}$ ash, while that in the trabecular femur was not determined. Assuming that the value was the same as that found in trabecular tibia, the Ra^{228} and Th^{228} values were corrected to amputation time.

The ratio of $\text{Th}^{232}/\text{Ra}^{228}/\text{Th}^{228} = 1.0:1.1:1.5$.

The relation between the three nuclides at amputation was:

Nuclide	$\mu\mu\text{c/g}$ ash	$\mu\mu\text{c}$ in skeleton*	% of Th^{232} injected (0.22 μc)
Th^{232}	0.55	1540	0.74
Ra^{228}	0.60	1680	0.76
Th^{228}	0.84	2352	1.07

* $\mu\mu\text{c/g}$ ash x 2,800 g skeletal ash

2. Trabecular Tibia

The weights of the organic ash from the femur and tibia were about the same, and the variation in the total Th^{228} in each was not great; the time factors for the different analyses varied considerably.

The Th^{232} present in trabecular tibia was $2\frac{1}{2}$ times greater than that found in cortical femur (284B-6), and the ratio in the tibia for $\text{Th}^{232}/\text{Ra}^{228}/\text{Th}^{228}$ was 1.0:1.65:1.60; the Ra^{228} and Th^{228} values at amputation were corrected as previously.

The relation between the nuclides in this case at amputation was:

Nuclide	$\mu\mu\text{c/g}$ ash	$\mu\mu\text{c}$ in skeleton*	% of Th^{232} injected (0.22 μc)
Th^{232}	0.55	1540	0.70
Ra^{228}	0.91	2540	1.15
Th^{228}	0.878	2458	1.12

* $\mu\mu\text{c/g}$ ash x 2,800 g skeletal ash

The ratios of $\text{Th}^{228}/\text{Ra}^{228}$ in cortical femur (284B-6) cortical tibia (284B-2) trabecular femur (284B-5) and trabecular tibia (284B-3), were 1.30, 1.02, 1.40 and 0.97 respectively. A ratio of less than unity is not very informative unless burden, time and actual equilibrium status of the Thorotrast at injection are known. A ratio >1.0 but <1.487 would imply that the Ra^{228} had not been available to the bone for at least 4.54 years, the time when the activity of Th^{228} growing in from an isolated source of Ra^{228} (5.80 year half-life),⁽¹²⁾ equals that of the parent.

These data, involving an atherosclerotic patient, whose bone examined microradiographically⁽¹³⁾ shows distinct evidence of limited circulation, do not furnish sufficient evidence of whether the differences in

the $\text{Th}^{228}/\text{Ra}^{228}$ ratios are a general phenomenon in specific loci of normal bone; they do suggest the possibility that these ratios may be indicative of circulation damage.⁽¹⁴⁾

3. Acetone-Extracted Trabecular Tibia

The significance of the acetone extraction of trabecular tibia indicates it is useless because no thorium activity was found in the DBP extract of the ash from the large volume of fatty material obtained. The total Th^{228} activity is about 18% less than in the ED-extracted bone although these were adjacent sections which differed in fresh weight by only 1 gram; the acetone and water extracts yielded only 12 mg of ash. The conclusion is definite that acetone is an inadequate extractant for the organic matrix of bone.

Ethylenediamine extractions have always yielded Th^{228} activity in both organic and inorganic portions. In the extractions cited in this report, trabecular bone yielded $3.0 \pm 0.5\%$ Th^{228} in the organic extract, and the organic ash was 1.6% of the total ash. Comparing the organic and inorganic portions in the single ED extraction for cortical bone cited here with the two ED extracted cortical femur samples from a dial painter cited previously,⁽¹⁰⁾ we note that the Th^{228} activity found in the organic phase in 284B-6 was 1.7% of the total, while only 0.6% of the Th^{228} activity from the femur from the dial painter was found in the organic portions. The comparison of organic ash to total ash also varied; 284B-6 organic ash was 3.5% of the total, while for those previously cited it was 1.9%. The dial painter samples were about 7.0 g fresh sample, with about 4.0 g average for the ashed weight. All of the extractions and treatment of organic and inorganic phases were similar; except for those previously cited, the Løsee soxhlet and sintered glass thimble were used.

This does not definitely answer the question as to whether thorium may be presumably in the matrix but left in an insoluble form with the inorganic bone during ED extraction. However, we have shown that ED extraction does not remove Th^{228} from the cortical femur. The Th^{228} content per gram of ashed bone in 284B-1a, directly ashed, and in 284B-6, ED extracted, was the same (see Table 15).

Trabecular bone, ED extracted, yielded more Th^{228} activity in the organic phase ($3.0 \pm 0.5\%$ of the total activity) than ED extracted cortical (1.5%); this result is expected since the proportion of organic matter in trabecular bone is much greater than in cortical. The $\text{Ra}^{228}/\text{Th}^{228}$ ratios in trabecular bone were close to theoretical equilibrium; hence, it appears that ED extraction does not plate out thorium under these conditions; or if it does, it plates it to the same extent as Ra^{228} . It does show definitely that acetone does not remove the thorium-bearing matrix during extraction.

D. Thorotrast Cases Previously Reported(1-3)

The analyses for Th^{228} and Th^{232} on a tooth from 04-101* were reported previously without a Ra^{228} determination because the nitric acid solution was used for a Ra^{226} analysis and discarded. The Ra^{226} present was $0.040 \pm 0.008 \mu\mu\text{g/g}$ ash, while the Th^{228} was $1.39 \pm 0.18 \mu\mu\text{c/g}$ ash on 7/11/58, 350 days after extraction.

This subject had received 10 ml Thorotrast i.v. in 1939. The in vivo gamma-ray spectrum of this patient was reported by Miller⁽¹⁵⁾ who stated, "Since the subject was found to contain 1.22×10^{-7} c MsThII and 1.1×10^{-7} c ThC", or 12% more MsThII than the source, it seems likely that the patient was eliminating about 12% of the daughter products between MsThII and ThC".

A male subject, 04-102,** who died about 1/28/59 at 32 years of age, had received 75 ml of Thorotrast more than 16 years previously. The Th^{228} value reported⁽²⁾ was made from an autoradiograph of a vertebral section a few weeks after death.

Th^{228} and Ra^{228} analyses were made on an ED-cleaned (by refluxing, not soxhlet) portion of vertebra. These analyses are summarized in Table 18; no Th^{232} analysis was made. The calculations were made using only growth and decay factors for Ra^{228} and Th^{228} .

Table 18

Th^{228} and Ra^{228} analyses on 04-102

Sample No.	46B	46B
Nuclide	Th^{228}	Ra^{228}
Fresh weight, g	2.296	2.296
Ashed weight, g	1.867	1.867
$\mu\mu\text{c}$ of nuclide per g ash	25.8 ± 1.50	32 ± 3.0
Date of analysis	5/13/59	6/19/59
$\mu\mu\text{c}$ of nuclide on 1/28/59	24.8 ± 1.40	34.0 ± 3.0
$\text{Th}^{228}/\text{Ra}^{228}$	0.73	

*References 1 and 2.

**Reference 2

L. D. Marinelli directed the experiments and measurements reported herein. Corrections for the assays and their interpretation for Thorotrast patients are his contribution.

References

1. L. D. Marinelli and A. F. Stehney. Radioactivity in Bones of Patients Injected with Thorotrast. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1958. ANL-5967, pp. 105-6.
2. L. D. Marinelli. Epidemiological Studies on Radiation Cancerogenesis in the Human Skeleton. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049, p. 30. [Presented at the Roentgen Ray Society Meeting, September 22-25, 1959, Cincinnati, Ohio.]
3. L. D. Marinelli. The Effect of Chronic Low Levels of Radiation in Man: The Contribution of Epidemiological Studies. Trans. 9th Intern. Congr. of Radiology, Munich, July 23-30, 1959. Ed. B. Rajewsky, Stuttgart: Georg Thieme Verlag, 1960, pp. 1234-1238.
4. H. F. Lucas and F. H. Ilcewicz. The Natural Ra^{226} Content of Illinois Water Supplies. J. Am. Water Works Assoc. 50 (11) 1523-32 (1958).
5. H. F. Lucas. The Correlation of the Natural Radioactivity in the Human Body and its Environment: Uptake and Retention of Ra^{226} from Food and Water. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1960. ANL-6297, pp. 55-66.
6. A. F. Stehney, D. E. Wallace, and F. H. Ilcewicz. Isolation of Th^{228} from Bone with Dibutyl Phosphate. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June 1956. ANL-5596, pp. 12-14.
7. D. P. Krause. Mesothorium Measurements on Some Radium-Bearing Potable Waters. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1958. ANL-5967, pp. 126-128.
8. R. B. Holtzman. Some Determinations of the RaD and RaF Concentrations in Human Bone. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June 1960. ANL-6199, pp. 94-106.
9. F. L. Losee and L. A. Hurley. Successful Cross-species Bone Grafting Accomplished by Removal of the Donor Organic Matrix. Research Report. National Naval Medicine Center, December 5, 1956. Volume 14, pp. 911-948.

10. D. E. Wallace, A. F. Stehney, and F. H. Ilcewicz. Th^{228} , Ra^{228} and Ra^{226} in Bone. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1958. ANL-5967, pp. 107-110.
11. J. B. Hursh, A. Lovaas, and E. Biltz. Ra^{226} in Bone and Soft Tissues of Man. U.S. Atomic Energy Commission Report, UR-581, October 31, 1960.
12. C. W. Mays, D. R. Atherton, F. W. Bruenger, B. J. Stover, W. M. Hammer and W. W. Parmley. Redetermination of the Half-period of Ra^{228} (Mesothorium). Radiobiology Division, Univ. of Utah, Report COO-222 (1960). pp. 27-48.
13. R. E. Rowland. The Microradiographic Appearance of Bones Subject to Chronic Vascular Insufficiency. This report, p. 36.
14. L. D. Marinelli, Personal communication.
15. C. E. Miller. The Human Spectrometer. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1957. ANL-5829, pp. 144-166.

APPENDIX I

Calculations for Ra²²⁸ and Th²²⁸ at t₁, time of sample collection.*

Example used, 284B-1a, cortical femur:

t₁ = 2/15/60, date of amputation

Th²²⁸ on 5/13/60, 90 days after t₁ = 0.595 ± 0.007 μμc/g ash

Ra²²⁸ on 1/12/61, 245 days after 5/13/60 = 0.45 ± 0.04 μμc/g ash

Th²³² on 7/12/61 = 0.22 ± 0.03 μμc/g ash.

Since the Ra²²⁸ solution measured on 1/12/61 did not contain Th²³²;

Ra²²⁸ on 5/13/60 = 0.45/0.92 (Ra²²⁸ decay in 245 days) = 0.49 μμc

Th²³² on 5/13/60, same as on 7/12/61, 0.22 μμc/g ash.

We may proceed to calculate the Ra²²⁸ (in the presence of Th²³²) from 5/13/60 to 2/15/60 by correcting for the decay of Ra²²⁸ present in excess of equilibrium, that is, at t₁, 2/15/60:

$$\begin{aligned} \text{Ra}^{228} &= \text{Th}^{232} - \text{Ra}^{228} + \frac{\mu\mu\text{c Ra}^{228} \text{ on } 5/13/60 - \mu\mu\text{c Th}^{232}}{\text{decay Ra}^{228} \text{ in 90 days}} \\ &= 0.22 + \frac{0.49 - 0.22}{0.97} = 0.496 \mu\mu\text{c/g ash} \end{aligned}$$

And similarly for the Th²²⁸ (if x is the Th²²⁸ on 2/15/60):

$$\begin{aligned} \text{Th}^{228} \text{ on } 5/13/60 &= \text{Th}^{232} - \text{Th}^{228} + \text{Ra}^{228} \text{ on } 5/15/60 - \text{Th}^{232} \times \frac{\text{Th}^{228} \text{ growth}}{\text{in 90 days}} + \frac{\text{Th}^{228} \text{ on } 2/15/60}{- \text{Th}^{232}} \times \frac{\text{Th}^{228} \text{ decay}}{\text{in 90 days}} \\ 0.595 &= (0.22) + (0.496 - 0.22) \times 0.086 + (X - 0.22) \times 0.914 \end{aligned}$$

X, or Th²²⁸ at t₁ = 0.600 μμc/g ash.

*Ra²²⁸ half-life used in calculations was 5.80 years.⁽¹²⁾

FURTHER OBSERVATIONS ON Th^{228} AND Ra^{226} IN BONES FROM DIAL PAINTERS AND MEDICAL PATIENTS

Dorothy E. Wallace

Results from Th^{228} and Ra^{226} analyses of four of Martland's bone specimens from dial painters have been completed. Autoradiographic measurements of hotspots and diffuse concentrations have been reported by Rowland;⁽¹⁾ these subjects are 03-997,* or E. E., 03-996* or I. L. and 03-999,* or K. S. and Case No. 6.

The samples E. E. and I. L. were very small pieces, partially or wholly imbedded in plastic; these early dial painters, described by Hoecker⁽²⁾ and originally by Martland,⁽³⁾ were believed to have terminal body burdens of 50 and 8 μg of radium, respectively, during the 1920's when they were examined. The sample K. S., Martland's Case No. 6,⁽⁴⁾ was preserved in formalin; this subject was a dial painter and instructor, whose terminal body burden of radium was estimated at 19 μg .

The fourth sample 03-998,* Martland's S. M. and Case No. 1,^(5,6) was the first case of death among the dial painters. Martland's statement about the gamma electroscope used to determine the radioactivity in the subject on 6/16/25 was "This is the first time, as far as we know, that this test was made on a human being the girl died a few days later; after autopsy in this case, it was estimated that the skeleton contained 180 μg of radioactive substances, about 70% being from mesothorium and its decay products." A subsequent report in 1931⁽⁴⁾ gave in "Table I, Deaths of So-called Radium Poisoning Occurring among 800 employees of a New Jersey Plant Manufacturing Luminous Watch Dials," this listing:

"No.	Name	Age	Work	Date of Death
9	S. M.	33	Dial Painter	6/18/1925"

The description on the label on the outside of the museum jar containing the specimens included the following, "1925. First reported death due to radium poisoning . . . equivalent of 90 μg of radium in the body, found during life and after death by electroscopic methods."

Portions of the white, ashed bone specimen, labelled femur, were used for chemical, α -pulse spectrometer** and autoradiographic measurements.⁽⁷⁾

*Atomic Energy Commission, Punched Card Code Number.

** α -pulse spectrometer analysis by H. F. Lucas, Jr.

A fifth specimen, 03-479, left metatarsal bones from a dial painter living in Illinois, was received in May 1959 following the removal of a bunion. The Ra^{226} body burden of this subject was measured by gamma-ray spectrometry⁽⁸⁾ on 10/31/58 and found to be 7×10^{-8} g Ra^{226} .

The analyses were made as follows: Th^{228} by dibutyl phosphate extraction of thorium from a nitric acid solution of ashed bone;⁽⁹⁾ Ra^{226} by the radon emanation method.⁽¹⁰⁾ Table 19 summarizes the results of these analyses.

Table 19
 Th^{228} and Ra^{226} content of bones from dial painters

Subject	Body burden, $\mu\text{g Ra}^{226}$	Years acquired, t_1	Date of Th^{228} analyses	$\mu\mu\text{c Ra}^{226}$ per g ash	$\mu\mu\text{c Th}^{228}$ per g ash	% Ra^{228} at t_1 relative to Ra^{226}
03-997 (E. E.)	50	1917-1920 1920	12/5/58	$3500 \pm 30^{**}$	$176 \pm 11^{**}$	$320 \pm 20^{**}$
03-996 (I. L.)	8	1917-1921 1921	12/19/58	2763 ± 15	5.8 ± 0.36	12 ± 0.7
03-999 (K. S.)	19	1916-1922 1922	4/22/59	2935 ± 58	5.55 ± 0.24	11 ± 0.73
03-998 (S. M.)	90	1917-1923 1923	1/25/61	$4789 \pm 85^*$	72.5 ± 0.40	96.4 ± 5.0
03-479	0.07	1924	7, 3/59	25 ± 0.05	0.0425 ± 0.006	7.6 ± 1.0

* Ra^{226} content, g ash by α -pulse spectrometer analysis = 4800 mg.

**90% confidence interval.

A medical case 03-214 or No. 114,⁽¹¹⁾ was reported to have a body burden of 1.6 μg radium in 1952; a subsequent measurement 3/30/59⁽⁸⁾ gave 1.4 μg . The specimen, pieces of jaw bone from a mastoid operation, received 4/59, only gave 0.126 g ash, with a total of 0.005 ± 0.009 $\mu\mu\text{c Th}^{228}$ for the sample. This is in the same range as that found per g of ash in normal bone,⁽¹²⁾ but with a far greater error, and as such is not significant. It confirms previous findings that the radium solutions administered medically some thirty years ago were relatively free of mesothorium. The 0.70% of Ra^{228} (6.7 year half-life) at t_1 , relative to Ra^{226} , agrees well with that of 0.66% reported in 1955 by Norris et al.⁽¹³⁾

The following summary is offered for comparative results.

Subject	Body burden, $\mu\text{g Ra}^{226}$	Year, t_1	Date of Th^{228} analysis	$\mu\mu\text{c Ra}^{226}$ per g ash	$\mu\mu\text{c Th}^{228}$ per g ash	% Ra^{228} at t_1 relative to Ra^{226}
03-214 (114)	1.4	1930	5/1/59	78 ± 3	0.0365 ± 0.070	0.70 ± 1.3

References

1. R. E. Rowland. Late Observations on the Distribution of Radium in the Human Skeleton. Argonne National Laboratory Radiological Physics Division Semiannual Report, (July through December 1959) ANL-6104, pp. 16-29; Radioisotopes in the Biosphere, Ed. R. S. Caldecott and L. A. Snyder, Minneapolis: Univ. Minn., 1960 pp. 339-353, (with J. H. Marshall) Radium in Human Bone: The Dose in Microscopic Volumes of Bone. Radiation Research 11 299-313 (1959).
2. F. E. Hoecker and P. G. Rooft. Studies of Radium in Human Bone. Radiology 56 89-98 (1951).
3. H. S. Martland. Occupational Tumors: (6) Bones. In International Labor Office, Geneva, Occupation and Health. Supplement. October, 1939, Brochure 13, pp. 1-23.
4. H. S. Martland. The Occurrence of Malignancy in Radioactive Persons. Am. J. Cancer 15 2435 (1931).
5. H. S. Martland. Some Unrecognized Dangers in the Use and Handling of Radioactive Substances. J. Am. Med. Assoc. 85 1769, (1925).
6. H. S. Martland. Occupational Poisoning in the Manufacture of Luminous watch Dials. J. Am. Med. Assoc. 92 466 (1929).
7. R. E. Rowland. Detailed Autoradiographic and Microradiographic Studies of Bone from Human Radium Cases. This report, page 5.
8. C. E. Miller. The Human Spectrometer. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1957. ANL-5829, pp. 144-166.
9. A. F. Stehney, D. E. Wallace and F. H. Ilcewicz. Isolation of Th^{228} from Bone with Dibutyl Phosphate. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June 1956. ANL-5596, pp. 12-16.
10. H. F. Lucas and F. H. Ilcewicz. The Natural Ra^{226} Content of Illinois Water Supplies. J. Am. Water Works Assoc. 50 (11) 1523-1532 (1958); The Correlation of the Natural Radioactivity in the Human Body and its Environment: Uptake and Retention of Ra^{226} from Food and Water. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1960. ANL-6297, pp. 55-66.

11. W. B. Looney, R. J. Hasterlik, A. M. Brues and E. Skirmont. A Clinical Investigation of the Chronic Effects of Radium Salts Administered Therapeutically. *Am. J. Roentgenol.* 73 1006 -1037 (1955).
12. D. E. Wallace, A. F. Stehney and F. H. Ilcewicz. Th^{228} , Ra^{228} , and Ra^{226} in Bone. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1958. ANL-5967, pp. 107-110.
13. W. P. Norris, T. W. Speckman and P. F. Gustafson. Studies of Metabolism of Radium in Man. *Am. J. Roentgenol.* 73 785-802 (1955).

Th²²⁸ AND Ra²²⁶ ANALYSES OF BONES FROM KERALA, INDIA

Dorothy E Wallace

Humans subject to external background gamma radiation approximately ten times higher than the average estimated for the world at large have been referred to in epidemiological studies on cancerogenesis by Marinelli.^(1,2)

Bones and flesh, preserved in formalin wrappings, from seven subjects in Kerala (formerly Travancore), India, were received by Dr. R. J. Hasterlik,* Argonne Cancer Research Hospital, University of Chicago, Illinois, in November 1954. The period or periods of elapsed time between procurement of the samples and their receipt is unknown.

Analyses

The entire mass of bone and flesh was subject to gamma-ray spectroscopic measurement, using an 8" x 4" NaI(Tl) crystal in the steel room; but it showed no detectable activity despite several attempts in 1957 and 1958.⁽³⁾

Some of the bone specimens were too small (0.5 to 5.0 g dry weight) for chemical analysis for Th²²⁸ by the dibutyl phosphate method.⁽⁴⁾ Three samples (our No. 36B) were analyzed for Th²²⁸ and Ra²²⁶. One of them, 36B-5, was a very poor sample; it looked corroded or "moth-eaten," was reddish-brown in color, and very irregular in structure. The other samples used were clean cortical specimens. An autoradiograph of a section of 36B-5 yielded no information.** Considerable interference due to ferric ions found in the nitric acid solution of this ashed bone made the Th²²⁸ values for 36B-5 questionable; the ferric ions do not interfere with the Ra²²⁶ determination by the radon emanation method of Lucas and Ilcewicz.⁽⁵⁾ The values for 36B-5 are included in the summary in Table 20.

From the percentage ratios of Th²²⁸/Ra²²⁶ in 36B-5 and 36B-6, even allowing for the ferric ion interference, which would cause the Th²²⁸ values to be low, it is obvious that these samples are from two different persons.

*The specimens were sent to Dr. R. J. Hasterlik by Dr. M. Thangavelu, Department of Pathology, Travancore-Cochin Medical School, Trivandrum, Kerala, India. For these, and many other bone samples, our thanks are expressed to Dr. R. J. Hasterlik.

**Autoradiography performed by R. E. Rowland.

The second determination on 36B-2 was made to verify the fact that this sample was from still another subject. It is obvious that the individual from whom this specimen was obtained had had an environmental exposure different from the other two.

Table 20
Th²²⁸ and Ra²²⁶ in dry cortical bone from Kerala, India

Sample No.	Nuclide	Dry weight, g	620°C ashed weight, g	$\mu\mu\text{C} \pm 0.9 \text{ E}^*$ in sample	$\mu\mu\text{C} \pm 0.9 \text{ E}^*$ per g ash	Date of analysis	% Th ²²⁸ /Ra ²²⁶ at date Th ²²⁸ analysis
36B-6	Th ²²⁸	7.131	4.204	0.117 ± 0.013	0.028 ± 0.003	12/8/58	36.8 ± 4.0
	Ra ²²⁶	7.131	4.204	-	0.076 ± 0.005	1/22/59	
36B-5	Th ²²⁸	5.585	2.747	0.058 ± 0.023	0.021 ± 0.008	12/8/58	69 ± 27
	Ra ²²⁶	5.585	2.747	-	0.0306 ± 0.003	1/22/59	
36B-2 (1)	Th ²²⁸	5.415	3.158	0.034 ± 0.013	0.011 ± 0.004	2/16/59	10.5 ± 4.2
	Ra ²²⁶	5.415	3.158	-	0.105 ± 0.005	4/6/59	
36B-2 (2)	Th ²²⁸	5.724	3.345	0.036 ± 0.013	0.011 ± 0.004	7/24/59	9.0 ± 3.6
	Ra ²²⁶	5.724	3.345	-	0.127 ± 0.004	9/8/59	

*90% confidence interval.

Comparison with Th²²⁸/Ra²²⁶ Ratios in Bones from Natural Ra²²⁶ Activity Sources

The natural radioactivity of humans in relation to the uptake and retention of Ra²²⁶ from food and water has been discussed by Lucas.⁽⁶⁾ Comparison of the Ra²²⁶ and the Th²²⁸ bone ash content and their ratios with the Kerala samples, is pertinent.

The lowest Ra²²⁶ value of $0.030 \pm 0.003 \mu\mu\text{g/g}$ ash is comparable to natural water levels for Ra²²⁶ in La Grange and Peoria, Illinois ($0.20 - 0.48 \mu\mu\text{g/l.}$). Webster, Iowa with $0.85 \mu\mu\text{g Ra}^{226}/\text{l.}$ falls in this range; but the sample from there (284B or 04-104*) is from a Thorotrast patient discussed in this report for thorium distribution in cortical and trabecular bone.⁽⁷⁾ The Th²²⁸/Ra²²⁶ ratio for the Kerala sample in this range is the highest, $69 \pm 27\%$. But for 284B the Th²²⁸ content is 175 times that of the Ra²²⁶; because of the body burden of the Th²³² and daughters, this specimen is not representative for the natural radioactivity expected for Th²²⁸ content.

The intermediate Ra²²⁶ content of $0.076 \pm 0.005 \mu\mu\text{g/g}$ ash is characteristic of samples reported from Aurora and Elmhurst ($5.0 \mu\mu\text{g/l.}$), and Peru, Illinois ($2.5 \mu\mu\text{g/l.}$).

The highest Ra²²⁶ content of $0.116 \pm 0.007 \mu\mu\text{g/g}$ ash (average of 2 analyses on 36B-2) can be compared with samples from Ottawa, Illinois

*Atomic Energy Commission, Punched Card Code No.

(4.6 $\mu\mu\text{g/l.}$). Another sample from Granville, Illinois (10.5 $\mu\mu\text{g/l.}$) which showed the highest Ra^{226} value reported by Lucas, gives a $\text{Th}^{228}/\text{Ra}^{226}$ ratio the same as for one Ottawa and Kerala sample 36B-2. The sample 36B-5 ratio, instead of falling into the lowest range, is the same as the highest of the Aurora ratios.

The tabulation of these comparisons in Table 21 shows that a considerable range can exist for a city; e.g., the Aurora range is from 20 to 70%. As has been cited by Lucas, much depends upon the source as well as the treatment of water within a city, and the residence history of the individual; the latter, in these cases is incomplete.

Table 21

Comparison of $\text{Th}^{228}/\text{Ra}^{226}$ ratios from natural radioactivity and Kerala specimens

Mean* $\mu\mu\text{g Ra}^{226}$ per g ash	$\mu\mu\text{g Ra}^{226}$ * per g ash	Sample No.	City	$\mu\mu\text{c Th}^{228}$ per g ash	% $\text{Th}^{228}/\text{Ra}^{226}$
0.024	0.033 ± 0.006	386	Peoria	0.0084 ± 0.006	25 ± 1.8
	0.030 ± 0.003	36B-5	Kerala	0.021 ± 0.008	69 ± 27
0.081	0.095 ± 0.003	113	Aurora	0.022 ± 0.002	23 ± 2
	0.078 ± 0.003	124	Elmhurst	0.021 ± 0.003	27 ± 4
	0.080 ± 0.002	150	Aurora	0.024 ± 0.003	30 ± 4
	0.042 ± 0.002	111	Aurora	0.016 ± 0.002	39 ± 8
	0.105 ± 0.002	118	Peru	0.043 ± 0.003	41 ± 3
	0.083 ± 0.004	165	Aurora	0.056 ± 0.006	67 ± 8
	0.076 ± 0.005	36B-6	Kerala	0.028 ± 0.003	37 ± 4
0.112	0.110 ± 0.008	120	Ottawa	0.016 ± 0.003	16 ± 3
	0.118 ± 0.008	164	Ottawa	0.043 ± 0.010	36 ± 9
	0.116 ± 0.007	36B-2	Kerala	0.11 ± 0.004	10 ± 4
-	0.472 ± 0.010	148	Granville	0.071 ± 0.010	15 ± 2

*Reference 6.

References

1. L. D. Marinelli. Epidemiological Studies on Radiation Cancerogenesis in the Human Skeleton. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049. pp. 30-42. [Presented at the Roentgen Ray Society Meeting, September 22-25, 1959, Cincinnati, Ohio.]
2. L. D. Marinelli. Radioactivity and the Human Skeleton (Janeway Lecture). Am. J. Roentgenol. 80 729-739 (1958).

3. C. E. Miller. Low Intensity Spectrometry of the Gamma Radiation Emitted by Human Beings. Progress in Nuclear Energy, Series VII, Medical Sciences, Vol. 2, New York: Pergamon Press, 1959, pp. 87-104; Proc. Second Intern. Conf. on Peaceful Uses of Atomic Energy, Geneva, 1958. Geneva: United Nations, 1958. Vol. 23, pp. 113-122.
4. D. E. Wallace, A. F. Stehney and F. H. Ilcewicz. Isolation of Th^{228} from Bone with Dibutyl Phosphate. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1956. ANL-5596, pp. 12-14.
5. H. F. Lucas and F. H. Ilcewicz. Natural Radium-226 Content of Illinois Water Supplies. J. Am. Water Works Assoc. 50 (11) 1523-1532 (1958).
6. H. F. Lucas, Jr. Correlation of the Natural Radioactivity of the Human Body to that of its Environment: Uptake and Retention of Ra^{226} from Food and Water. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1960. ANL-6297, pp. 55-66.
7. D. E. Wallace, H. F. Lucas, Jr., D. P. Krause, and F. H. Ilcewicz. Radioactivity in Bones of Patients Injected with Thorotrast: II. Variations in Distribution in Cortical, Trabecular and Marrow Portions. This report, page 51.

FURTHER STUDIES ON NATURAL RADIOELEMENTS IN BOVINE BONES AND TEETH

Elvira R. Di Ferrante

Results published in a previous report⁽¹⁾ suggested that the concentration of Ra^{226} in the ash of bovine teeth formed after the weaning age is very nearly equal to that in bone. As a consequence, the estimation of body burden can be based on tooth analysis, if the limitation in question is kept in mind. In order to confirm this assertion, further analyses were performed, and the results are reported here.

It is known from the literature^(2,3) that there is a different chronology in the eruption of permanent teeth. Therefore, as calcification takes place, their Ra^{226} content reflects mainly the Ra/Ca in the diet at that time since there is little change after mineralization is completed. If two teeth are formed within a certain interval, it is possible to infer, from their Ra^{226} content, whether or not a sensible change in diet occurred in the interim.

Among the 25 samples of bovine jaw bones with teeth, 11 belonging to animals with permanent dentition (≥ 5 y old) were chosen for a study of Ra^{226} content in all the teeth available.

The data obtained are graphically represented in Figure 11, where radium contents are plotted vs tooth formation times. The formation time, corresponding to a certain age of the animal, has been arbitrarily set as the midtime between tooth emergence into oral cavity and completion of root formation. The data give evidence of some fluctuations in the radium content of teeth from the same animal; however, they also show that reliable information about skeletal burden may be obtained from teeth formed between approximately 2 and 4 years of age, thus confirming the previous findings.

The first molars, M1, because of their partial formation before birth and completion within the first year of age of the animal, are subject to a Ra^{226} -to-Ca discrimination of placental, milk, and intestinal origin.⁽⁴⁾ They may, therefore, have a content different from that present in bone at a more advanced age and from that existing in permanent teeth formed after substantial dietary changes have occurred.

Sample 23C is representative of a case of high discrepancy between bone and first molar contents: although the latter gives a value of $0.800 \mu\mu\text{g Ra}^{226}/\text{g ash}$, the bone gives $2.58 \mu\mu\text{g/g ash}$. The second incisor, I2, with a formation time at about 3 years, has a content of $2.52 \mu\mu\text{g Ra}^{226}/\text{g ash}$. These results are indicative of a great increase in radium intake after formation of the first molar. The analysis of the fourth incisor, I4, formation

time ~ 4 years, gives $2.35 \mu\mu\text{g/g}$ ash for the crown and $3.5 \mu\mu\text{g/g}$ ash for the root: since the complete calcification of the root occurs after that of the crown, the higher value suggests a further enrichment of Ra^{226} in the diet (including water), although this enrichment is not confirmed by the value obtained for the bone. This apparent contradiction may be due to the fact that the age at the death was close to that of the formation of the tooth and, therefore, the skeletal burden could not be affected yet by the change because of the long turnover time. Also for Sample 16C, a great difference exists between bone and first molar radium contents, but in this case the tooth presents a much higher content: $1.92 \mu\mu\text{g/g}$ ash, vs. a bone value of $0.970 \mu\mu\text{g/g}$ ash, close to that of the other permanent teeth. This case strongly contrasts with the previous one and suggests that the specific radium intake of the mother (affecting the content of teeth partially formed at birth) must have been higher than that of its progeny by a factor substantially greater than would be expected from the placental discrimination. For other samples (1C, 5C, 11C, 25C) discrepancies between bone and first molars have been observed, but they are much less evident than those observed for Samples 23C and 16C.

Figure 11

Ra^{226} content in permanent teeth and bone

A new sample, 26C, belonging to a young animal ($2\frac{1}{2}$ y old), was analyzed. The results are shown in Table 22. The Ra^{226} content is higher in the deciduous premolars, DP1, DP2, DP3, than in the permanent molars, M1, M2, M3. Since deciduous teeth are almost completely formed at birth and do not reflect the radium concentration of the diet as well as permanent teeth, a decrease in the radium intake must have occurred after the weaning age. The decrease, probably due to a diet of Ra^{226} content lower than that in the diet of the mother, is not revealed by the bone whose content is still representative of an early radium intake. Samples 12C, 15C, 20C, 24C⁽¹⁾ were also from young animals, and Ra^{226} analyses were performed using deciduous teeth; comparing the teeth content with that in the bones, there is an indication of an increase of skeletal burden with age. In these cases, during the prenatal period, discrimination factors probably controlled the radium intake, which, after the weaning age (as the calves passed to a normal diet) increased in value and affected the bone content.

Table 22

Ra^{226} in Sample 26C ($\mu\mu\text{g/g}$ ash)*

Location and age	Bone	DP1**	DP2	DP3	M1	M2	M3 (not erupt.)
Coal Valley, Ill. 2y 6m	0.464	0.460	0.500	0.480	0.338	0.300	0.226

*Average value of two or more analyses; range of estimated error = ± 0.005 - $0.02 \mu\mu\text{g/g}$ ash.

**DP = deciduous premolar; M = molar.

In order to establish the entity of the discrimination factors, efforts were made to obtain some samples of cows with fetuses. Only recently a jaw bone with teeth from a cow approximately 10 years old and the jaw bone of its 8-month fetus were received. Analyses are currently in progress. It was observed that, in accordance with classical data, the jaw of the fetus presented second and third premolars and first, second and third incisors already erupted. All the teeth were not completely calcified and the roots were just forming.

All observations made thus far lead to the conclusion that in cases in which great changes in the radium content of the diet are suspected, or whenever samples from very young animals or first molars are available, the analysis of tooth specimens can be informative only about the period of formation of the tooth itself. The knowledge of animal's history facilitates a proper selection of the tooth to be analyzed and helps avoid a wrong estimate of body burden.

Some results have been obtained on pig and deer bones (see Table 23). Pigs show a much lower, and deer a higher radium bone content than the average value found in bovine samples. The differences noticed are certainly due to the different diets of the animals, the deer being herbivorous and the pigs, omnivorous. Quite remarkable is the large difference between the Ra contents of the porcine fetus bone ash and that of the mother sow.

Table 23

Ra²²⁶ content of pig and deer bones

Animal identification and location	Type of bone	$\mu\mu\text{g Ra}^{226}/\text{g ash}$
Pig 1 Chicago, Illinois	vertebra	0.036 ± 0.005
Pig 2 Chicago, Illinois	phalanges	0.055 ± 0.006
Pig 3 (pregnant) Chicago, Illinois	vertebra	0.196 ± 0.008
Pig 3a (fetus)	long bone	0.006 ± 0.003
" "	jaw bone	0.006 ± 0.003
" "	teeth	0.005 ± 0.004
Deer 1 ANL site	tibia	0.565 ± 0.07
Deer 2 ANL site	tibia	0.863 ± 0.02

The 25 bovine jaw bones were analyzed also for Po²¹⁰. The determinations of polonium content were performed by plating technique following a nitric-perchloric acid digestion of the samples.⁽⁵⁾ Table 24 lists the Po²¹⁰ concentrations found in the bones at the time of the analyses. The range of the values is quite large and irregular, and without any connection with the Ra²²⁶ content. It is difficult to draw a conclusion with the data available; other studies would be necessary. In some regions, frequent rainfalls that contribute to the deposition of Po²¹⁰ and its parent Pb²¹⁰ on grass could furnish an explanation for the cases of high content.⁽⁶⁾ In Table 24, results obtained from the Th²²⁸ analyses* performed for 21 samples are also shown. Ra²²⁶ contents previously reported are included for the convenience of the reader.

Special thanks are due to Mr. L. D. Marinelli for advice and suggestions during the preparation of this paper. Acknowledgement is made to Janet A. Kann and Frank Markun for the numerous Ra²²⁶ analyses.

*The dibutyl phosphate (DBP) - BaSO₄ procedure⁽⁷⁾ was used for the determination of Th²²⁸.

Table 24
 Po^{210} , Th^{228} , Ra^{226} content of bovine bones

Sample No.	Location and age	Po^{210} $\mu\mu\text{c/g ash}^*$	Th^{228} $\mu\mu\text{c/g ash}^{**}$	Ra^{226} $\mu\mu\text{g/g ash}$	Sample No.	Location and age	Po^{210} $\mu\mu\text{c/g ash}^*$	Th^{228} $\mu\mu\text{c/g ash}^{**}$	Ra^{226} $\mu\mu\text{g/g ash}$
1C	Elk Grove, California 17 y	0.055	analysis not performed	0.185	14C	Charleston, S. Carolina (horse) 9 y	1.06	analysis not performed	0.136
2C	Elk Grove, California 17 y	0.223	analysis not performed	0.191	15C	Columbus, Ohio 2 y	0.438	0.234	0.511
3C	Spokane, Washington 4y 7m	0.606	0.143	0.380	16C	Augusta, Georgia 8 y	0.609	0.179	0.970
4C	Inland Empire, Washington 3 y	0.370	0.105	0.270	17C	Lyndon, Kansas 5 y	2.45	0.110	0.431
5C	Chicago, Illinois 5 y	0.369	0.130	0.395	18C	Lyndon, Kansas 5 y	1.147	0.223	0.483
6C	Amarillo, Texas 3 y	0.667	0.385	0.405	19C	Shawnee, Oklahoma 5 y	0.445	0.147	0.396
7C	Lyndon, Kansas 6 y	1.672	0.189	0.389	20C	Fayetteville, N. Carolina 2 y	0.658	0.431	1.345
8C	White Bear, Minnesota 5 y	1.277	0.135	0.288	21C	Fayetteville, N. Carolina 1 y	0.503	0.489	1.25
9C	Omaha, Nebraska aged cow	0.756	0.207	0.414	22C	Salt Lake City, Utah 10 y	0.608	analysis not performed	0.232
10C	Anthony, New Mexico 6 y	0.489	0.182	0.245	23C	Augusta, Georgia 5 y	0.562	0.777	2.58
11C	Wellston, Oklahoma 6 y	0.673	0.253	0.333	24C	Wapakoneta, Ohio 2y 6m	0.366	0.095	0.526
12C	Glyndon, Minnesota 3y	0.653	0.126	0.331	25C	Zumbro Valley, Minn. 7y	1.02	0.154	0.333
13C	Cogswell, North Dakota 7y	1.115	0.149	0.327					

*Range of estimated error = $\pm 0.01 - 0.04 \mu\mu\text{c/g ash}$.

**Range of estimated error = $\pm 0.009 - 0.02 \mu\mu\text{c/g ash}$.

References

1. E. R. Di Ferrante. Ra^{226} in Bovine Bones and Teeth. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1960. ANL-6199, pp. 88-93.
2. N. L. Garlick. The Teeth of the Ox in Clinical Diagnosis. Am. J. Vet. Research 15 385-394 (1954).
3. W. A. Brown, P. V. Christofferson, M. Massler, and M. B. Weis. Postnatal Tooth Development in Cattle. Am. J. Vet. Research 21 7-34 (1960).
4. C. L. Comar and R. H. Wasserman. Differential Behavior of Substances in Metabolic Pathways. Radioisotopes in the Biosphere. Ed. R. S. Caldecott and L. A. Snyder. Minneapolis: University of Minn. Press, 1960. pp. 526-539.
5. R. B. Holtzman. Analysis of Tissue Samples for Po^{210} . Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1960. ANL-6199, pp. 107-110.
6. C. R. Hill. Pb^{210} and Po^{210} in Grass. Nature 187 211-212 (1960).
7. A. F. Stehney, D. E. Wallace and F. H. Ilcewicz. Isolation of Th^{228} from Bone with Dibutyl Phosphate. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1956. ANL-5596, pp. 12-14.

STABLE LEAD IN HUMAN BONE

Douglas P. Krause

Holtzman^(1,2) has determined Pb^{210} (RaD) in human bone tissue. In connection with his work a program to determine stable lead in some of the same bone specimens has been initiated. Tipton⁽³⁾ has determined stable lead in bone tissue by a spectrographic method, and recently Hill and Jaworowski⁽⁴⁾ have published the results of a study of stable lead and RaD in human bone.

In the work presented here the method of separation and determination of the lead was similar to that used by Hill and Jaworowski.⁽⁴⁾ The separation of the lead was accomplished by a modification of the solvent extraction technique using diethyl dithiocarbamate as described by Maynes and McBryde⁽⁵⁾ in their determination of lead in certain minerals. The separated lead was determined colorimetrically by the dithizone method as detailed by Snyder.⁽⁶⁾

Bone samples in the order of 1 to 2 grams are ashed in platinum at 600°C, then dissolved in 6N hydrochloric acid, diluted to 25 ml volume, and the acidity of the solution is adjusted to 0.9-1.0 M. The lead is extracted with a 1% solution of diethyldithiocarbamate (diethylammonium salt) in chloroform and then partitioned back into an aqueous solution by contact of the organic solvent with 4.5 M hydrochloric acid. This acid solution containing the lead is evaporated to dryness and then fumed with nitric and perchloric acids to remove any organic material. The isolated lead is transferred to a separatory funnel with 10.0 ml of a 1% nitric acid solution. 30.0 ml of an ammonium hydroxide solution containing potassium cyanide and sodium sulfate are added to the nitric acid solution in the separatory funnel. The lead is then extracted as the lead-dithizone complex by shaking the above solution with 10.0 ml of 0.006% dithizone solution in chloroform for one minute and is quantitatively determined by measurement of the absorbance of the chloroform solution containing the lead-dithizone complex at 515 m μ on a Beckman Model DU spectrophotometer. For greater accuracy a reagent blank for each sample is carried through the complete procedure.

The results of the determination of the lead content of 12 specimens of cortical bone are shown in Table 25. The range of the data is from 4.0 μ g Pb/g ash to 59.0 μ g Pb/g ash with an average value of 24.8. Medical histories show 8 cases of osteogenic sarcoma, 3 cases of atherosclerotic gangrene and 1 case of melanoma. In all cases the patients were born and lived in the midwestern states in both rural and urban areas. From the information available, no positive statement can be made regarding occupational exposure to lead; but it has been assumed that none of the cases was so exposed.

Table 25

Sample No.	Bone tissue	Pathology	Age of Donor	Sex	Pb, $\mu\text{g/g ash}$	Pb, $\mu\mu\text{c/g ash}$
280-B	Femur	OS	9	M	4.0	0.075 ± 0.009
283-B	Femur	OS	12	M	7.0	0.420 ± 0.005
539-B	Humerus	OS	17	M	10.8	-
483-B	Femur	OS	27	M	6.3	-
375-B	Femur	OS	30	F	20.6	0.092 ± 0.011
343-B	Femur	AG	42	M	32.0	0.076 ± 0.006
276-B	Talus	AG	48	M	34.8	0.120 ± 0.012
549-B	Femur	OS	56	M	35.0	-
286-B	Tibia	M	61	M	29.6	0.092 ± 0.006
484-B	Humerus	OS	64	M	30.0	0.0 -
413-B	Femur	OS	68	M	29.0	0.100 ± 0.009
341-B	Femur	AG	85	M	59.0	0.079 ± 0.004
Average			43	-	24.8	0.085

OS = Osteogenic sarcoma

AG = Atherosclerotic gangrene

M = Melanoma

Also included in Table 25 are the RaD data obtained by Holtzman on some of the samples.

Table 26 is a summary of the average values for lead obtained by Hill and Jaworowski,⁽⁴⁾ Tipton⁽³⁾ and Kehoe⁽⁷⁾ on various types of bone. Holtzman⁽²⁾ has converted the data of Tipton and Kehoe to an ash basis and these figures are shown in the last column of the table. The data of Hill and Jaworowski have been converted to an ash basis using the factors of 21% ash/g bone for the vertebrae (trabecular) samples and 60% ash/g bone for the tibia (cortical) sample.

Table 26

Investigator	Type of bone sample	No. of samples	Average $\mu\text{g/Pb/g}$ wet base	Average $\mu\text{g/Pb/g}$ bone ash
Tipton	Trabecular	99	6.6	33.0*
Kehoe	Trabecular	6	6.9	32.9*
Kehoe	Cortical	6	20.0	33.3*
Hill and Jaworowski	Trabecular	5	6.6	31.0
Hill and Jaworowski	Cortical	1	4.5	7.5
This report	Cortical	12	-	24.8

*Holtzman⁽²⁾ calculation

As can be seen, the calculated average values for lead on a bone ash basis give remarkably close agreement in the area of about $33 \mu\text{g/g}$ ash as compared to the $25 \mu\text{g/g}$ ash obtained in this work. However, except for the work of Tipton⁽³⁾ in which the range of values found was very wide ($45 \mu\text{g/g}$ wet bone to $0.2 \mu\text{g/g}$ wet bone) only a very small number of samples has been analyzed.

The data in Table 25 suggest a possible correlation between concentration of lead in bone and the age of the donors living in the Midwest. Figure 12 is a linear plot of age versus concentration. Kehoe⁽⁷⁾ found no such relationship. However, his samples were taken from a higher age group, i.e., the average for cortical specimens was 53 years, whereas the average age for the samples shown in Table 25 is 43 years.

Figure 12

Pb concentration in cortical bone as a function of age of the donor. OS, osteogenic sarcoma; AG, atherosclerotic gangrene; M, melanoma.

However, again because of the small number of samples which have been analyzed, no definite conclusions regarding the variation of skeletal lead with time can be drawn until further work has been done.

References

1. R. B. Holtzman. Some Determinations of the RaD and RaF Concentrations in Human Bone. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June 1960. ANL-6199, pp. 94-118.
2. R. B. Holtzman. Critique on the Half-Lives of Lead and RaD in the Human Body. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1960. ANL-6297, pp. 67-80.

3. I. H. Tipton, et al. U.S. Atomic Energy Commission Reports, ORNL-CF-54-12-66; ORNL-CF-56-3-60; ORNL-CF-57-2-2; ORNL-CF-57-2-3; ORNL-CF-59-2-4; ORNL-CF-57-11-33; ORNL-CF-58-10-15; ORNL-CF-59-8-106.
4. C. R. Hill and Z. S. Jaworowski. *Nature* 190 353-354 (1961).
5. A. D. Maynes and W. A. E. McBryde. *Anal. Chem.* 29 1259-1263 (1957).
6. L. J. Snyder. *Ind. Eng. Chem. Anal. Ed.* 19 684-687 (1947).
7. R. A. Kehoe. *Occupational Med.* 3 156 (1947).

FISSION AND NATURAL RADIOACTIVITY IN GROUND LEVEL AIR

Philip F. Gustafson,* Michael A. Kerrigan and S. S. Brar*

Analyses of monthly composites of HV-70 filter paper exposed on Sutorbilt air pumps located on and surrounding the Argonne site have been continued.⁽¹⁾ Weighed portions of filters, exposed to known volumes of air, were partially ashed, compressed into a cylindrical disk, and analyzed by NaI scintillation spectrometry. Quantitative determinations were made of Ce^{144} , Ru^{106} , Cs^{137} , Sb^{125} , W^{181} , Rh^{102} , and Be^7 radioactivity. The latter radionuclide is produced by cosmic ray interactions with atmospheric oxygen and nitrogen and is not a fission product.

In Figure 13 the ground level concentrations of Cs^{137} and Be^7 are indicated; values for Cs^{137} extend from January 1958 through May 1961, and those for Be^7 run from September 1959 through May 1961. Periodic variations in Cs^{137} concentration are readily apparent, maximal levels existing during the spring months. The observance of this phenomenon in 1960 and again in 1961 supports the contention that the "spring maximum" in air-borne fission product concentration and ground deposition rate is attributable to meteorological causes and is not due to periodicity in the testing of nuclear weapons. At the moment, it is not possible to state on the basis of the data shown in Figure 13 whether the 1961 rise has reached its peak; however, it is apparent that the 1961 maximum is as high if not somewhat higher than that observed in 1960.

Figure 13.
 Cs^{137} and Be^7 in surface air

*Division of Biological and Medical Research.

A possible cause of the higher level observed in 1961 will be given presently. The higher concentration of Be^7 seen in the spring months may also be explained on the basis of the mass transfer of stratospheric air into the troposphere during these months. The increased levels of Be^7 present throughout the summer months may arise from the higher production rate occurring in the troposphere during this time. Such increased rate is due to the inclusion of regions of higher Be^7 production within the troposphere during the summer at middle and polar latitudes. These regions, which in summer are part of the higher troposphere, become the lower strata of the stratosphere during the winter time. A discussion of the change in production rate of Be^7 within the troposphere and some of its implications regarding the removal of stratospheric debris is presented elsewhere.⁽²⁾

Cesium-137, as well as other fission products, present in surface air at any given time is the result of nuclear tests conducted prior to the time of observation. By using appropriate radioactive tracers and activity ratios between various fission products, it is possible to resolve the total Cs^{137} present during a given month into that due to specific weapons tests. The starting point in such a procedure is the use of W^{181} , produced by neutron activation in a number of shots in the Hardtack I series, as a tracer for Hardtack debris.⁽¹⁾ In a similar way, Rh^{102} , also produced by neutron activation, in a high altitude shot of the Hardtack series may be used as a tracer for high altitude debris. In Figure 14, the concentrations of W^{181} and Rh^{102} in surface air are shown. The activity values have been corrected for radioactive decay back to the time of production, which were taken to be June 15, 1958 for W^{181} , and August 15, 1958 for Rh^{102} . It should be noted that W^{181} and Rh^{102} also show a spring maximum similar in character to that exhibited by Cs^{137} and Be^7 .

Figure 14.
 W^{181} and Rh^{102} in
surface air.

Activity ratios between $\text{Zr}^{95}/\text{Cs}^{137}$, $\text{Ce}^{144}/\text{Cs}^{137}$, $\text{Ru}^{106}/\text{Cs}^{137}$, $\text{Zr}^{95}/\text{Ce}^{144}$, and $\text{Ce}^{144}/\text{Ru}^{106}$ have been determined for Hardtack, high altitude, Soviet October (1958), Soviet spring (1958), as well as for residual debris. During 1958 and up until early 1960 the $\text{Zr}^{95}/\text{Cs}^{137}$ ratio proved to be the most useful for determining the apparent age of the residual debris.⁽¹⁾ With the decay of Zr^{95} to levels precluding accurate measurements, the $\text{Ce}^{144}/\text{Cs}^{137}$ ratio has been employed, as shown in Figure 15. The $\text{Ce}^{144}/\text{Cs}^{137}$ ratio for Hardtack, high altitude, and the two Soviet series are indicated in Figure 15 by solid lines. The plotted points are the empirical values of the ratio for non-Hardtack, and non-high altitude Ce^{144} and Cs^{137} . During early 1958 the ratio values indicate the presence of predominantly recent debris, presumably arising from the spring Soviet tests. In the summer of 1958, Hardtack I is the dominant source, with lesser amounts of older debris as indicated by the ratio for residual debris. Commencing in November 1958 and continuing through the spring of 1959, the residual activity is mostly due to the Soviet October series. Caution must be exercised, however, because conclusions regarding the source of debris, based upon apparent age determinations, may be quite erroneous. For example, the apparent age during September 1959 is slightly pre-Hardtack for the residual component. This does not preclude the presence of Soviet debris of shorter age, mixed with considerably older material, giving rise to the apparent age observed. A variety of mixtures may all lead to the same apparent time of origin.

Figure 15.
 $\text{Ce}^{144}/\text{Cs}^{137}$ activity ratio.

The various activity ratios previously listed were used in pairs in an attempt further to resolve the residual fraction in surface air. The results of this procedure are shown in Figure 16, where the breakdown of Cs^{137} into that pertaining to Hardtack, high altitude, and Soviet October is shown. The total activity is also shown to provide an indication of the amount of Cs^{137} not accounted for by any of these sources.

Figure 16.

Partition of Cs^{137} in surface air.

Each of these three components indicates a spring maximum in 1960, whereas no high altitude debris was apparent in the spring of 1959 (in fact no Rh^{102} was seen in surface air until September 1959); the other two sources, however, did display maxima in the spring of 1959. A number of observations may be made from further consideration of Figure 16. First is the rapidity with which debris from the Soviet October series diminished during the second and third quarters of 1959. After accounting for the major share of Cs^{137} during the spring months, this source contributed less than 10 percent of the total by the end of the year. A slight peak in debris from this source, observed in the spring of 1960, was less than one percent of

the magnitude seen during the corresponding period in the previous year. The Hardtack series continues to be an important source of activity although there is a decrease in absolute magnitude. Perhaps the most striking feature is the increasing proportion of high altitude debris. The similarity in the rate of increase of high altitude Cs^{137} and total Cs^{137} in surface air during spring 1961 suggests that a larger portion than is indicated in Figure 16 may actually arise from this source. The validity of this suggestion is supported by the $\text{Ce}^{144}/\text{Cs}^{137}$ ratio during March, April, and May 1961 as shown in Figure 15.

The data presented in Figure 16 may also be used to estimate the mean stratospheric residence time for gross debris and for that pertaining to individual test series. In addition to the spring maximum, there is also a period of minimal concentration in surface air which occurs during the fall and winter months. It is presumed that activity seen during this period is a measure of the leakage of debris through the tropopause, occasioned by minor fluctuations in tropopause height, and represents a steady-state condition as opposed to the period of tropopause disruption and mass transfer.

If this be so, then comparison of the concentration of Cs^{137} in surface air between successive minima should provide a measure of stratospheric residence time.⁽²⁾ The mean stratospheric residence time for all tests found by comparing the Cs^{137} levels in the fall 1958 with those in the fall 1959 was 14 months; a mean residence time of 23 months was found between fall 1959 and the fall 1960. The mean residence time of W^{181} -labeled Hardtack debris increased from 8 to 10 months in 1958-59 to in excess of 15 months in 1959-60. Such increases in residence time with time is to be expected as the lower-altitude (and thus more readily removable) debris descends into the lower atmosphere. The mean residence time of Soviet October debris is relatively short, being on the order of 2 to 4 months. It is likely that the removal of the bulk of Soviet October debris by the end of 1959 materially influenced the lengthening of the gross residence time by almost a factor of two. It should also be mentioned that roughly 50 percent of the Cs^{137} found in surface air during the fall of 1960 appeared to be pre-Hardtack in origin, suggesting that the initial estimates of stratospheric residence time of 5 to 10 years based upon measurements of debris from Castle may not have been wide of the mark.

References

1. P. F. Gustafson and M. A. Kerrigan. Argonne National Laboratory, Radiological Physics Division Semiannual Report, January through June 1960. ANL-6199, pp. 62-65.
2. P. F. Gustafson, M. A. Kerrigan and S. S. Brar. A Comparison of Be^7 and Cs^{137} in Ground Level Air. *Nature* 191 454-456 (1961).

MULTIPLE REGRESSION EQUATIONS RELATING METEOROLOGICAL VARIABLES WITH EIGHT-HOUR AVERAGE RADON CONCENTRATIONS

Harry Moses

I. Introduction

In the last decade, meteorologists have given renewed emphasis to the use of statistical techniques for forecasting the magnitude of one meteorological variable from others in the space-time domain. For example, Miller⁽¹⁾ applied the "screening technique" of multiple regression analysis developed by Bryan⁽²⁾ and Efroymsen⁽³⁾ to forecasting pressure, air temperature or dewpoint 24 hours in advance from meteorological measurements at each of 48 stations over the United States. Weather studies using other statistical techniques, such as orthogonal polynomials⁽⁴⁾ or multiple discriminant analysis,⁽⁵⁾ have been conducted with a certain degree of success at the Massachusetts Institute of Technology and the Travelers Weather Research Center. In this study, the "screening technique" was used, not to forecast, but to relate the magnitude of observed eight-hour average radon concentrations with simultaneous values of weather elements occurring in the immediate vicinity.

The concentration of radon a short distance above the ground is a function of: 1) the rate of vertical diffusion, 2) horizontal advection of radon from upwind locations, 3) the radioactive decay rate, and 4) the emanation rate from the soil surface. Of these, the decay rate of radon with a half-life of 3.825 days is known, and the emanation rate apparently varies but little except for times when rain or snow inhibit its flow. The vertical diffusion varies greatly from time to time and is of prime importance in controlling the magnitude of radon in the lowest layers of the atmosphere. Of lesser importance, but by no means negligible, are the changes brought about by advection. This investigation concerns itself essentially with the effect of vertical diffusion on radon concentration, but account is also taken of the effect of precipitation. Multiple regression equations are developed relating radon concentrations with meteorological variables pertinent to the diffusion process.

II. Experimental Technique

Average values of radon concentration were obtained for consecutive eight-hour periods beginning at 0845 on Monday and ending at 1645 on Friday of each week from August 20, 1956 through September 30, 1957. These measurements were made under the supervision of A. F. Stehney and H. F. Lucas, Jr. of the Radiological Physics Division of the Argonne National Laboratory.

The intake duct through which the samples were obtained was located 3 feet above ground and 1 foot from the east wall of a one-story wing of Building 203. From the intake duct the air sample passed through a Drierite-Ascarite column to remove water and carbon dioxide, and, in addition, through two dry-ice-cooled dip-type freeze traps to remove the final traces of water vapor. The sample then passed through a tube containing 7 grams of dry-ice-cooled activated coconut charcoal on which the radon was adsorbed. A zinc sulphide scintillation counting chamber developed by Lucas⁽⁶⁾ and a Dumont 6292 photomultiplier were employed for making the radon concentration determinations.

III. Meteorological Data

The field site⁽⁷⁾ at which the meteorological measurements were made is located about one mile southwest of the point at which the radon samples were taken. Only those variables which have a bearing on the rate of vertical diffusion and consequently on radon concentrations were selected. These are: 1) wind speed at 19 feet, 2) wind speed at 150 feet, 3) the ratio of the 150-foot to the 19-foot wind speed, 4) ambient air temperature at 5 $\frac{1}{2}$ feet, 5) relative humidity at 6 feet, 6) station pressure at 746 feet above sea level, 7) atmospheric stability as indicated by the difference between temperature measurements at 144 and 5.5 feet, 8) intensity of solar radiation on a horizontal surface, 9) soil temperature at 1 cm below the surface, 10) difference between ambient temperature and 1-cm soil temperature, and 11) the water-vapor pressure at 6 feet. The meteorological data were averaged for the consecutive eight-hour periods: 0100 to 0900, 0900 to 1700, and 1700 to 0100, to correspond to the periods during which the radon measurements were made.

An examination of the radon measurements shows that when the ground is wet, the amount of radon in the air is smaller, no doubt, because of the clogging of the soil pores or capillary passages through which the radon emerges, and, to a lesser extent, the solution of radon in the added soil water. For this reason, the data were stratified in accordance with whether the ground was wet or dry. At the present point in this study, it is not clear how best to stratify the data. For example, the same amount of rainfall during the warm part of the year may be far less effective than during the cold part of the year because the difference in the evaporation rate is appreciable. For the first part of the study (Table 27) the data were separated in accordance with the following criterion: the ground was considered dry if less than 0.05 inches of precipitation fell within the 24 hours preceding the end of the current eight-hour period; the ground was considered wet if 0.05 or more inches of rain fell during this period. In the second part of the study, Tables 28 and 29, the criterion was changed to the following: the ground was considered dry if less than 0.03 inches fell during the 24-hour period and wet if 0.10 or more inches of precipitation occurred. With the latter criterion, there is a set of data with precipitation from 0.03 to 0.09 inches which were not considered in the studies.

Table 27

Regression coefficients based on data for total period

August 20, 1956 - September 30, 1957

Based on data taken Monday, Tuesday, Thursday and Friday															
Period	Ground condition	Meteorological variables												No. of cases	Percent of explained variance (R^2)
		Constant	U_{19}^*	$\frac{U_{150}}{U_{19}}$	T_a	RH	P	ΔT	S	T_s	$T_a - T_s$	U_{150}	e		
All three 8-hour periods	Wet and dry	4.440	-0.011				-0.140	0.076						544	0.34
	Dry	5.799			-0.018	0.004	-0.195	0.093	0.0002	0.016	0.026	-0.006		411	0.41
	Wet	0.114		0.127				0.055				-0.006		133	0.36
0045 - 0845 C.S.T.	Wet and dry	0.393	-0.014					0.150	-0.004	0.008				159	0.60
	Dry	9.849	-0.017				0.322	0.136	-0.004	0.009				115	0.73
	Wet	0.153	-0.011	0.167				0.135	-0.003					44	0.64
0845 - 1645 C.S.T.	Wet and dry	0.398	-0.049					0.040	-0.0002		0.006	0.030		219	0.15
	Dry	0.397			0.004			0.105	-0.0004					165	0.20
	Wet	0.329	-0.057									0.034		54	0.19
1645 - 0045 C.S.T.	Wet and dry	4.935	-0.007	0.029			-0.161	0.059	-0.004				0.003	166	0.48
	Dry	8.269		0.025			-0.273	0.059	-0.004			0.006	0.005	131	0.53
	Wet	-0.081		0.160										35	0.38

* For meaning of symbols see Section IV.

Table 28

Regression coefficients based on data taken during warm part of year

August 20, 1956 - September 30, 1956;
April 1, 1957 - September 30, 1957

Based on data taken Monday, Tuesday, Thursday and Friday															Based on data taken Wednesday		
Period	Ground condition	Meteorological variables												No. of cases	Percent of explained variance (R ²)	No. of cases	Percent of explained variance (R ²)
		Constant	U ₁₉	$\frac{U_{150}}{U_{19}}$	T _a	RH	P	ΔT	S	T _s	T _a -T _s	U ₁₅₀	e				
All three 8-hour periods	Wet and dry	-0.009	-0.007	0.038		0.001		0.042		0.007				307	0.37	92	0.39
	Dry	4.746		0.035	0.005	0.003	-0.165	0.047						225	0.38	65	0.42
	Wet	0.261	-0.019		0.005									59	0.27	20	0.25
0045 - 0845 C.S.T.	Wet and dry	0.199			0.005			0.166						118	0.51	32	0.75
	Dry	0.316						0.154						82	0.50	26	0.76
	Wet	0.315	-0.025		0.005									24	0.59	6	0.02
0845 - 1645 C.S.T.	Wet and dry	0.053			0.008			0.031						99	0.26	30	0.20
	Dry	0.034			0.009			0.035						78	0.32	20	0.21
	Wet	No significant coefficients - Mean radon concentrations = 0.18μC/liter												16	-	-	-
1645 - 0045 C.S.T.	Wet and dry	0.290	-0.034	0.042						0.010				90	0.50	30	0.62
	Dry	0.184	-0.047			0.004				0.010				65	0.49	19	0.65
	Wet	0.435	-0.031											19	0.16	7	0.32

Table 29
Regression coefficients based on data taken during cold part of year
October 1, 1956 - March 31, 1957

Based on data taken Monday, Tuesday, Thursday and Friday															Based on data taken Wednesday		
Period	Ground condition	Meteorological variables												No. of cases	Percent of explained variance (R ²)	No. of cases	Percent of explained variance (R ²)
		Constant	U ₁₉	$\frac{U_{150}}{U_{19}}$	T _a	RH	P	ΔT	S	T _s	T _a -T _s	U ₁₅₀	e				
All three 8-hour periods	Wet and dry	0.267	-0.010	-0.049		0.002		0.109		0.010				239	0.47	74	0.40
	Dry	0.044			0.007	0.004		0.094						181	0.44	60	0.53
	Wet	-0.241		0.301	0.006									40	0.50	5	-1.07
0045 - 0845 C. S.T.	Wet and dry	0.606	-0.016	-0.095	0.006			0.203						71	0.66	23	0.41
	Dry	-0.106				0.004		0.198		0.014				58	0.67	19	0.71
	Wet	No significant coefficients - Mean radon concentrations = 0.23 μμC/liter												12	-	-	-
0845 - 1645 C. S.T.	Wet and dry	0.126	-0.015	0.274				0.175		0.009				95	0.43	26	0.13
	Dry	0.267				0.002		0.259		0.011				73	0.47	21	0.46
	Wet	0.162			0.006									14	0.19	1	-
1645 - 0045 C. S.T.	Wet and dry	0.072		0.115										73	0.35	25	-0.29
	Dry	0.120		0.100										53	0.30	20	-0.53
	Wet	5.179					-0.173	0.186		0.008				14	0.85	3	-6.18

Use of solar radiation intensity as a variable should be made with caution since the first and third eight-hour periods, i.e. 0045 to 0845 and 1645 to 0045, are partly in darkness. For this reason, solar radiation intensity was omitted as a variable in these periods during the second portion of the study; however, it was included in the second period (0845 to 1645) which is usually entirely during daylight.

The data were further stratified in accordance with the warm and cold seasons of the year. The warm portion of the year was considered as the period from April 1 to September 30 and the cold part as the remainder of the year - from October 1 to March 31. Tables 28 and 29 show the number of measurements in each grouping for which multiple regression equations were calculated. It is to be noted that regression equations were also calculated for combinations of groups, such as, the combination of wet and dry conditions or the group in which the information from the first, second and third periods were treated together.

IV. Statistical Analysis

A linear multiple regression equation such as

$$Y = \text{Constant} + b_1 U_{19} + b_2 U_{150} + b_3 \frac{U_{150}}{U_{19}} + b_4 T_a + b_5 (\text{RH}) + b_6 P + b_7 (\Delta T) + b_8 S + b_9 T_s + b_{10} (T_a - T_s) + b_{11} e$$

provides a means for calculating the magnitude of radon concentration from pertinent meteorological variables. The symbols used in the above equation and those appearing in Tables 27 to 30 are:

Y	radon concentration in $\mu\mu\text{c/liter}$
b_i	where $i = 1$ to 11 are the regression coefficients
U_{19}	wind speed in miles per hour at 19 feet above ground
U_{150}	wind speed in miles per hour at 150 feet above ground
U_{150}/U_{19}	ratio of 150-foot to 19-foot wind speed
T_a	ambient air temperature in degrees Centigrade
RH	ambient relative humidity in percent
P	atmospheric pressure in inches of mercury
ΔT	temperature difference (144- minus 5.5-foot levels) in degrees Centigrade
S	intensity of solar radiation on a horizontal surface in langleys per minute
T_s	soil temperature in degrees Centigrade at one centimeter below the ground surface
$T_a - T_s$	difference between air and 1-cm soil temperature in degrees Centigrade
e	vapor pressure in millibars

In Tables 27 to 30 only those variables which contributed significantly to the estimate are included since the "screening technique" eliminates the others. The data taken on Wednesday were not used in calculating the regression equations but were reserved as a relatively independent set of measurements to test the equations.

An indication of the effectiveness of the regression equations may be obtained from the "estimated" multiple correlation coefficients shown in Tables 27 to 29. The multiple correlation coefficient, $R_{Y, 1 \ 2 \dots n}$ provides a measure of how well the equation fits the data since

$$R_{Y, 1 \ 2 \dots n}^2 = 1 - \frac{S_{Y, 1 \ 2 \dots n}^2}{\sigma_Y^2}$$

where $S_{Y, 1 \ 2 \dots n}^2$ is the mean square of the deviations of the observed radon concentrations from the hyperplane in a space of the same number of dimensions as independent variables and σ_Y^2 is the mean square of the deviations about the average of radon concentration measurements. The "estimated" $R_{Y, 1 \ 2 \dots n}$ incorporates a correction for sampling error and is calculated from the expression

$$R_{Y \cdot 1 \ 2 \dots n}^2 = 1 - \left(\frac{S_{Y \cdot 1 \ 2 \dots n}^2}{\sigma_Y^2} \right) \frac{N-1}{N-n}$$

where N is the number of cases and n is one plus the number of independent variables used in the regression equation. When N is large and n is small, the correction is negligible.

Tables 28 and 29 show how effectively the regression equations predict radon concentrations from the independent set of data, i.e., those taken on Wednesday

$$R_{Y_w \cdot 1 \ 2 \dots n}^2 = 1 - \frac{\frac{\sum (Y_w [\text{observed}] - Y [\text{calculated}])^2}{N}}{\frac{\sum (Y_w [\text{observed}] - \bar{Y}_w)^2}{N}}$$

$Y_w [\text{observed}]$	radon values observed on Wednesday
$Y [\text{calculated}]$	radon values calculated from the regression equations
$\bar{Y}_w$	average of observed Wednesday radon values for the grouping under consideration.

The screening technique of multiple regression analysis used in this study is based on a SHARE program for the IBM 704 computer written by Efroymsen and is similar to the technique proposed by Bryan⁽²⁾ and adapted to meteorological problems by Miller.⁽¹⁾ In carrying out the procedure, the zero order correlation coefficients between the dependent and each of the independent variables are computed, and the independent variable having the largest absolute value of the correlation coefficient is selected. The linear regression equation with this selected independent variable is formed, and the Fisher F-test at the 5% level with 1 and $N-2$ degrees of freedom used to test significance. The computed F value

$$F_{\text{computed}} = \frac{\text{Explained variance}}{\text{Unexplained variance}} = \frac{\frac{1}{N} \sum (Y [\text{calculated}] - \bar{Y})^2}{\frac{1}{N} \sum (Y [\text{observed}] - Y [\text{calculated}])^2}$$

is compared with the Fisher F value from statistical tables. If

$$F_{\text{computed}} > F_{\text{table}}$$

the variable is retained, otherwise, the process is terminated.

In the second step the partial correlation coefficients between the dependent variable and each of the remaining unselected variables, holding the selected one constant, are calculated, and the variable with the largest coefficient selected as the second variable. The increase in explained variance is compared with the unexplained variance and an F test with 1 and $N-3$ degrees of freedom used to determine if the second selected variable is significant.

The process is continued by calculating the partial correlation coefficients between the dependent and each of the unselected independent variables, holding all of the previously selected variables constant, and selecting the variable with the largest coefficient. When a step is reached in which the F test with 1 and $N-n-1$ degrees of freedom shows that the selected variable is insignificant, the regression equation consisting of the significant variables is formed and the procedure is stopped. N is the number of cases and n is the number of independent variables selected.

In this preliminary study, some of the refinements suggested by Efroymson and by Miller were not used; for example, Miller indicates that in using the F test the 5% significance must be modified in accordance with the number of independent variables used. As a result, our test for significance is somewhat less stringent than the 5% level. Actually this is not too serious since in most all of our cases, the computed F values were appreciably higher than the critical tabular values. Further, Efroymson indicates if all of the remaining variables in a given step of the screening technique are insignificant, a combination of variables may be significant and suggests steps for carrying out this calculation. These improvements will be incorporated in an extension of this work.

V. Discussion of Results

It is of interest to point out that in Table 27 where the data for the warm and cold parts of the year are considered together, stability (ΔT) or the wind speed ratio, which is strongly correlated with stability, appears as a significant variable in nearly all of the equations. Throughout this study, wind speed and a measure of the air or soil temperature enter into the equations quite often, but pressure quite infrequently.

Tables 27 to 29 indicate that the regression equations are most reliable during the first period (0045 - 0845) and less reliable during the second (0845 - 1645). Because of the paucity of data, the equations are least satisfactory when the ground is wet.

When no independent variable is significant, one may use the average value as the best estimate of the radon concentration. After carrying out the experimental calculations presented in this report, a number of improvements suggest themselves. For example, the amount of radon present in

one period appears to be highly correlated with the amount appearing in a succeeding period. It is therefore desirable that radon concentration values measured in the preceding eight hours be considered as one of the independent variables. Further, no separation was made between cloudy and sunny days, which represent two distinct meteorological regimes; stratification in accordance with this criterion should be tested. Some of the low values of explained variance may be due to the relatively long averaging period of eight hours. During an eight-hour interval, marked changes may occur, especially during some parts of the year, i.e. half the period may be in sunlight and under highly diffusive conditions, whereas, the other half may be under a strong inversion. The advection of radon from an upwind source, no doubt, also influences the effectiveness of the equations. This difficulty may be alleviated by taking wind direction into account. Again, an average direction can be meaningless, especially if the wind blew 4 hours from the south and the following 4 hours from the north. Caution must, therefore, be exercised to use only data based on wind directions which are uniform throughout the eight-hour averaging period.

VI. Conclusions

In spite of the difficulties presented in the previous paragraphs, the equations represent a practical means for estimating radon concentrations based on meteorological information. The improvement over using a mean value is indicated by the ratio of explained variance to total variance or the square of the multiple correlation coefficient - the coefficient of determination (see Tables 27 to 29). Regression analysis is a tool which may be used not only for studying the relation between radon concentrations and weather elements, but for investigating the relations between the meteorological variables themselves.

The help of Mr. Willard G. Greenhow of the Applied Mathematics Division in programming this problem on the IBM 704 is gratefully acknowledged. Thanks are also due to Mr. Frank C. Kulhanek for his assistance in carrying out some of the computations.

APPENDIX A

The following regression equations shown in Tables 30a and 30b were excerpted from Tables 28 and 29 and may serve as a practical means for estimating the eight-hour average radon concentrations:

Table 30a

For data from August 20, 1956 through September 30, 1956
and April 1, 1957 through September 30, 1957

Warm part of year

Period	Ground Condition	Regression Equation
0045 - 0845	Dry	$Y = 0.316 + 0.154 \Delta T$
0045 - 0845	Wet	$Y = 0.315 - 0.025 U_{19} + 0.005 T_a$
0845 - 1645	Dry	$Y = 0.034 + 0.009 T_a + 0.035 \Delta T$
0845 - 1645	Wet	Use mean value $\bar{Y} = 0.18 \mu\mu c/l$
1645 - 0045	Dry	$Y = 0.184 - 0.047 U_{19} + 0.004 RH + 0.010 T_s$
1645 - 0045	Wet	$Y = 0.435 - 0.031 U_{19}$

Table 30b

For data from October 1, 1956 through March 31, 1957

Cold part of year

Period	Ground Condition	Regression Equation
0045 - 0845	Dry	$Y = -0.106 + 0.00428 RH + 0.198 \Delta T + 0.0142 T_s$
0045 - 0845	Wet	Use mean value $\bar{Y} = 0.231 \mu\mu c/l$
0845 - 1645	Dry	$Y = 0.267 + 0.002 RH + 0.259 \Delta T + 0.011 T_s$
0845 - 1645	Wet	$Y = 0.162 + 0.006 T_a$
1645 - 0045	Dry	$Y = 0.120 + 0.100 U_{150}/U_{19}$
1645 - 0045	Wet	$Y = 5.179 - 0.173 P + 0.186 \Delta T + 0.008 T_s$

References

1. R. G. Miller. Statistics and the predictability of weather. Studies in Statistical Weather Prediction, Final Report, AFCRC-TR-58-272. Ed. B. Shorr; Project Director, T. F. Malone. Travelers Weather Research Center, Hartford, Conn., 1956. pp. 137-153.
2. J. G. Bryan. Special techniques in multiple regression. Unpublished manuscript, 1944.
3. M. A. Efroymson. Stepwise procedure for calculation of multiple regression, presented at the Gordon Research Conference on Statistics, Aug. 8-12, 1955.
4. E. N. Lorenz. Prospects for statistical weather forecasting. AFCRC-TR-59-224, Massachusetts Institute of Technology, Cambridge, Mass., 1959.
5. R. G. Miller. Selecting variates for multiple discriminant analysis. Final Report, AFCRC-TR-60-254, The Travelers Weather Research Center, Hartford, Conn., 1960.
6. H. F. Lucas, Jr., Improved low-level alpha scintillation counter for radon. Rev. Sci. Instr. 28, 680-683 (1957).
7. H. Moses, and J. H. Willet. 1957: Five-Year climatological summary, July 1949 - June 1954, Argonne National Laboratory, DuPage County, Illinois. Argonne National Laboratory Report ANL-5592.

THE ARGONNE METEOROLOGICAL MODEL TOWING TANK; TWO-DIMENSIONAL JETS IN A STRATIFIED FLUID

Paul Frenzen

Introduction

Studies of the behavior of streams of fluid injected into larger volumes of similar fluid initially at rest (circulation models sometimes referred to as "submerged jets") have contributed significantly to the available knowledge of free turbulent flows, that is of fields of turbulence not influenced by the presence of near-by boundary surfaces. That such circulations are of practical as well as theoretical interest is demonstrated by the existence of prototype examples ranging from the mixing of small quantities of one ingredient injected into another to the flow of planetary jet streams which circle the earth within both the atmosphere and the ocean. Intermediate in this scale range, the injection of stack plumes into the lower layers of the atmosphere is one phase of the problem which has been the subject of an extensive field investigation at this laboratory.⁽¹⁾

In recent years a number of theoretical and experimental studies have concerned themselves with the effects exerted upon these flows by stable density gradients.⁽²⁻⁴⁾ This particular aspect of the problem is of special importance to geophysical circulations since in nature, large bodies of fluid generally are stably stratified except for local, temporary departures near their boundaries. Thus, the average decrease of density with height in the free atmosphere is only about two thirds of that corresponding to neutral equilibrium; in other words, the average lapse rate is about two thirds that of the dry adiabatic. Similarly, the thermohaline structure of the ocean is such that the density of sea water normally increases with depth. These facts have given rise to the widely-accepted impression that the occurrence of narrow, jet-like currents as prominent features in the large-scale circulations of the sea and the air is, at least in part, due to the presence of stably stratified fluid layers. Indeed a number of theoretical explanations for this association have been advanced,⁽⁵⁻⁷⁾ but no complete solution exists.

In the Argonne towing tank horizontal jets have been observed to form in the later stages of turbulent decay within stably stratified water in the lee of a moving, rectangular grid, once a critical value of a shear-stability ratio is attained.⁽⁸⁾ The current report summarizes the results of a series of experiments designed to examine further the apparent dependence of these stratified jet flows upon this ratio, the "internal" Froude number.

Theoretical Remarks

When the density of a fluid is permitted to vary, the field of motion is affected in two ways, for local variations in the mass per unit volume contribute proportional variations to both the inertial and the gravitational (or buoyant) accelerations undergone by the fluid parcels. Theoretical studies have found that critical values of a nondimensional parameter describing the relative magnitudes of these two effects are closely associated with characteristic changes in the properties of analytical solutions.^(3,4,6,7) In the laboratory, experimental studies of the flow of stratified fluids^(2,3,8) have verified similar associations between changes in circulation patterns and values of this ratio, a generalized form of the Froude number.

The ratio may be written by first noting that in the interior of a nonhomogeneous fluid ($d\rho/dz \neq 0$) the acceleration of gravity is, in effect, reduced in proportion to the vertical percentage change of density by the effects of buoyancy. One may write

$$g^* = g(\Delta \rho / \bar{\rho})$$

where g^* is often termed the "reduced" gravity. Then a representative ratio of inertia force to reduced gravitational force within such fluids can be written:

$$\frac{\bar{\rho} u^2}{L} / \bar{\rho} g^* = \frac{\bar{\rho} u^2}{g \Delta \rho L} = \frac{\bar{\rho} (u/L)^2}{g (d\rho/dz)} = Fi^2 \quad (1)$$

where u and L are representative scales of velocity and length, respectively, and Fi is the internal Froude number. It may be noted in passing that the expression above is equivalent to the reciprocal of the meteorologically more familiar parameter, the Richardson number, i.e., $Fi^2 = 1/Ri$.

When the velocity, u , is small enough and/or the stable gradient is strong enough (in other words, when Fi is smaller than some critical value), the inertia forces are negligible in comparison with those of gravity; the flow is then dominated by the latter effects. As a result, vertical motions in opposition to the stabilizing buoyancy forces are damped, and fluid disturbances tend to be confined to their level of origin.

For stratified flows with $Fi > 1/\pi$, solutions of the equations of motion have been found which correspond to internal waves; these have been verified by Long's experimental observations of stratified flow over barriers.⁽³⁾ Yih⁽⁴⁾ gives an analytical solution for $Fi < 1/\pi$ for the case of stratified flow into a line sink which also indicates waves, but he points out that in experiments by Debler⁽²⁾ no such waves were observed. Instead, the fluid developed a discontinuity between a stagnant and moving

layer, the dimensions and velocity of the latter determining an internal Froude number, $Fi \approx 0.28$, a fair approximation to the theoretical critical value of $1/\pi = 0.32$. Moreover, the observed flow pattern closely resembled an alternative non-wave solution to the line sink case given earlier by Yih.⁽⁹⁾ In both theoretical treatment and experiment, these were steady, two-dimensional, laminar flows.

Rossby⁽⁷⁾ and concurrently Craya⁽⁶⁾ have discussed the vertical concentration of momentum from another point of view. Their analyses indicate that jet-like velocity profiles will form in stably-stratified, fully-developed turbulent flow in open channels under conditions of constant volume transport and minimum transfer of momentum; Craya extended the treatment to the case of minimum energy flux. In both cases, initially uniform horizontal velocity distributions along the vertical are shown to concentrate into jet profiles because of momentum losses at the boundary in the one case and energy losses throughout the cross section of the fluid in the other. Again in both cases, the development of these profiles tends toward critical conditions of speed and velocity that are specified by critical values of the internal Froude number.

With an analytical model consisting of a moving, stably-stratified layer bounded above and below by stagnant, neutrally-stratified regions, Rossby found that the minimum momentum transfer principle predicted the ultimate creation of a jet profile whose dimensions would adjust to a critical internal Froude number, $Fi = 1/\sqrt{24} \sim 0.20$. With the energy principle, Craya found a slightly different value, $Fi \sim 1/\sqrt{17} \sim 0.24$.

Thus it is seen that there are both theoretical reasons and experimental precedents for the observation of two dimensional jets in the interior of stably-stratified fluids, jets whose vertical dimensions and maximum velocities might be expected to adjust to critical values of the internal Froude number. Surprisingly, this seems to apply equally well to both laminar and turbulent flows in a quantitative as well as a qualitative sense, since the critical values of the internal Froude number for either case lie in the relatively narrow range $0.2 < Fi < 0.3$.

Apparatus and Procedure

For this series of experiments the towing-carriage and suspended-grid assembly of the Argonne tank were removed, and the slit source and sink arrangements shown schematically in Figure 17 were installed. The tank (9.15 m long, 66 cm wide, and filled to a depth of 36 cm for these tests) is well suited to these trials since it has been designed to provide the necessary temperature control. The lower boundary contains a full-length heat exchanger through which water of a desired temperature can be continuously circulated, while the upper boundary covering 95% of the free surface effectively restricts evaporation cooling and supplies uniform heat

when desired by means of an array of electrical resistance cables. In addition, the temperature of the laboratory space itself can be held within close limits: during the critical, neutral and near-neutral stratification trials, the free air temperature remained within half a degree C of the mean temperature of the water in the tank.

Figure 17.

Schematic representation of slit source and sink apparatus installed in the Argonne towing tank.

Vertical temperature gradients were measured with a twelve-junction, copper-constantan thermopile which was mounted in the center of the tank, just downstream from the test section (Figure 17). For each degree C difference in temperature impressed across its 10-cm length, this device supplied 240 microvolts to a Brown self-balancing, recording potentiometer. Regular calibration checks made during the course of the experimental program resulted in gradient measurements which were reproducible within plus or minus 0.01 deg cm^{-1} .

The slit source was constructed by cementing two pieces of Lucite ($26" \times 10" \times 3/8"$) together with two thin strips of the same material to make a narrow box, open along its two longer sides. Water introduced between the Lucite plates through a perforated pipe sealed tightly along one of the assembly's longer sides was passed through a fine mesh stilling screen before it issued from the opposite, open side of the source. The device generated an approximately two-dimensional turbulent jet, initially 1 cm deep, that extended very nearly across the full width of the tank. The slit sink was simply a flattened funnel fashioned of sheet aluminum. It appeared that its shape was not at all critical; it was only necessary that the intake be positioned at the same central level as the source in order that water of the proper temperature could be continuously circulated.

The rate of flow was controlled by means of a large needle valve and measured with a rotameter, as shown in Figure 17. Flow variations observed in the system throughout the one-hour long experimental runs were negligible.

Observations were made by photographing the horizontal displacement of vertical dye streaks which had been placed in the center of the tank's test section by dropping minute, soluble crystals of fluorescein dye through a small port in the transparent overhead lid. The camera shutter remained open long enough for two flashes from a high speed, electronic flash unit that were triggered in succession by an accurate timing device. Thus, from its position at the side of the tank 92 cm from the glass wall, the 35-mm camera recorded two successive positions of the moving dye streak. From the displacements of the two images relative to one another on the same negative (see Figure 18), the known time interval and the scale factors, the average maximum velocity (u) and the vertical width (L) of the principal jet were readily obtained.

In each run photos were taken at three-minute intervals during the first quarter of an hour, five minutes apart for the next 25 minutes, and ten minutes apart during the final 20 minutes. Measurements were made from images of the 35-mm negatives optically enlarged to about half the size of the original experimental region. The maximum horizontal deflection along the axis of the principal jet was measured first, this giving the velocity term, u . Then two points above and below, where the dye streak had moved just half as far, were located (see inset, Figure 18). Twice the vertical distance between these levels was recorded for the scale L .

Experimental Observations

These experiments were suggested not only by the appearance of jet profiles in the stratified fluid-turbulence experiments mentioned previously, but also by some unpublished observations of John Francis, Imperial College of Science and Engineering, London, which were brought to our attention by Professor D. Fultz of The University of Chicago (consultant to the Argonne Meteorology Group). Francis set up a jet pipe directed along the central axis of a channel containing water at rest. He found that although the jet leaving the pipe was fully turbulent and that, therefore, ink placed in the region extending up to several feet from the orifice was rapidly mixed, there could be observed after a few moments "a long V-shaped tongue or wedge of dyed water" projecting slowly downstream from the turbulent area.⁽¹⁰⁾ Above and below this wedge, the water remained clear. Surprisingly, Francis was of the opinion that the water in his tank was not stably stratified.

The first trial runs in the Argonne tank duplicated the Francis' arrangement in order to test this particular point. As will be seen in the more detailed discussion of the slit-source experiments below, Francis' wedge (actually the leading edge of a plane jet) was not observed to form in a fluid with a truly neutral (isothermal) stratification, but it did appear when the fluid in the tank was stable.

Figure 18

An example of photographs taken in two-dimensional jet experiments; inset sketch shows details of measurements made for u and L . Conditions: $d\theta/dz = 0.24 \text{ deg cm}^{-1}$; $Q = 168 \text{ cc/sec}$; $t = 20 \text{ min}$ after start; $\Delta t = 20 \text{ sec}$; distance between vertical reference marks $\approx 10 \text{ cm}$; $F_1 = 0.21$ (on decreasing trend after attaining $F_{1\text{crit}}$, see Figure 20c).

Since under suitable conditions large bodies of fluid often tend to assume stable density distributions when left undisturbed for long periods of time (Francis generally allowed 24 hours to elapse after filling the tank and before starting the jet circulation), it seems likely that these early "V-wedge" observations actually were made under weakly stratified conditions.

The Argonne experiments were carried out over a range of flow rates and in a variety of fluid stability conditions. The results fall into a number of categories that may best be summarized in the internal Froude number trend curves of Figures 19 and 20.

As mentioned above, no patterns of flow clearly resembling plane jets were observed when the density stratification was very nearly neutral ($d\theta/dz = 0 \pm 0.01/\text{cm}$); turbulence introduced by the slit source was instead propagated downstream in a divergent jet. If the vertical extent of the tank had been greater, it was evident that the diffusing jet at any section would have grown to a thickness directly proportional to its distance from the orifice, as predicted by mixing-length considerations and verified experimentally under neutral conditions (cf. Reference 11, Ch. 23).

Figure 19.

Internal Froude number trends under conditions of moderate stability ($d\theta/dz \approx 0.1$ to 0.2 deg cm^{-1}) and increasing flow rate; figures at ends of curves are flows in $\text{cm}^3 \text{ sec}^{-1}$.

Figure 20

Internal Froude number trends for several conditions of stability and flow rate. Note tendency for curves to change from overdamped to underdamped (oscillatory response) when mean Froude number exceeds $Fi_{crit} \approx 0.3$. Figures at ends of curves are average vertical temperature gradients in deg cm^{-1} .

However, when the experiment is carried out in stably-stratified fluid, the restriction of the flow issuing from the slit source to the central levels of the tank is quite marked. Secondary circulations that soon appear both above and below the central jet (Figure 18) evidence some vertical transfer of momentum. But these remain essentially closed circulations, isolated respectively in the upper and lower portions of the tank. On several occasions a cloudy precipitate which had formed in the pump and its associated lines overnight remained in the principal jet after several hours of continuous recirculation through the system; the frictionally driven secondary loops above and below remained virtually clear.

The development and trends of characteristic internal Froude numbers for the central jet under moderately stable conditions ($0.1 \text{ deg cm}^{-1} \leq d\theta/dz < 0.2 \text{ deg cm}^{-1}$) and in response to several rates of flow from the slit source are shown in Figure 19. Note that the weakest flow ($Q = 84 \text{ cc/sec}$) results in what might be termed an "over damped oscillation" response; the Fi value rises to about 0.2 and then remains approximately constant. An increase in flow rate to 126 cc/sec results in a curve roughly resembling the response of a system with near-optimal damping. After a rapid rise to a peak overshoot and one or two damped swings, the Fi value settles down to an average value just under 0.3. Further increases in flow rate ($Q = 168, 227$ and 265 cc/s) result in underdamped oscillatory behavior in which Fi swings around average values in excess of 0.4.

Figure 20, a through d, shows a number of experimental runs with similar flow rates. Here, the damping effect of increasing stability may be seen in each set, each individual curve originating from a different stability condition. There is some scatter in this relation; curves for nominally more stable conditions sometimes exhibit larger average values of Fi than do members of the same set corresponding to lesser stabilities. However, as may be seen in Figure 20a ($Q = 84 \text{ cc/sec}$), the general trend is in the expected direction.

A most interesting trend in these curves, one which may not be immediately apparent to the reader's eye, concerns the tendency for the "critically-damped" condition to occur when Fi is in the range $0.26 < Fi < 0.34$. In order to show this effect more readily we note first that the curves generally exhibit a monotonic trend downward with time, probably because of the continuing loss of momentum to the secondary circulations which continued to grow throughout the period of observation. This trend in Fi is only partially offset by a similar steady decrease in the lapse rate resulting from limited vertical mixing that takes place across the central jet.

Using only those data taken at equal intervals of five minutes (usually the measurements made between five and twenty minutes after the circulating pump had started), linear regression lines for Fi on t

were computed for the decreasing Froude number trends. Standard errors of estimate (σ_e) were then calculated for the departures of the observed Fi values from these straight lines, the object being to employ the square of these statistics as measures of the degree each curve oscillated around its general trend.

Figure 21 plots σ_e^2 obtained from these 15-minute segments of each experimental run against the average internal Froude number maintained by the principal jet during the period of observation.

Figure 21

Variance (standard error of estimate squared) of the departures of observed Fi values from linear regression lines fitted to trend curves of Figure 20 between 15 and 40 minutes after pump start. Note increase in variance observed in cases with $Fi > Fi_{crit} \approx 0.3$.

Note the pronounced tendency for this variance estimate to remain small until the mean Froude number has exceeded some value in the range $0.26 < \overline{Fi} < 0.34$; after passing through this zone of transition, σ_e^2 increases rapidly. In other words, as long as $\overline{Fi} < Fi_{crit} \approx 0.3$, the $Fi(t)$ trend curves rise asymptotically to some near-constant, subcritical level, somewhat in the manner of the response characteristic of an overdamped system. On the other hand, when $\overline{Fi} > Fi_{crit}$, the trends undergo a rapid initial rise and then continue to oscillate about some supercritical Fi level, their behavior resembling underdamped response. Intermediate between these extremes, critically damped response occurs when $\overline{Fi} \approx Fi_{crit}$.

Observations of ink traces also revealed qualitative changes in flow behavior when the internal Froude number exceeded the critical value. Dye streaks in flows with small Fi remained approximately parallel to the side walls of the tank as they deflected downstream in response to the layered, horizontal jet circulations. However, those associated with larger values skewed laterally as much as 30 or 40 degrees, indicating the appearance of slow, periodic transverse currents. In addition, the noses of dye streaks in supercritical flows exhibited tendencies to rotate slowly one way or the other; therefore, small vertical velocity components were present as well. Similar behavior has been observed in wind tunnels by Strom; but, whereas these have been attributed to the effects of small temperature differences across the duct, the motions in the Argonne tank are evidently purely kinematic in origin. Their occurrence appears to be associated with gradual increases in the vertical shear term (u/L) and not with any changes in the horizontal temperature field.

The transition from a steady, two-dimensional flow pattern featuring plane jets to an unsteady regime exhibiting slow transverse oscillations showed some tendency to take place when a form of the Reynolds number had attained a value near 10^3 . For the Reynolds number

$$Re = \frac{uL}{\nu} \quad (2)$$

we use the central velocity and twice the vertical width of the principal jet at the half-velocity point for characteristic measures of u and L as before; in the usual notation, ν is the kinematic viscosity. Nine of the ten runs in the transition zone of Figure 21 exhibited average Reynolds numbers (during the period of five-minute observations used to calculate σ_e^2) that ranged from 0.7×10^3 to 1.3×10^3 ; seven of them were either 1.0×10^3 or 1.1×10^3 . Only one was as large as 1.9×10^3 , this run resulting in the most divergent point in Figure 22.

Since a characteristic value of the internal Froude number ($Fi \approx 0.3$) also obtains at transition, the two expressions may be combined.

Figure 22.

Average scale lengths L of the principal jet observed in the ten runs located in the transition zone of Figure 21 vs the prevailing vertical temperature gradient; solid line represents the relation $L^2 = (2\nu / \sqrt{d\theta/dz}) (Re/Fi)$ for $Re = 10^3$, $Fi = 0.3$, and $\nu = 9 \times 10^{-3} \text{ cm}^2 \text{ sec}^{-1}$.

For this purpose, it is convenient to rewrite Fi in a form in which the coefficient of volume expansion for water has been included in order to convert the gradient of density ($d\rho/dz$) to the experimentally measured gradient of temperature ($d\theta/dz$). Thus numerically,

$$Fi \approx \frac{2u}{L\sqrt{d\theta/dz}} \quad (3)$$

where the constant includes both $d\rho/d\theta$ and g . Upon eliminating u between (2) and (3), we obtain

$$L^2 = \frac{2\nu}{\sqrt{d\theta/dz}} \left(\frac{Re}{Fi} \right) \quad (4)$$

Thus it appears that the scale (L) of the principal jet at transition is dependent solely upon the viscosity and the density gradient of the fluid.

Figure 22 plots the average scales observed for the ten runs in the transition zone of Figure 21, the solid line representing the L^2 relation (Eq. 4) in water at 25°C with $Fi_{\text{crit}} \approx 0.3$ and $Re_{\text{crit}} \approx 10^3$. The fairly good

fit is, of course, an indication of the degree with which these simultaneous criteria were actually seen to occur. However, it cannot claim to demonstrate the general validity of this particular form of the L^2 relation; for there remains the possibility that the relative nearness of the upper and lower boundary surfaces in the Argonne tank significantly influenced the magnitude of L , thereby affecting the magnitudes of both Fi and Re at transition. A program of experimentation in a significantly deeper volume of stratified fluid could contribute further to this point.

One can also eliminate L between (2) and (3) to obtain

$$u^2 = \frac{\nu \sqrt{d\theta/dz}}{2} (Re_{crit}) (Fi_{crit}) \quad (5)$$

Thus, the indicated simultaneous occurrence of both Reynolds and Froude criteria implies a central jet velocity again dependent only upon the viscosity and vertical gradient of density. Therefore, it appears that transition is characterized by the attainment of a critical profile since both u and L are specified by the condition of the fluid medium. In fact, since transition to the regime of slow oscillations must inherently involve an increase of energy losses over those of the preceding steady flow pattern, the critical configuration would probably tend to preserve itself in cases where the circulation has had just sufficient energy to attain the transitional state, but not enough to maintain the less economical oscillatory condition.

Finally, it is of interest to remark that plane jet flow in quiet, stably-stratified air can lead to the horizontal layers of smoke often found in rooms occupied by the more contemplative users of tobacco. Such a pattern of tracer striation can be seen in the lightly-shaded, relatively-stagnant regions marking the upper and lower boundaries of the principal jet in the experimental photograph of Figure 18. The repeated introduction of vertical dye streaks for each successive photo has resulted in the gradual accumulation of dye in the shearing zones between the principal jet and the lesser counter currents above and below. In the stably-stratified air of a quiet room where one might find a lapse rate of (say) 1°C per meter, the L^2 relation predicts a vertical spacing between stagnant smoke layers of 52 cm, assuming that the residual currents in the room were such that the flow tended to remain near transition. More practically, these relations may be of value for the estimation of the characteristics of maximum, steady two-dimensional flows under the stratified conditions characteristic of the flow of shallow wedges of cold air over sloping terrain or of density currents in the ocean depths.

The writer would like to express his thanks to Professor Dave Fultz of The University of Chicago for suggesting these experiments by bringing to our attention the unpublished work of Francis. The experimental program and the reduction of the resultant data were largely carried out by

Mr. William Prepejchal, Radiological Physics Division of Argonne National Laboratory, who also contributed a number of improvements to both the apparatus and to the procedures employed in reducing the data.

References

1. H. Moses and G. H. Strom. A Comparison of Observed Plume Rises with Values Obtained from Well-known Formulas. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1960. ANL-6297, pp. 81-108.
2. W. R. Debler. Stratified Flow into a Line Sink. Proc. Am. Soc. Civil Engrs. 85 51-65 (1960).
3. R. R. Long. The Motion of Fluids with Density Stratification. J. Geophys. Research 64 2151-63 (1959).
4. C. S. Yih. Effect of Density Variation on Fluid Flow. J. Geophys. Research 64 2219-23 (1959).
5. F. K. Ball. Long Waves, Lee and Gravity Waves. Quart. J. Roy. Meteorol. Soc. 85 24-30 (1959).
6. A. Craya. Critical Regimes of Flows with Density Stratification Tellus 3 24-42 (1951).
7. C. G. Rossby. On the Vertical and Horizontal Concentration of Momentum in Air and Ocean Currents. Tellus 3 15-27 (1951).
8. P. Frenzen. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December 1960. ANL-6297, pp. 109-113.
9. C. S. Yih. On the Flow of a Stratified Fluid. Proc. Third Natl. Congr. Appl. Mech., 1958 pp. 857-61.
10. J. Francis. Personal letter communicated by D. Fultz.
11. H. Schlichting. Boundary Layer Theory. New York: McGraw-Hill, 1955.

ANALYSIS OF SOIL- AND AIR-TEMPERATURE DATA BY FOURIER TECHNIQUES

James E. Carson

The annual and diurnal cycles of air and soil temperature at the DuPage County site of the Argonne National Laboratory have been described in previous reports of this series.(1-3) A set of case studies showing the thermal response of the soil to typical weather conditions was used to illustrate the diurnal cycles of soil and air temperature on individual days. It was shown that the temperature of the air and soil varies in a somewhat regular pattern, reflecting both the annual and diurnal cycles of solar radiation. Superimposed on these regular cycles are fluctuations of variable duration and amplitude created by changing weather conditions, such as cloudiness, rain, warm and cool spells, etc. These fluctuations can be essentially removed from the data and the regular cycles isolated and studied by use of data averaged over a suitable time interval. This paper, which is an extension of the previous reports, deals with Fourier analyses of the soil- and air-temperature data and with estimates of the thermal diffusivity of the soil computed from the soil temperature record.

Figure 23 shows the annual cycle of air temperature 5.5 feet above the ground and of soil temperature at seven depths (1, 10, 20, 50 and 100 cms, 10 and 29 feet) during 1955, based on the average daily value at each level.

Figure 23a
Average air temperatures, 1955

Figure 23b-h
Average daily soil and air temperature, 1955

This average is defined as the mean of the twenty-four hourly observations and effectively removes the daily cycle from the record. The dominant feature of these seven curves is, of course, the annual cycle created by the cyclic variation in the amount of solar energy reaching this area. Deviations from this dominant cycle, created by the individual weather situations seen on the daily weather maps, are very pronounced at the 1-, 10-, 20- and 50-cm levels; only extended periods of unseasonable temperatures deviate the annual progression at 100 cm. At the two deepest levels, 10 and 29 feet, only minor departures from the annual march are observed; these are more probably caused by slight drifts in the temperature-measuring system than by real temperature variations. At 29 feet, the seasons are reversed, with the warmest months being February and March, the coolest July and August.

The use of consecutive decade (10-day) averages reduces, but does not eliminate, the effect of these irregular fluctuations in the annual cycle down through the 100-cm level; at 10 and 29 feet, the curves are nearly sinusoidal (Figure 24). This chart shows only slight amplitude and phase variations between the 1- and 20-cm levels. Curves of the annual progression of soil temperature based on monthly averages for a given year are more nearly smooth sine curves, but still are not regular. These fluctuations are eliminated from the annual cycle by the use of several years of data. Figure 25, which shows the annual progression of soil temperature based on three-year averages for each month for the period January 1953 through December 1955, shows no departures from regular waves. The temperature curves below 20 cm exhibit the amplitude and phase variations with depth predicted by the theory of heat flow in semi-infinite solids. In the layer of soil from 1 to 20 cm, however, the variation of phase with depth is less than one month and is, therefore, masked by the averaging procedure. The decrease in amplitude in the soil temperature wave with depth between 1 and 10 cm is less than predicted by heat-flow theory, a point which will be discussed in a later paragraph.

The diurnal cycles of air and soil temperatures, based on average hourly values, are shown in Figure 26 for the months of January, April, July and October for the year 1954. These months are typical of the four seasons. These curves show that irregular fluctuations in the daily cycle of soil temperature (but not of air temperature), created by variable sunshine, rainfall, etc., are effectively removed from the record by the use of only one month of data.

During January, which is normally the coldest month in this area, the amplitude of the diurnal temperature cycle in the soil is very small and much smaller than that of the air above. The daily heating cycle is unable to penetrate as far as 20 cm into the soil in midwinter.

Figure 24

Annual progression of soil temperature at indicated levels below surface based on 10-day averages.

Figure 25

Annual progression of soil temperature based on 3-year averages for each month. Jan. 1953-Dec. 1955

Figure 26

Hourly average air temperature and soil temperatures at indicated levels.

This small amplitude is caused by the low sun and short days in this season, the high moisture content (and, therefore, high specific heat) of the soil, the blanketing effect of a snow cover when present, and the release (absorption) of the heat of fusion as the soil water freezes (thaws). The temperature gradient within the soil and between the air and soil shows that the heat stored in the soil during the previous summer is being returned to the atmosphere.

In the spring, the greater amount of solar energy reaching the soil surface due to a higher sun and longer days warms the upper 10 feet of soil and creates a diurnal temperature wave in the soil (and in the atmosphere) which is easily measureable at 20 cm. In summer, the drier soil plus greater insolation per day create temperature waves of considerable amplitude in the soil and air. This diurnal heating cycle is able to penetrate about 50 cm into the soil. In the fall, the amplitude of the temperature waves decreases considerably as the amount of solar radiation per day decreases. An important feature of this diagram is the rapid decrease of amplitude and the increasing lag in the time of extremes with depth. It is clear that the temperature waves in the air and soil are not symmetrical since the heating portion of the cycle is shorter than the cooling. An interesting feature of this diagram is the rather flat maximum in the air-temperature curve (as compared to that of the soil at 1 and 10 cm) in midsummer. This flatter peak is caused by the intense daytime convection in the atmosphere during the hot season.

Harmonic (Fourier) analysis provides a completely objective and quantitative description of any regular cycle, such as the soil temperature waves described above. In addition, a study of the variation of the Fourier coefficients with depth in the soil provides information of the heat-transfer processes operating and the thermal properties of the soil.

It can be shown that any time series consisting of a finite number of equally-spaced data points can be completely accounted for by a finite number of sine and cosine terms in a Fourier analysis. For example, if the average temperature of the soil for each hour of the day is given, then 12 cosine and 11 sine terms plus the mean are sufficient to describe the diurnal cycle completely. The usual form of the Fourier series is:

$$T = \bar{T} + \sum_{n=1}^{N/2} \left[A_n \cos \left(\frac{360}{P} nx \right) + B_n \sin \left(\frac{360}{P} nx \right) \right] , \quad (1)$$

where $\bar{T}$ is the average value of the variable in the time interval studies; n is the number of the harmonic; N is the total number of equally-spaced data points; A_n and B_n are the amplitudes (half-range) of the various terms; P is the period of the fundamental cycle (usually but not always equal to N);

and x is the time factor. The period of the second harmonic is $P/2$, etc., and there are $N/2$ harmonics. The time factor, x , varies from 0 to $(N-1)$. A more convenient representation of a Fourier analysis for meteorological purposes is:

$$T = \bar{T} + \sum_{n=1}^{N/2} \left[C_n \sin \left(\frac{360}{P} nx + \Phi_n \right) \right] , \quad (2)$$

where C_n is the amplitude of the n th harmonic and equal to $(A_n^2 + B_n^2)^{1/2}$ and Φ_n is a phase angle equal to $\arctan (B/A)$. The computational uncertainty created by the fact that $\arctan (B/A)$ has two values between 0° and 360° can be eliminated by the additional requirement that the phase angle be also equal to $\arcsin (B/C)$.

The details of the computational procedures and the method of finding the phase angle may be found in standard texts, such as Conrad and Pollak.⁽⁴⁾ This form of the Fourier equation is better for meteorological data since the amplitude of each harmonic is easily seen and the time of extreme values of each harmonic can be easily computed. The maximum value of any harmonic is reached when

$$\left[\frac{360}{P} nx + \Phi_n \right] = 90^\circ \text{ or } 450^\circ \quad (3)$$

The air and soil temperatures collected by the Meteorology Group have been subjected to a series of harmonic analyses. The calculations were performed on an IBM 650 computer using a program prepared by H. Muller and M. H. Dipert of the Biology Division.

The first program was concerned with the annual cycles of air and soil temperature for the three years of available soil temperature data (1953-1955), based on monthly averages. The resulting coefficients are given in Table 31. In this table, $N = 12 = P$ and x varies from 0 (January) to 11 (December). Thus the data for $x = 0$ correspond to mid-January and $x = 11$ to mid-December, etc. Figure 27 shows the variation with depth of the amplitude of the first harmonic for each of the three years and the date that this harmonic reached its maximum value. This figure shows that the temperature cycle in the soil follows the expected pattern of a damped, lagging wave, except in the layer from 1 to 10 cm. The time of warmest soil at the 1- and 10-cm depths is about 20 July, a few days after the maximum air temperature is reached. At deeper levels the temperature maximum is delayed, coming on August 15th at 1 meter, about October 10th at 10 feet, and not until the following January or February at 29 feet. Perhaps the most remarkable feature of Table 31 is the small variation from one year to the next in the time of maximum of the first harmonic at all soil levels (except 29 feet).

Table 31

Fourier coefficients for the annual march of air and soil temperature based on monthly averages.

1953

n	Air		1 cm		10 cm		20 cm	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	23.72	262	23.16	260	22.99	259	21.82	255
2	0.24	294	0.84	95	1.37	108	1.15	112
3	1.79	357	0.73	349	1.14	347	1.05	342
4	0.24	354	0.25	238	0.25	253	0.25	242
5	0.95	126	0.94	150	1.00	140	0.67	135
6	0.00	0	0.06	0	0.03	0	0.02	0
Mean	49.56		53.11		52.72		52.52	
n	50 cm		100 cm		10 feet		29 feet	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	19.05	247	15.52	236	7.23	193	Missing	
2	0.83	108	0.49	107	0.07	330		
3	0.82	336	0.47	302	0.29	223		
4	0.23	0	0.25	242	0.19	219		
5	0.28	101	0.15	336	0.22	277		
6	0.06	0	0.09	0	0.09	0		
Mean	52.36		52.29		51.79			

1954

n	Air		1 cm		10 cm		20 cm	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	23.28	265	23.65	262	23.19	261	21.90	258
2	1.13	3	1.15	72	1.22	70	1.07	59
3	1.63	9	0.71	9	0.77	14	0.71	9
4	0.45	96	0.72	115	0.88	118	0.87	113
5	3.36	346	1.42	0	1.39	2	1.07	5
6	1.64	0	0.89	0	0.76	0	0.59	0
Mean	49.02		53.34		52.65		52.48	

Table 31 (Cont'd.)

1954 (Cont'd.)

n	50 cm		100 cm		10 feet		29 feet	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	18.96	250	15.15	238	6.99	193	0.73	82
2	0.73	45	0.68	44	0.35	354	0.13	251
3	0.70	359	0.41	14	0.15	283	0.08	259
4	0.69	110	0.45	113	0.09	349	0.09	240
5	0.46	351	0.20	254	0.12	144	0.02	160
6	0.34	0	0.12	0	0.06	0	0.05	0
Mean	52.33		52.48		52.49		51.60	

1955

n	Air		1 cm		10 cm		20 cm	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	27.26	268	24.37	264	24.18	264	22.82	261
2	2.75	325	2.00	35	1.78	36	1.54	32
3	1.29	169	1.11	135	1.41	128	1.36	119
4	2.74	70	2.36	64	2.46	62	2.21	61
5	0.82	282	0.21	223	0.17	229	0.13	177
6	0.41	0	0.07	0	0.01	0	0.04	0
Mean	48.63		53.18		52.56		52.38	

n	50 cm		100 cm		10 feet		29 feet	
	Amp	Angle	Amp	Angle	Amp	Angle	Amp	Angle
1	19.41	253	15.50	241	6.98	195	0.93	62
2	1.20	22	0.94	11	0.28	351	0.07	67
3	1.16	103	0.80	79	0.29	13	0.09	158
4	1.53	51	0.91	27	0.19	303	0.22	247
5	0.34	118	0.26	114	0.09	195	0.06	302
6	0.07	0	0.12	0	0.01	0	0.06	0
Mean	52.33		52.43		52.51		51.78	

Only two years of data are available at the 29-foot level due to an instrument failure. The greater variability in the time of extreme values at this level is due in part to the small amplitude of the wave. An unexpected detail is the small phase and amplitude variation between 1 and 20 cm.

Figure 28 is a plot of the variation with depth of the higher harmonics of the annual soil temperature wave. The same harmonics for the annual cycle of air temperature are also entered for completeness. An unusual feature of this chart is the large value of the fifth harmonic in the air and upper layer of soil. As one progresses further into the soil, the relative magnitude of the various harmonics falls more into line with the expected pattern. An examination of the data in Table 31 shows that the year 1954 was rather peculiar in that this large value for this harmonic was not present in the other years. The very warm February in 1954 (10.2°F above normal) was actually warmer than March (2.0°F cooler than normal), creating a non-typical annual sequence of soil and air temperatures; these conditions are, of course, reflected in Fourier coefficients.

Figure 27

Fourier analysis. Amplitude and date of maximum temperature of the first harmonic of the annual air and soil temperature cycle, based on monthly averages.

Figure 28

Fourier analysis. Amplitude variation with depth of the higher harmonics for the air and soil temperature cycle in 1954.

Table 31 and Figures 27 and 28 show that the annual temperature cycle in the soil is fairly well described by the first harmonic alone.

Since the variation of amplitude and phase of the first harmonic in the upper 20 cm of soil using monthly averages was much less than what would be expected from the theory of heat flow in solids, the annual march of soil temperature at 1-, 10- and 20-cm levels using decade (10-day) averages also was subjected to Fourier analysis. It was hoped that this procedure would yield a more accurate description of the change of phase of the soil temperature wave with depth in the layer of soil near the surface. The results of this study, using three times as many data points to describe the annual cycle, show practically the same phase and amplitude variation with depth. The date of maximum for the first harmonic at a given level never varies by more than one day between the two analyses. Thus, the use of the additional points does not alter the pattern of the phase changes and amplitude ratios in the upper soil layers.

The Fourier coefficients for the daily cycle of air and soil temperature, based on average hourly data for each, have also been computed. The results of this calculation are too lengthy to be included in this article, but will be given in an Argonne report to be published soon. Since the daily soil temperature cycle is completely damped out in the upper two feet of soil, no analysis was made of the soil temperature record at 100 cm and deeper.

The daily wave of soil temperature, as contrasted with the annual wave, is not truly represented by a single harmonic. Typically, the primary heating mechanism, net radiation, is rather uniform and negative from just before sunset to just after sunrise and reaches a high positive value about noon. The heating function and the resulting daily temperature wave are therefore not symmetrical. Except on days of great air mass change, the maximum temperature of the soil surface is usually reached at 1 to 2 P.M. and the minimum near sunrise, making the cooling part of the cycle about twice as long as the warming. The use of averages tends to remove the effects on the daily cycle of those days where air mass changes and other non-periodic meteorological conditions force the curve from the usual pattern.

Figure 29 shows the amplitude of the first 5 harmonics of the diurnal soil and air temperature cycles (some harmonics with amplitudes less than 0.1°F have been omitted for clarity) and the time of maximum of the first and second harmonics for alternate months in 1953. The chart for January shows that the amplitude of each harmonic is much greater in air than in the soil, while the soil temperature at 20 cm and below shows no daily variation. This pattern is repeated in February.

Figure 29

Fourier analysis of daily air and soil temperature wave. Amplitude and time of first maximum air and soil temperature.

In the other months, when the soil is not frozen and/or covered with snow, the amplitude of the daily cycle decreases rapidly with depth and is almost completely damped out in the upper 50 cm of soil. The amplitude of the first harmonic of air temperature is almost always greater

than the corresponding harmonic in the soil; the reverse is often true for the second, however. The first harmonics in the air and at 1 cm are about in phase. In contrast with the annual cycle, the phase change in the soil from 1 to 20 cm is quite pronounced. There is considerable variation in the time of maximum of the first harmonic at 50 cm, due to the small amplitude of the temperature variation, random fluctuations not created by the radiation cycle, and errors of observation. At 50 cm, the extremes are about 14 to 18 hours behind those of the soil surface. Thus the maximum at this level is reached about the same time as the minimum in the air and upper levels of soil; night and day are reversed at this shallow depth.

The original data can be reconstructed from the Fourier coefficients by plotting all of the harmonics and then adding the resulting curves. If all $N/2$ harmonics are used, the original data are fitted exactly. Figure 30 shows several of the harmonics of the daily soil temperature cycle at the 1-cm level for June 1953. Also shown are the observed average hourly temperatures and the sum of the first two and the first three harmonics. The sum of the first two harmonics fits the observed data quite well.

A frequently asked question when dealing with harmonic analysis is, "How many harmonics should be computed?" Statistically this question may be asked: "How many harmonics are needed to reduce the variance below a selected value?" The variance is given by

$$\text{VAR} = \sigma^2 = \frac{\sum d^2}{N} \quad (4)$$

where d is the difference between data and the sum of the first n harmonics, and σ is the standard deviation. The variance of the 0th harmonic is the variance of the data about the average value $\bar{T}$. It can be shown⁽⁵⁾ that the variance accounted for by a given harmonic is $C_n^2/2$, except for the last where it is C_n^2 . The ratio of this quantity to the total variance shows the fractions of the variability accounted for by each harmonic.

An IBM program to reconstruct the original curve from the Fourier coefficients has been prepared by Mr. M. H. Dipert of the Biology Division. This program, a Fourier synthesis, computes the value of the first harmonic at each data point (that is, for each value of x) and the variance and standard deviation of the observed values from this curve. The program then computes the sum of the first two harmonics and the variance and standard deviation from this curve, etc. A feature of the program is that the synthesis can be stopped at any time that the remaining variance falls below a pre-selected level.

Figure 30

Fourier synthesis of 1-cm soil temperature wave for June 1953.

The results of a Fourier synthesis computation are given in Table 32 using the annual cycles of air and soil temperature based on monthly averages. The table lists the standard deviation of the data points from the reconstructed curves based on the sum of the first n harmonics. The table also gives the percentage of the total variance accounted for by the sum of these harmonics. As much as 99.8 percent and as little as 93.0 percent of the total variance of the annual cycle is accounted for by the first harmonic although it requires the first 5 harmonics to account for 99 percent of the variance of air temperature in 1954.

Table 32

Fourier synthesis of the annual cycle of air and soil temperature.
Standard deviation of data from the sum of the first n harmonics
and the fraction of the total variance accounted for by these
coefficients.

Level	Year	Standard Deviations								Percent of Variance Accounted for by Sum of Indicated Harmonics				
		Harmonic								Harmonic				
		0	1	2	3	4	5	6		1	2	3	4	5
Air	1953	16.83	1.45	1.44	0.69	0.67	0	0		99.3				
	1954	16.77	3.22	3.12	2.90	2.88	1.64	0		96.3	96.5	97.0	97.1	99.0
	1955	19.50	2.98	2.25	2.07	0.70	0.40	0		97.7	98.7	98.9	99.9	
1 cm	1953	16.41	1.05	0.86	0.68	0.66	0	0		99.6				
	1954	16.81	1.72	1.52	1.44	1.34	0.89	0		99.0				
	1955	17.39	2.32	1.85	1.67	0.15	0	0		95.1	98.9	99.1		
10 cm	1953	16.32	1.45	1.08	0.72	0.70	0	0		99.2				
	1954	16.49	1.72	1.50	1.39	1.24	0.76	0		98.9	99.2			
	1955	17.26	2.36	2.01	1.74	0.10	0	0		98.1	98.6	99.0		
20 cm	1953	15.47	1.21	0.89	0.50	0.46	0	0		99.4				
	1954	15.56	1.45	1.24	1.14	0.96	0.59	0		99.1				
	1955	16.28	2.13	1.83	1.56	0.08	0	0		98.3	98.7	99.1		
50 cm	1953	13.50	0.86	0.63	0.26	0.20	0	0		99.5				
	1954	13.44	0.99	0.84	0.68	0.47	0.33	0		99.5				
	1955	13.82	1.62	1.38	1.11	0.24	0	0		98.6	99.0			
100 cm	1953	10.98	0.53	0.40	0.21	0.13	0	0		99.8				
	1954	10.73	0.67	0.47	0.36	0.17	0.10	0		99.6				
	1955	11.01	1.10	0.88	0.67	0.20	0.10	0		99.0				
10 ft	1953	5.12	0.30	0.30	0.21	0.17	0	0		99.7				
	1954	4.95	0.29	0.15	0.10	0.08	0	0		99.7				
	1955	4.94	0.31	0.24	0.14	0	0	0		99.6				
29 ft	1953	-	-	-	-	-	-	-		-				
	1954	0.54	0.12	0.08	0.06	0	0	0		95.1				
	1955	0.68	0.18	0.18	0.15	0	0	0		93.0				

The theory of heat conduction in semi-infinite, homogeneous media predicts that the logarithm of temperature amplitude should decrease linearly and that the lag of extreme values should increase linearly with

increasing depth.⁽⁶⁾ Therefore, plots of the amplitude against depth on semilogarithm paper and phase (expressed in radians) on linear graph paper should be straight lines, with a slope of magnitude

$$\left(\frac{\pi}{\tau\kappa}\right)^{1/2} = \left(\frac{\omega}{2\kappa}\right)^{1/2} \quad (5)$$

when ω is the frequency (rad/sec) and τ is the period (seconds) of the harmonic, and κ is the thermal diffusivity (cm^2/sec) of the medium.

Plots of the first harmonic of the annual soil temperature wave on the proper paper show a nearly linear variation with depth, except for the layer from 1 to 10 cm. It would appear that the annual temperature variations are consistent with the semi-infinite homogeneous model, except in the top 10 cm of the soil. Plots of the first harmonic of the daily cycle (Figure 29) are not straight lines and thus do not fit the predicted pattern. Hence, the model is unsatisfactory for cycles of this period, indicating that other modes of heat transfer are operating, such as vapor transfer of heat, percolation of water, etc., or that the top several inches of soil are not homogeneous.

If heat is conducted through the soil in accordance with the theory of heat flow in semi-infinite homogeneous media, the thermal diffusivity of a layer of soil may be computed by comparing the temperature amplitude at two depths by means of the following formula:⁽⁶⁾

$$\kappa = \frac{\pi}{\tau} \frac{(z_2 - z_1)^2}{(\ln A_1/A_2)^2} \quad (6)$$

where A_1 and A_2 are the amplitudes at depths z_1 and z_2 and τ is the period of the wave.

The thermal diffusivity may also be computed from the phase variation with depth:

$$\kappa = \frac{\tau}{4\pi} \frac{(z_2 - z_1)^2}{L^2} \quad (7)$$

where L is the phase variation between the two depths (in time units).

The thermal diffusivity of the layer of soil between each of the soil temperature probes has been computed, using the first harmonic of the annual soil temperature wave and the above formulae. The results of these calculations are given in Table 33. Except for the layer from 1 to 10 cm, fairly good agreement in the values for a given year by the two

methods is seen, with the amplitude variation method usually yielding somewhat smaller values. The values are also in reasonable agreement with the diffusivity values found by other investigators for clay soils.⁽⁷⁾ The year-to-year variations are also relatively small. It is clear that the diffusivity increases with depth. An unexpected result of these calculations is the large diffusivity value found for the soil layer from 1 to 10 cm. This may be due to a failure of the Fourier analysis to detect a real phase and amplitude variation between the two levels using only 12 data points. Similar calculations using decade Fourier coefficients did not alter the results. It would appear that other heat transfer processes, such as the movement of soil moisture and the freezing-thawing cycle, mask the transfer of sensible heat in this layer, or that this layer is a very good conductor of heat, despite the presence of roots and organic materials which are relatively poor conductors.

Table 33

Thermal diffusivities of the soil computed from the annual march of soil temperature based on monthly averages (cm^2/sec).

Layer	1953		1954		1955	
	Amp	Phase	Amp	Phase	Amp	Phase
1 cm-10 cm	0.149	0.0917	0.0210	0.0327	0.134	0.0219
10 cm-20 cm	0.00365	0.00267	0.00304	0.00266	0.00298	0.00300
20 cm-50 cm	0.00487	0.00496	0.00432	0.00537	0.00343	0.00510
50 cm-100 cm	0.00593	0.00598	0.00492	0.00540	0.00492	0.00558
100 cm-10 feet	0.00718	0.00798	0.00706	0.00664	0.00658	0.00690
10 feet-29 feet	-	-	0.00655	0.00900	0.00868	0.00737
10 cm-100 cm	0.00525	0.00427	0.00446	0.00456	0.00408	0.00456

The thermal diffusivities of the first 50 cm of soil have also been computed from the first harmonic of the daily cycle of soil temperature for each month, using both the phase and amplitude relations. During the warmer part of the year (May to September), the computed values of thermal diffusivity for the layers 10 to 20 and 20 to 50 cm are in reasonable agreement with those computed by means of the annual cycle and with the published data. In the 1- to 10-cm layer in all seasons and the other layers during the cooler portion of the year, the values computed by the two formulae are very poor, being as much as a factor of ten too large (especially in the 1- to 10-cm layer in all but January and February) or too small. The results of these calculations show that heat-flow theory developed for homogeneous media does not apply in the soil.

Many other investigators have estimated the thermal diffusivity of the soil from the soil temperature record. Good agreement between the two formulae (phase and amplitude) and with measured diffusivities have been found by some of these researchers⁽⁸⁻¹⁰⁾ and poor agreement by others.⁽¹¹⁻¹⁴⁾ The discrepancies are, as pointed out by Swinbank⁽¹¹⁾ and others, due in part to heat transfer due to water vapor movement within the soil, and in part to the non-homogeneous nature of the soil since the thermal properties of soil vary with depth as well as with time as the moisture content varies. Lettau,⁽¹⁴⁾ Aichi,⁽¹³⁾ de Vries,⁽¹⁵⁾ and others have treated the problem of heat flow in soils in detail and have offered more realistic models of the heat transfer mechanisms.

It is interesting to note that, in agreement with this study, those investigators who computed thermal diffusivities from the annual soil temperature record usually found realistic and consistent values, while those using the daily cycle did not. This would seem to indicate that non-conductive heat-transfer mechanisms in the soil are quite large in the upper foot or so of soil, but that the averaging process over a year (except for the top four inches) minimizes their importance.

References

1. J. E. Carson and H. Moses. Soil Temperature Studies. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1957. ANL-5829, pp. 187-199.
2. J. E. Carson and H. Moses. Soil Temperature Studies. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1958. ANL-5967, pp. 210-218.
3. J. E. Carson. Soil Temperature Studies. Argonne National Laboratory Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049, pp. 55-68.
4. V. Conrad and L. W. Pollak. Methods in climatology, 2nd ed. Cambridge: Harvard Univ. Press, 1950.
5. H. A. Panofsky and G. W. Brier. Some applications of statistics to Meteorology. University Park, Penn.: State Univ. Press, 1958.
6. H. S. Carslaw and J. C. Jaeger. Conduction of heat in solids, 2nd ed. Oxford: Clarendon Press, 1959.
7. J. H. Chang. Ground temperature. Cambridge: Harvard Univ. Press, 1958.
8. D. C. Pearce. Ground temperature in studies at Saskatoon and Ottawa, Canada. Extrait des comptes rendus et rapports, Assoc. Intern. Hydrol. Sci. UGGI, Toronto 1957, IV, 279-290, 1958.

9. D. C. Pearce and L. W. Gold. 1959: Observations of ground temperature and heat flow at Ottawa, Canada. *J. Geophys. Research* 64 1293-1298 (1959).
10. N. K. Johnson and E. L. Davies. Some measurements of temperatures near the surface in various kinds of soils. *Quart. J Roy. Meteorol. Soc.* 53 45-59 (1927).
11. W. C. Swinbank. Note on the direct measurement of the thermal conductivity of soil. *Quart. J. Roy. Meteorol. Soc.* 74 409-410 (1948).
12. S. T. Tamura. Observations of earth temperature in Japan. *Monthly Weather Rev.* 33 296-302 (1905).
13. K. Aichi. On the new method of reduction of observations of underground temperature. *Proc. Phys. Math. Soc. Japan, 3rd Ser.*, 1 2-7 (1919).
14. H. H. Lettau. Improved models of thermal diffusion in the soil. *Trans. Am. Geophys. Union* 35 121-132 (1954).
15. D. A. de Vries. On the integration of the heat conduction equation with periodic variation of temperature. *J. Meteorol.* 14 71-76 (1957).

PUBLICATIONS

January through June 1961

- Berlman, I. B. Transient Dimer Formation by 2,5-diphenyloxazole. *J. Chem. Phys.* 34 1083-1084 (1961).
- Berlman, I. B. Pyrene Dimers. *Nature* 191 595 (1961).
- Failla, G. Recovery from Radiation Damage at the Cellular Level. *Radiation Research* 14 465 (1961) (Abstract).
- Failla, G., and P. Failla. Long-Term Effects of Radiation. Trans. IX International Cong. Radiology, Munich, Germany, 1959. Stuttgart, Germany: Georg Thieme Verlag, 1960 Vol. II, pp. 1195-1203.
- Gustafson, P. F., S. S. Brar and M. A. Kerrigan. Radioactivity in Food. *Radiation Research* 14 471 (1961) (Abstract).
- Gustafson, P. F., M. A. Kerrigan and S. S. Brar. A Comparison of Be^7 and Cs^{137} in Ground Level Air. *Nature* 191 454-456 (1961).
- Marinelli, L. D., C. E. Miller, H. A. May and J. E. Rose. The Use of Low Level Gamma Scintillation Spectrometry in the Measurements of Activity in Human Beings. Radioactivity in Man, ed., G. Meneely. Springfield, Illinois: C. C. Thomas, 1961 pp. 26-29.
- Marinelli, L. D. The Effect of Chronic Low Levels of Radiation in Man: The Contribution of Epidemiological Studies. Trans. IX International Cong. Radiology, Munich, Germany, 1959. Stuttgart, Germany: Georg Thieme Verlag, 1960. Vol. II pp. 1236-1238.
- May, H. A. Supplement to the Use of Low Level Gamma Scintillation Spectrometry in the Measurements of Activity in Human Beings, by L. D. Marinelli, et. al. Radioactivity in Man, ed., G. Meneely. Springfield, Illinois: C. C. Thomas, 1961 pp. 84-96.
- May, H. A., and L. D. Marinelli. Factors Determining the Ultimate Detection Sensitivity of Gamma Scintillation Spectrometry. Proc. of the Total Absorption Gamma-Ray Spectrometry Symposium, Gatlinburg, Tennessee, May 10-11, 1960, ed., G. Chapman. U.S. Atomic Energy Commission Report TID-7594 (1961) pp. 236-248.

- Miller, C. E. Sensitivity of Scintillation Scanners. Radioactivity in Man, ed., G. Meneely. Springfield, Illinois: C. C. Thomas, 1961. pp. 75-83.
- Finkel, A. J., and C. E. Miller. Early Metabolism of Intravenously Injected Radium in Mice. Radiation Research 14 466 (1961) (Abstract).
- Platzman, R. L. Probabilité d'ionisation par transfert d'énergie d'atomes excités à des molécules. J. phys. radium 21 853-851 (1960).
- Platzman, R. L. Total Ionization in Gases by High-Energy Particles: An Appraisal of Our Understanding. Intern. J. Appl. Radiation and Isotopes 10 116-127 (1961).
- Hart, E. J., and R. L. Platzman. Radiation Chemistry. Mechanisms in Radiobiology, ed., M. Errara and A. Forssberg. New York: Academic Press, 1961 Vol. 1, Chap. 2, pp. 93-257.
- Rowland, R. E. Microscopic Metabolism of Ra^{226} in Canine Bone and Its Bearing on the Radiation Dosimetry of Internally Deposited Alkaline Earths. Radiation Research 15 126-137 (1961).

PAPERS TO BE PUBLISHED

- Anderson, W. R., and I. B. Berلمان. Radiant Flux From a Cylindrical Distributed Source. *J. Opt. Soc. Am.* (November, 1961).
- Brar, S. S., P. F. Gustafson, M. A. Kerrigan, B. C. Bridenstein, U. S. Garrigus, and W. R. Usburne. Radioactivity in Wool. *Nature*.
- Gustafson, P. F., S. S. Brar and M. A. Kerrigan. Air-Borne Radioactivity Due to Nuclear Weapons Tests. *Science*.
- Machta, L., and H. F. Lucas. Radon in the Upper Atmosphere. Submitted to *Science*.
- Marinelli, L. D., and H. F. Lucas. Translocation of Thorium Daughters to Bone. Symposium on "Some Aspects of Internal Irradiation," Heber, Utah, May 8-11, 1961. Pergamon Press.
- May, H. A., and L. D. Marinelli. Sodium Iodide Systems: Optimum Crystal Dimensions and Origin of Background. Symposium on Whole Body Counting, Vienna, June, 1961.
- Miller, C. E. An Experimental Evaluation of Multiple-Crystal Arrays and Single-Crystal Techniques. Symposium on Whole Body Counting, Vienna, June 1961. Submitted to *Health Physics*.
- Remenchik, A. P., and C. E. Miller. The Measurement of Total Body Potassium in Man and Its Relation to Gross Body Composition. Symposium on Whole Body Counting, Vienna, June 1961.
- Durup, J., and R. L. Platzman. Role of the Auger Effect in the Displacement of Atoms in Solids by Ionizing Radiation. Discussions of the Faraday Society (1961).
- Durup, J., and R. L. Platzman. La contribution de l'ionisation en couche profonde a la radiolyse des hydrocarbures et de quelques substances biologiques. *J. Chim. Phys.*
- Rowland, R. E. Skeletal Retention of the Alkaline Earth Radioisotopes and Bone Dosimetry. Symposium on "Some Aspects of Internal Irradiation," Heber, Utah, May 8-11, 1961. Pergamon Press.

PATENTS

Atmospheric Eddy Disturbance Detector

AEC Case S-17, 312
Inventor, Harry Moses

STAFF ACTIVITIES

Lucas, Henry F., Jr. Epidemiologic Aspects of Background Exposure from Radium. Summer Postgraduate Course, Epidemiology of Radiation Injury. St. Louis University School of Medicine August 7-18, 1961.

Robert E. Rowland was Chairman of the Session on Selected Communications of the Gordon Research Conference on Bones and Teeth, Meriden, N. H., July 10-14, 1961.