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Argonne National Laboratory

RADIOLOGICAL PHYSICS DIVISION

SEMIANNUAL REPORT

July through December, 1960

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ARGONNE NATIONAL LABORATORY
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Argonne, Illinois

RADIOLOGICAL PHYSICS DIVISION
SEMIANNUAL REPORT

July through December, 1960

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

February 1961

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A STUDY OF TRANSIENT DIMERS IN A SOLUTION CONTAINING NAPHTHALENE

Isadore B. Berlman

Introduction

Forster⁽¹⁾ has demonstrated that an excited molecule (in this case pyrene was excited with uv radiation) can combine with a similar unexcited molecule to form a transient dimer. The process may be written as follows:

where A , A^* , $(AA)^*$, and ν represent respectively the unexcited molecule, the excited molecule, the transient dimer, and the frequency of the emitted quantum of radiation. Dammers-de Klerk⁽²⁾ has used this concept to explain the high quantum yield of naphthalene in various solutions at very high solute concentrations: namely, she postulated that the excited naphthalene molecules form transient dimers with unexcited naphthalene molecules and thereby become immune to external quenching. The formation of transient dimers in solutions containing large concentrations of PPO has been reported recently.⁽³⁾

As the solute concentration is increased, 1) a shift of the emission spectrum to longer wavelengths and 2) the presence of a second and longer component in the mean fluorescent lifetime are clues to the existence of dimers. Dimer formation by PPO⁽³⁾ and pyrene⁽¹⁾ is favored in aliphatic solvents much more than in aromatic solvents and varies in an inverse manner with the viscosity of the solution. In the present report the formation of transient dimers in solutions of naphthalene in cyclohexane (an aliphatic compound of low viscosity) as a function of the solute concentration was studied.

Materials and Methods

The naphthalene was recrystallized three times from ethanol solutions,⁽⁴⁾ and the cyclohexane, Eastman spectro grade, was vacuum distilled once. Both aerated and nitrogen-bubbled solutions were used. The apparatus for measuring the emission spectrograms and the mean emission lifetimes are fully described in Reference 5.

Results

Emission spectrograms of naphthalene-cyclohexane solutions were made to investigate the tell-tale "fingerprint" of dimers, namely a shift of the emission spectrum to longer wavelengths. As the concentration of the solute was varied from 1 mg/liter to 100 g/liter, no change in the emission spectrum was noted, except at the very highest concentrations (≥ 50 g/liter) where a slight hint of dimer formation is presented by the small shift in the emission spectrum to longer wavelengths, as shown in Figure 1. Warming the concentrated (100 g/liter) solution to approximately 40°C removed the new long wavelength portion of the emission spectrum, as shown in Figure 2. The same emission spectrum was obtained with the following excitation wavelengths, 3130 Å, 2890 Å, and 2650 Å.

Figure 1

The spectrograms of solution of naphthalene in cyclohexane. Curve A, 1-mm sample of a nitrogen-bubbled solution of 1 g/liter of naphthalene in cyclohexane. Curve B, a 0.05-mm sample of a nitrogen-bubbled solution of 50 g/liter of naphthalene in cyclohexane. Curve C, a 1-mm sample of an aerated solution of 1 g/liter of naphthalene in cyclohexane. These curves have been corrected for the wavelength response of the equipment.

Figure 2

Curve A is a spectrogram of a 0.05-mm sample of a 100-g/liter solution of naphthalene in cyclohexane at 25°C. Curve B is the spectrogram of the same solution at approximately 40°C.

The effect of oxygen quenching on the quantum yield from naphthalene is very pronounced, as shown in Figure 1; it has been estimated that an excited naphthalene molecule is approximately 50 times more vulnerable to quenching by oxygen than an excited PPO molecule.⁽⁶⁾ The excessive susceptibility of the excited naphthalene to quenching is mainly due to its long lifetime.

Mean lifetime measurements of several of the above solutions were made to ascertain the presence and enhancement of a longer component in the fluorescent lifetime with increasing solute concentration. The measured mean emission lifetimes of all of the deoxygenated solutions were found to have at least two components, 65 ± 8 nsec and $0.7 \pm 0.3 \mu\text{sec}$, irrespective of the solute concentrations. The equipment however was not capable of measuring longer components, should they exist. Oxygen in aerated solutions reduced the shorter component of the fluorescent lifetime to 21 ± 3 nsec, and erased the longer lived component, as shown in Figure 3.

Figure 3

Curve A is a semilog plot of the fluorescent lifetime of a nitrogen-bubbled solution of 1 g/liter of naphthalene in cyclohexane. Curve B is a similar plot of the lifetime of an aerated solution.

Conclusion

Since neither the magnitude and the shape of the emission spectrum nor the shorter component (the only one which can be conveniently measured) of the emission lifetime is affected by changes in the solute concentration, we find it necessary to assume that naphthalene molecules have negligible self-quenching, and that no appreciable formation of transient dimers at moderate (> 0.1 g/liter) solute concentration takes place, as is assumed by Dammers-de Klerk.⁽²⁾ At the very highest concentrations (≥ 50 g/liter) the slight change in the emission spectrum favors assumption of the initiation of dimer formation.

The pronounced quenching by oxygen of the fluorescent light, invariant at all concentrations of naphthalene, militates against the contention that dimers are necessary to explain the large quantum yield from

naphthalene, unless the transient dimers themselves are quenched just as effectively as single molecules. Further work is contemplated along these lines.

References

1. Th. Forster and K. Kasper. Z. Electrochem. 59 976 (1955).
2. A. Dammers-de Klerk. Molecular Phys. 1 141 (1958).
3. I. B. Berlman. J. Chem. Phys., to be published.
4. Performed in 1956 by W. L. Buck and kindly provided by Dr. A. Weinreb.
5. I. B. Berlman. J. Chem. Phys. 33 1124 (1960).
6. I. B. Berlman. J. Chem. Phys. 34 598 (1961).

BONE STUDIES ON AN EXHUMED RADIUM PATIENT

R. E. Rowland

Exhumation of radium poisoning victims has long been considered as a means of studying the early effects of radium in human bone. However, although many of the cases who died a few years after acquiring this isotope undoubtedly could be found, it is questionable whether the metabolic and dosimetric information obtained from such studies would be sufficiently precise unless the quantity of radium and the route of administration were known.

In this respect the Elgin State Hospital patients who were given Ra^{226} under controlled conditions in 1931⁽¹⁾ are ideal. After 20 or more years retention studies have been made on these cases,^(2,3) and the distribution of radium in their bones has been studied by quantitative autoradiographic techniques.⁽⁴⁾ In addition, a clinical investigation of these patients was reported in 1955⁽⁵⁾ and is continuing to date.⁽⁶⁾ Hence, any additional information that could be developed about this series would be of considerable value to the over-all problem of the effects of radium on man.

A careful review of the records of these patients has recently been made,* from which eight early deaths among the cases have been found. These patients are identified by code number in Table 1, with the date and quantity of radium administered, when known, and the date of death.

Table 1

Deceased Elgin State Hospital patients injected with Ra^{226}

Patient code	Date of death	Quantity of Ra^{226} administered	Year in which Ra^{226} was given	Remarks
03132	10/4/1931	Unknown	1931	Suicide Cremated
03129	5/6/1933	Unknown	1931	
03137	9/10/1934	Unknown	1932	
03130	3/26/1936	Unknown	1931-32	
03138	3/31/1936	Unknown	1932	
03123	2/5/1937	90 μc	1931	
03140	4/26/1937	Unknown	1933	
03120	6/25/1937	110 μc	1931	

*We are greatly indebted to the superintendent, Dr. Daniel Haffron, and the Clinical Director, Dr. Werner Tuteur, of the Elgin State Hospital, for permission to study the files of these cases.

Unfortunately, for six of these cases the quantity of radium administered was not explicitly stated. In fact, three of these patients did not even receive their radium in 1931, but later, during 1932 or 1933. Consequently, the method of administration of Ra^{226} to these latter groups may not have been the same as it was for the 1931 group when the following schedule prevailed: $10 \mu\text{c Ra}^{226}$, I.V., once a week for periods ranging from 8 to 45 weeks.

Nevertheless, when it appeared possible to obtain a bone sample from Case 03140, an exhumation was performed. The pertinent details of this case are listed in Table 2. In the records the following notation was found:

"This patient has been on this ward about six weeks. Has received 11 injections of radium chloride. 4/10/33."

Table 2
Description of patients

	Case 03140	Case 03139	Case 03141
Sex	Male	Male	Male
Date of birth	February 9, 1905	1908	1907
Radium acquired	Age 28, 1933	Age 25, 1933	Age 26, 1933
Physical data	129 lbs., 5'7" (12/3/31)	140 lbs (10/23/31)	127 lbs (5/23/30)
Date of death	April 26, 1937, age 32	Living	Living
Cause of death	Pulmonary tuberculosis		
Exhumation date	October 13, 1959		

At exhumation a sample was removed from the right femur for study. It was found that the burial vault had not been penetrated by ground water, and that the preservation of the body tissues was remarkably good, considering that 22 years had elapsed since death. The following studies have been completed on the sample:

1. Total Ra^{226} content and radon retention in a 79-g cortical bone sample, by the spectroscopic measurement of the γ -rays emitted.
2. Specific activity measurements of the hotspots and the diffuse distributions by means of quantitative alpha-track autoradiographs.
3. Bone changes by means of microradiographs of 100-micron thick bone sections.
4. The microscopic distribution of radium by means of stripping-film autoradiographs.

The results of the γ -ray and the α -ray autoradiographic measurements are summarized in Table 3. From each set of measurements an extrapolation to the total skeletal radium content has been made; the γ -rays yield $0.53 \mu\text{c}$, and the autoradiographs yield $0.55 \mu\text{c}$. For both measurements the extrapolations are based on the assumption that the total skeletal mass is one tenth the body weight; usually extrapolations from gamma-ray measurements of small masses of bone are poor, but it is interesting to note that both measurements yield a similar body content.

Table 3

Summary of measurements (Case 03140)

From γ -ray measurements	
Radon retention	25.9%
Ra ²²⁶ content (79 g bone)	$0.090 \mu\mu\text{c}/\text{mg bone}$
Extrapolated body burden	$0.53 \mu\text{c}$
From autoradiographic measurements	
Maximum hotspot intensity	$10.8 \mu\mu\text{c}/\text{mg bone}$
Average diffuse concentration	$0.044 \mu\mu\text{c}/\text{mg bone}$
Ratio: hotspot to diffuse	246.
Extrapolated body burden	$0.55 \mu\text{c}$

The autoradiographic extrapolation is based on the observation that the diffuse level is 0.47 of the average skeletal content;⁽⁷⁾ this is probably the more reliable of the two methods mentioned above.

The stripping film autoradiographs showed that the intense deposits of radium in the bone samples were in narrow rings in haversian systems, averaging less than 50 microns in thickness. This is in marked contrast to the deposits in the other Elgin cases which have been studied,⁽⁴⁾ where completely labeled osteons were often observed. It thus appears that the radium injections did take place over only a short time interval and that the average linear growth rate in replacement haversian systems was somewhat less than 10 microns per week.

The narrow width of these bands proved to be a serious deterrent to quantitative evaluation of the specific activity levels in the "hotspots." Since the average range of the radium alpha particle is about 30 microns,

the tracks covered an area in the autoradiograph larger than the radium deposit actually covered in the bone section; therefore, the specific activity level listed in Table 3 is probably too low.

An indication that this is the case is given by the ratio of the maximum hotspot to the average diffuse level, which was found to be 246 in this patient. Previously a ratio of 218 was found in an Elgin patient who had been injected over an 18-week interval (Case I, Reference 7). Since this ratio is independent of the magnitude of the radium injections (as long as they are equal), but dependent upon the length of time the injections are continued, we would predict a six-week period of injections to produce a hotspot to diffuse ratio of magnitude close to 500.

The microradiographs of these bone sections showed no changes characteristic of radium damage. This was to be expected, for few such changes are seen in patients with 30-year burdens of less than $1 \mu\text{c}$,⁽⁸⁾ and, by comparison with the radium now present in Case 03139, (see below) the 30-year burden for our exhumed case would have been a small fraction of $1 \mu\text{c}$. Thus, from this study we could not expect to determine how early the typical radium changes actually occur in human bone; an examination of bone from a case who had received a much larger quantity of radium is required.

One of the unusual features about this case is the fact that another patient, who received radium at the same time, is still alive and has been extensively studied and measured, and had been considered to be one of the classic 1931 cases. This latter case, coded as 03139,* was of similar age and build as 03140, as can be seen from the data in Table 2.

In the hospital records the following notations for case 03139 were found:

"Patient is receiving (radium)** chloride intravenously. 3/31/33."

"Patient has been on this ward about six weeks and has received 11 injections of radium chloride intravenously. 4/10/33."

A third case, whom we code as 03141, was also found who had received radium in 1933. In his records the notation read:

"Patient has received 11 injections of radium chloride intravenously. Has been on this ward about six weeks. 4/10/33."

*This patient was coded as Paul N. in References 2, 5, and 9.

**On the typewritten record sheet the word "radium" had been inserted in ink, above the line, with a caret (^) to indicate that it belonged before the word "chloride."

This patient is still living in a state institution, but his total body content of Ra^{226} has not yet been measured.

These three cases, then, appear to have received identical radium treatments at the same time. Although we have no indication that the treatments actually terminated on 4/10/33, they may well have, for the fact that the date of the final notation in all three cases was the same may indicate that this treatment was being summarized.

It is of interest to compare the findings in our exhumed case with observations on the living patient in regard to total body burden. We first ask: How much radium did these cases actually receive? Granted that they received 11 injections over a 6-week period (i.e., two per week), and assuming that these were equal in magnitude, how large were they? Three possible magnitudes are suggested below with the reason for considering each alternative:

- 1) $10 \mu\text{c Ra}^{226}$ each injection:
(the same quantity as given in the 1931 series.)
- 2) $5 \mu\text{c Ra}^{226}$ each injection:
(half the magnitude given in 1931, to yield the same weekly dose.)
- 3) $3.3 \mu\text{c Ra}^{226}$ each injection:
(three cases were found who received radium at this time; one $10 \mu\text{c}$ ampule could provide three $3.3\text{-}\mu\text{c}$ doses each time the injections were given.)

In Table 4 are tabulated the expected body burdens at several times following administration, calculated from

$$R_t = 0.54t^{-0.52} \quad (\text{Reference 2})$$

for the three possibilities suggested above. Included for comparison are the measured values for Case 03139. The values of the total body content reported for 1951 and 1953 are now known to be incorrect, for subsequent gamma-ray measurements with large NaI(Tl) crystals in the low background room have yielded higher values for retained RaB and RaC than were found in the earlier measurements. Fortunately, it appears that the early radon breath measurements were valid, so that they may be employed to calculate the total Ra^{226} retained in the body. In Table 4 both the published and the recalculated values are shown; for the latter the assumption was made that 70% of the radon formed in the body is exhaled. The 1960 measurement was of the retained RaB and RaC only; the total Ra^{226} content is calculated from this measurement with the same assumption, namely, that 70% of the radon is lost. (See also a summary by C. E. Miller⁽⁹⁾ regarding the early measurements on this and other Elgin cases.)

Table 4

Power function prediction of total body content of Ra^{226}
following administration during March of 1933

Date	Predicted retention for three injection levels, μc			Measured retention, μc	
	36.3	55	110	Case 03140 (exhumed)	Case 03139 (living)
April 1937 (4 years)	0.446	0.615	1.35	0.53, 0.55	-
May 1951 (18 years)	0.205	0.309	0.62	-	0.46* (0.56)**
July 1953 (20 years)	0.194	0.292	0.585	-	0.52* (0.60)**
1960 (21 years)	0.167	0.250	0.50	-	(0.56)***

* Published total retention in Reference 2.

** Values from Reference 2 recalculated from radon breath measurements.

*** Based on an unpublished measurement of $0.167 \mu\text{c}$ ($\text{RaB} + \text{RaC}$) by C. E. Miller.

The conclusion is reached from examination of Table 4 that the total body measurements from Case 03139 are in excellent agreement with the assumption that he received 11 injections, $10 \mu\text{c}$ Ra^{226} each, totaling $110 \mu\text{c}$ Ra^{226} . It thus seems quite logical to assume that our exhumed case received the same total dose since the medical records are identical. Why, then, do the estimates of total skeletal content differ so from the predictions based on the power law?

Let us re-examine the concepts from which we extrapolated the total skeletal content. It is quite possible that the total radium content of our 79-g sample is not representative of the total skeleton, so that little reliability is based on this estimate. However, the magnitude of the diffuse level has been a reliable guide (within a factor of two) to the total skeletal content as measured in the human cases that have carried radium for 20 years or longer. If the total burden were actually $1.35 \mu\text{c}$ at time of death, as the power law predicts, then the observed diffuse level is only

0.19 times the average skeletal concentration. No value this low has been observed as yet although ratios as low as 0.23 have been found twice⁽⁷⁾ (once for another Elgin case who had carried radium for 24 years, Case R).

The power law itself, however, was derived from data that show a spread of a factor of two over a 22-year period.⁽²⁾ Thus, the difference between the expected and the calculated total body burden at time of death could be within the normal spread to be expected.

Measurement of Case 03141, the third patient who received radium in 1933, will be of great significance in the interpretation of the differences between the retention of radium in the exhumed (03140) and the living case (03139). Until further clarification of the administered and retained radium doses has been achieved, the validity of the total body radium content extrapolated from exhumation samples must be held in doubt.

References

1. 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.* 30 515-22 (1933).
2. 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).
3. C. E. Miller. Unpublished measurements (1960).
4. R. E. Rowland and J. H. Marshall. Radium in human bone: The dose in microscopic volumes of bone. *Radiation Research* 11 299-313 (1959).
5. W. B. Looney, R. J. Hasterlik, A. M. Brues and E. M. Skirmont. A clinical investigation of the chronic effects of radium salts administered therapeutically (1915-1931). *Am. J. Roentgenol.* 73 1006-1037 (1955).
6. A. J. Finkel, C. E. Miller, and R. J. Hasterlik. Status report of study of long-term effects of radium in humans. Argonne National Laboratory Radiological Physics Division Semiannual Report, July through December, 1959, ANL-6104 pp. 87-88.
7. 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. pp. 339-353.

8. R. E. Rowland, J. H. Marshall, and Jenifer Jowsey. Radium in human bone: The microradiographic appearance. Radiation Research 10 323-334 (1959).
9. 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.

RADIOACTIVE HOTSPOTS, BONE GROWTH AND BONE CANCER*

Self-Burial of Calcium-like Hotspots

John H. Marshall

The problem of estimating the effects of internally-deposited radioisotopes upon the human skeleton is being pursued in several ways. One of these is the investigation of the long term effects of radium upon human bone, together with experiments to determine the relative effects of radium and other radioisotopes upon animals such as mice and dogs. The ratio of the effectiveness of a given radioisotope to that of radium in producing an effect in animals can then be applied to the known effect of radium in man in order to estimate the effect of other radioisotopes. An advantage of this procedure is that the existence of human cases of radium poisoning gives us an experimental measurement of the sensitivity of human bone to long-continued bombardment by internal radiation.

In determining the effectiveness of a radioisotope - and let us narrow the discussion to the induction of osteogenic sarcomas - one needs a number to describe the quantity of radioactivity in, or radiation to, the skeleton. The simplest quantity is the total microcuries of activity in the skeleton or the activity per unit skeletal weight. One can then perform many different comparisons of the bone tumor incidence in animals resulting from different kinds of radioactivity and different schedules of intake, and so derive the ratio of effectiveness of each radioisotope to that of radium under different conditions.

This is a straightforward program likely to produce the desired results, but it has two natural limitations. First, the number of different combinations of isotope, level of injection, and schedule of injections is very large. Second, the implicit assumption is made that the ratio of the effectiveness of two radioisotopes in animals under given conditions will equal their ratio in man under similar conditions. Can we make better approximations to the effective quantity of radiation in the skeleton and so reduce the number of possibilities that must be tested by experiment? Can we make any generalizations that would give us a basis for predicting changes in the ratio of the effectiveness of two radioisotopes from species to species?

One important generalization is that, other things being equal, the effectiveness of a radioisotope should depend on the total energy of the radiation absorbed in the skeleton. Thus, there should be less difference between the effectiveness of different radioisotopes on an energy basis than on a microcurie basis. We then ask for the ratio of effectiveness of two radioisotopes for the same average skeletal dose.

*Presented at the Conference on Radioisotopes in Bone, Princeton, New Jersey, August 29, 1960.

Can we take one more step and find generalizations that can take into account the different distributions of dose within the skeleton? This step from the average skeletal dose to the dose distribution introduces immense complexity. We must understand temporal as well as spatial non-uniformities in dose, for they are usually interrelated. We must consider what microscopic locations in the skeleton are likely to be particularly sensitive to the carcinogenic effect of radiation. Finally, we must understand the relations between the physiological structure and dynamics of bone and the deposition of radioisotopes.

Useful generalizations on this level of microscopic complexity are difficult to find, and they are likely always to be only rough approximations of the tremendous variety of biological phenomena. However, they will serve their purpose if they give us consistent explanations for the difference in tumor incidence in the different animal experiments with which we might strengthen the prediction of the relative effects of different radioisotopes upon man.

An attempt at one such generalization is the subject of this paper. It concerns a relation between the growth of a typical bone surface, the formation of a hotspot there when Ra^{226} or Ca^{45} is injected into the blood, and the subsequent radiation dose delivered to the growing surface. The possible value of the relation stems from the fact that the details of growth rate, shape of blood curve, and particle type or range drop out, leaving an expression that should be approximately true for most hotspots in a skeleton following a single injection of a calcium-like radioisotope of short particle range.

The present approach to the problem of hotspots and bone cancer has been suggested by experiments concerning the incidence of bone tumors following the injection of Pu^{239} , Ra^{226} , Sr^{90} , or Ca^{45} into mice.^(1,2) It seems that plutonium is at least several times more effective than radium in inducing osteogenic sarcomas for comparable average doses to the skeleton. The dosimetry for this experiment has not been completed, but it seems likely that the main reason plutonium is more effective than radium is that plutonium deposits primarily on bone surfaces⁽³⁾ and that the surfaces of bone are particularly sensitive to radiation. This same conclusion is suggested by comparison of the bone tumor incidences due to injected Ca^{45} and Sr^{90} .⁽⁴⁾ Comparable average doses of Ca^{45} produce fewer tumors than Sr^{90} , not more, in spite of the presence of Ca^{45} hotspots. The most probable explanation is that most of the Ca^{45} hotspots bombard the bone matrix and the osteocytes, and not the bone surfaces, and thus are not particularly effective in inducing tumors.

Here, I would like to consider one particular point. The conclusion that bone surfaces are the sensitive regions for the induction of osteogenic sarcomas introduces somewhat of a paradox. We know that most Ca^{45} and

Ra^{226} hotspots are formed at the microscopic surfaces of bone that are growing at the time of injection. Therefore, these hotspots will initially bombard the sensitive surface region. A consistent explanation of the comparative dosimetry and tumor incidence of Ra^{226} versus Pu^{239} and of Ca^{45} versus Sr^{90} should somehow account for the fact that the Ca^{45} and Ra^{226} hotspots formed at growing surfaces do not seem to be very effective in inducing tumors.

It is the thesis of this paper that such calcium-like hotspots are buried rapidly enough by continuing growth that the dose accumulated at the bone surface during the process of their formation and of their subsequent burial is not many times more than the dose that would be accumulated by the bombardment of the diffuse component alone. This burial process is here termed "self-burial" in order to emphasize the difference between hotspots which are formed by the growth process - the calcium-like hotspots - and those which do not require that the surface be growing. A hotspot whose formation depends upon the growth of the bone surface at the time of injection will usually be covered by continuing growth. If the particle range is short enough, this burial should be quite effective in reducing the dose rate at the surface of the bone. Contrast with this the situation for plutonium or for a rare earth radioelement of short particle range. Plutonium deposition on bone surfaces does not require growth.⁽³⁾ Therefore, for long periods after injection, many of the deposits of plutonium may be expected to remain on the surface.

The problem of the burial of calcium-like hotspots and the consequent reduction of the dose to the soft tissue outside bone has been mentioned recently by Lamerton.⁽⁵⁾ Here I would like to try to estimate the order of magnitude of this effect. Just how much radiation dose does a hotspot deliver to the surface of bone during the process of its formation and subsequent burial?

Consider what happens at a typical, growing surface of bone when a single radioactive injection of a calcium-like isotope is made. Figure 4 shows an autoradiograph of the femur of a mouse injected with Ra^{226} . The time is 30 days after the radioactive injection. The outlined area is enlarged in Figure 5.

Figure 4

Alpha particle autoradiograph of a longitudinal section through the femur of a mouse 30 days after an injection of Ra^{226} .

Figure 5

Enlargement of outlined area in Figure 4

Part of the bone surface along the edge of the cortical bone is growing and has deposited Ra^{226} intensely. This radioactive surface of bone is called a hotspot. In addition to this hotspot deposition, you can see the diffuse uptake within the cortical bone that existed at the time of injection. Notice that on one side of the bone growth stopped shortly after injection, leaving the radioactive deposit near the surface, but that on the other side of the bone growth continued at a rate of about 1 micron a day so that after 30 days the deposit is buried by about 30 microns of bone. The radium alpha particles can just reach the present surface.

As a first approximation to this situation let us calculate what the dosage distribution would be near a plane of activity. Here we make the assumption that all of the activity in the hotspot is located on the plane that corresponded with the position of the surface at the time of injection. Figure 6 shows the results of this calculation for Ca^{45} , Ra^{226} , and Sr^{90} . The dose values have been averaged over a 10-micron interval. Notice that, if a surface is growing at the rate of a few microns a day, the dose rate for Ca^{45} or Ra^{226} will drop very quickly as the surface grows out from under the point of maximum dose rate. This is particularly true for the alpha-particle dose from radium. Sr^{90} , due to the long beta-particle range, gives so uniform a dose distribution that the following analysis does not apply.

The distribution of activity deposited by a growing bone surface is not quite as simple as a plane of activity. Following a single injection of the radioisotope, the blood specific activity drops rapidly but still remains significant for many days after injection. Therefore, the activity is not confined to just one plane but is deposited within a slab of bone at specific activities which drop rapidly in the direction of growth.

Figure 6

Calculated dose distribution for a plane source of activity deposited within bone. An average over 10- μ intervals has been used to prevent the dose from becoming infinite at zero microns. Ra^{226} values were obtained assuming straight tracks and constant rate of energy loss. Ca^{45} and $\text{Sr}^{90}\text{-Y}^{90}$ values were calculated from the point source function.⁽⁶⁾ The integral doses for all three isotopes are the same.

Figure 7 shows the result of a calculation of this situation assuming a constant rate of surface growth of 3 microns per day, a blood specific activity decrease with time that is very much like that observed in animals, and a particle range in bone of 30 microns. This is the dose rate profile to be expected following a single injection of Ra^{226} . The alpha particle dose is a maximum near the location of the growing surface at the time of injection, and it decreases sharply on each side. Now such detailed calculations as this for a whole skeleton become unmanageable because there are hotspots formed by many different rates of surface growth and because different animals may have different curves of blood specific activity versus time. Any generalizations would be very difficult.

Figure 7

Calculated dose distribution for the deposition of Ra^{226} at a growing surface of bone assuming a power function blood specific activity curve. A uniform bone medium is assumed (i.e. growth has buried the deposit).

However, it is possible to make an approximate calculation for calcium-like hotspots in general. We make three basic assumptions:

1. That the radioactivity is deposited at the specific activity of the blood.
2. That the particle range is short.
3. That whatever the rate of surface growth at the time of injection, this same rate of growth is maintained for a few days or weeks after injection. Mathematically, we will assume that the given rate of surface growth is maintained indefinitely. (Surfaces which stopped growing abruptly soon after injection would receive higher doses than those estimated here.)

Let the growth rate of the bone surface have a constant value ℓ day^{-1} . Then, following a single injection of calcium-like radioactivity, the total activity deposited per unit surface area is

$$L = \ell \rho c \int_0^t B \, dt \quad (1)$$

where

L = total activity deposited between injection and time t ($\mu\text{c}/\text{cm}^2$)

ℓ = growth rate (cm/day)

ρ = density of bone (g/cm^3)

c = calcium content of bone (gCa/g)

B = blood specific activity ($\mu\text{c}/\text{gCa}$)

Expression (1) states that the total activity deposited by the growing surface is proportional to the linear growth rate and to the integral of the blood specific activity. In other words, this activity is being deposited by accretion; $\ell\rho c$ is the calcium accretion rate. This expression, suggested by the work of Bauer, Carlsson, and Lindquist⁽⁷⁾ has been verified by direct measurements of Ca^{45} uptake and linear growth rate.⁽⁸⁾ Let t be any time greater than a few days; then L represents the greater part of the activity deposited by the growing surface and practically all the activity which contributes to the hotspot.

What dose rates will the activity L produce? If we assume, for simplicity, that the medium around the growing surface is uniform (neglecting the effect of the difference between bone and soft tissue), then application of the conservation of energy yields the relation

$$\int_{-\infty}^{\infty} R \, dx = KL/\rho \quad (2)$$

where

R = dose rate as a function of x (rads/day)

x = distance from the position of the bone surface at the time of injection (cm)

K = conversion factor ($\text{rad-g}/\mu\text{c-day}$) which depends upon the energy emitted per disintegration

Expression (2) states that the integral of dose rate, the area under the curve of dose rate versus position (Figure 8), is proportional to the activity L . It depends upon the fact that the total energy absorbed in the vicinity of the hotspot is equal to the energy of the particles emitted.

Figure 8

Sketch of the dose rate distribution near a growing bone surface produced by the deposition of calcium-like radioactivity following a single injection.

Given this integral dose rate [Expression (2)], what is the dose accumulated at the bone surface as it grows away from the radioactive deposit? Assuming that the growth rate is still ℓ cm/day, we find

$$P = \int_0^{\infty} R \, dt = \int_0^{\infty} R \, dx/\ell = (1/\ell) \int_0^{\infty} R \, dx \quad (3)$$

where

P = the dose accumulated at the bone surface (rads)

R = the dose rate at the bone surface (rads/day)

x = the distance the bone surface has moved since the time of injection (cm)

The expression for accumulated dose in terms of dose rate and time has been converted into an integral of R with respect to x similar to that in Expression (2) by means of the substitution $\ell = dx/dt$.

Now we can tie all these expressions down to a measured quantity and simplify the result by relating P to the dose rate at a non-growing surface due to the diffuse component:

$$S = \frac{1}{2} f K c \int_0^t B \, dt \quad (4)$$

where

S = diffuse surface dose rate (rads/day)

f = augmentation rate (day^{-1}) obtained by dividing the specific activity of the diffuse component ($\mu\text{c/gCa}$) by the integral of

the blood specific activity, $\int_0^t B \, dt$. (9)

Expression (4) merely states that the dose rate at the surface of bone due to the diffuse component alone is proportional to the specific activity of the diffuse component, $f \int_0^t B \, dt$. The factor $\frac{1}{2}$ takes into account the reduction of dose rate at the surface of a thick source.

Finally, let us combine all four expressions and solve for P . With the approximation that

$$\int_0^{\infty} R \, dx \approx \frac{1}{2} \int_{-\infty}^{\infty} R \, dx \quad (5)$$

(which assumes the dose distribution to be symmetrical about $x = 0$). We find simply that

$$P \approx S/f \quad (6)$$

where

P = rads (surface dose from a calcium-like hotspot during burial)

S = rads/day (surface dose rate due to diffuse component)

f = day⁻¹ (augmentation rate)

All the integrals and even the growth rate ℓ have dropped out. The integral of the blood specific activity dropped out because it enters both the expression for hotspot uptake (1) and the expression for diffuse uptake (4). In other words, the injection of different quantities of radioactivity will not change the hotspot-to-diffuse ratio. The growth rate ℓ dropped out because although hotspot activity is proportional to ℓ (1), the accumulated dose P is inversely proportional to ℓ (3). More rapid growth produces a more intense hotspot, but the bone surface is exposed to the high dose rate for a shorter time.

Considering the doses at bone surfaces, we find that the accumulated dose due to a calcium-like hotspot is approximately equal to the diffuse dose rate divided by the augmentation rate (f).

Now what does this expression mean? What values are there for f ? Figure 9 shows some of the measured values for augmentation rate, the rate of long-term transfer of calcium from blood to bone. These values were obtained by dividing Ca^{45} diffuse specific activity (measured autoradiographically) by the integral of the blood specific activity following a single injection of Ca^{45} . For the bone of a young mouse, the augmentation rate should be roughly 90% per year, so P equals S times about 400 days.

Figure 9

Augmentation rate f , the rate of long term calcium transfer from blood to bone, as a function of bone age. The values correspond to the Ca^{45} diffuse specific activity divided by the integral of the blood specific activity. The unmarked points are for canine cortical bone. The figure has been taken from Reference 8.

In other words, the accumulated surface dose due to a hotspot, during the process of its burial, is comparable to the surface dose due to the diffuse component accumulated over a period of 400 days. The time between a radioactive injection and death with an osteogenic sarcoma in mice is the order of magnitude of a few hundred days, so we may interpret this relation as indicating that a calcium-like hotspot, during the process of its burial, gives an accumulated dose to the bone surface of not more than a few times the dose due to the diffuse component. It must be remembered that a constant growth rate has been assumed. If growth stops and leaves calcium-like hotspots bombarding the bone surfaces, the dose will be larger than our estimate. Furthermore, this result applies directly only to single injections. However, the result indicates that the self-burial of calcium-like hotspots should be quite significant in that it reduces the bone surface dose far below the maximum hotspot dose (which may be tens or even hundreds of times the diffuse dose). Also the result suggests that the relative effectiveness of hotspots and the diffuse component may be species dependent through the age-dependent factor f although the greater tumor induction times usually observed for the longer-lived animals may counteract such a species dependence. The conclusion cannot be clear-cut without an understanding of both the spatial and temporal factors. The calculation stands as a particularly simple result in an extremely complicated phenomenon. Its simplicity recommends it as a possibly useful piece in the puzzle relating the formation of radioactive hotspots, bone growth, and the induction of osteogenic sarcomas. Whether the hypotheses presented in this paper will quantitatively fit the dosimetric and tumor incidence information remains to be seen.

References

1. M. P. Finkel. Relative Biological Effectiveness of Radium and Other Alpha Emitters in CF No. 1 Female Mice. *Proc. Soc. Exptl. Biol. Med.* 83 494-498 (1953).
2. M. P. Finkel and B. O. Biskis. The Induction of Malignant Bone Tumors in Mice by Radioisotopes. *Acta Union Intern. Contra le Cancer* 15 99-106 (1959).
3. J. S. Arnold and W. S. S. Jee. Bone Growth and Osteoclastic Activity as Indicated by Radioautographic Distribution of Plutonium. *Am. J. Anat.* 101 367-418 (1957).
4. J. H. Marshall and M. P. Finkel. Autoradiographic Dosimetry of Mouse Bones Containing Ca^{45} , Sr^{90} , or Ra^{266} . II. The Sensitive Region in the Induction of Osteogenic Sarcomas. Radiological Physics Division Semiannual Report, January through June 1960. ANL-6199, pp. 44-54.

5. L. F. Lamerton. Radioisotopes in the Skeleton: Considerations of Radiation Dosage in Relation to Bone Damage, Radioisotopes in the Biosphere, ed. R. S. Caldecott and L. A. Snyder. Minneapolis: University of Minnesota Press, 1960. pp. 382-400.
6. R. Loevinger. The Point Source Function. Radiation Dosimetry, ed. Hine and Brownell. New York: Academic Press, 1956. pp. 709-715
7. G. C. H. Bauer. A. Carlsson and B. Lindquist. Evaluation of Accretion, Resorption, and Exchange Reactions in the Skeleton. Kgl. Fysiograf. Sällskap. Lund Forh. 25(1) (1955).
8. J. H. Marshall. Microscopic Metabolism of Calcium in Bone. Bone as a Tissue, ed. K. Rodahl, J. T. Nicholson, and E. M. Brown, Jr. New York: McGraw-Hill Book Co., Inc., 1960. pp. 144-155.
9. J. H. Marshall, R. E. Rowland and Jenifer Jowsey. Microscopic Metabolism of Calcium in Bone. V. The Paradox of Diffuse Activity and Long-Term Exchange. Radiation Research 10 258-270 (1959).

MEASUREMENT OF K^{42} IN HUMANS

C. E. Miller

The gamma-ray spectra of 20 subjects who contained known quantities of K^{42} were obtained with several different sized NaI(Tl) crystals located at various points around the body. These spectra were analyzed to determine the variations in counting rates (per μc of K^{42} in the body) that resulted with each patient-crystal arrangement when subjects of widely differing body builds were measured in order to assess errors inherent in various "geometries."

Twelve subjects were measured both prone and supine while curved on a 175-cm arc* and while seated in the tilting chair.⁽¹⁾ Also, six of these subjects were measured with a 3" x 5" NaI(Tl) crystal located 12 cm from the back at a sitting height elevation of 27 cm. The same deviations from the average counting rates were noted with the 175-cm arc technique as with the tilting chair technique. The counting rates (per μc in the body) were $2 \pm 2\%$ higher than the average for those subjects who weighed between 130 and 140 pounds, $2 \pm 2\%$ lower than the average for those who weighed between 180 and 200 pounds, and within $\pm 2\%$ of average for those who weighed between 150 and 160 pounds. With the 3" x 5" NaI(Tl) crystal at the small of the back, the counting rates varied from 12% above the average for the smaller subjects to 15% less than the average for the larger subjects. A variation of $\pm 5\%$ was noted for subjects of approximately the same height and weight.

Six subjects were measured while seated in the tilting chair first with an 8" x 4" crystal and then a 9" x 4" crystal over the chair above the subject. They were then measured with a 3" x 5" crystal located under the seat of the chair. The same variations from the average counting rate were observed with both the 8" x 4" and the 9" x 4" crystals. The counting rates observed with the 3" x 5" crystal deviated $\pm 10\%$ for subjects who gave counting rates within 1% of the average observed with the larger crystals.

The gamma-ray spectra were obtained from six patients, lying supine and prone with a 9" x 4" crystal placed at 7 equally spaced locations 24.5 cm apart along the body. (See Figure 10.) The crystal was first placed 30 cm and then 40 cm above the surface of the bed. The variations in counting rates between subjects observed at each crystal position were calculated as well as the variations that existed if combinations of crystals were used. The same percentage deviation occurred for each subject regardless of whether one crystal was used or whether an array of two or more crystals was used front and back, or both. The percentage deviations were about three times those observed when the same subjects were counted with the tilting chair technique.

*An 8" x 4" NaI crystal was located at the center of the 175-cm arc.

Figure 10

The counting rate observed with the crystal 40 cm above the surface of the bed was 71% of that observed with the crystal 30 cm above the bed. Thus it is not possible to eliminate or reduce the deviations from one subject to another by choosing a particular array of crystals at staggered heights from the subject. These data indicate that minimal variability is attained when patients are measured in the tilting chair.

Reference

1. C. E. Miller. Measurement of Gamma Rays in Vivo. Radiological Physics Division Semiannual Report, January through June, 1956. ANL-5596, pp. 26-33.

CESIUM-137 POTASSIUM-40 RATIOS IN EIGHT CONTROL SUBJECTS OVER A SPAN OF 30 MONTHS

C. E. Miller and J. B. Corcoran

The total-body gamma-ray spectra of 12 unexposed employees of the laboratory were measured with Human Spectrometer No. 1 on two dates during 1960. These subjects have been measured periodically, usually every 3 months, since March of 1955.

Their Cs^{137} body contents, expressed in units of $\mu\mu\text{c Cs}^{137}$ per gram of natural body potassium, have been determined, and the contents of 8 are given for 8 dates during the past 30 months in Table 5. It will be noted that the average Cs^{137} content dropped from $55 \mu\mu\text{c Cs}^{137}/\text{g K}$ during December of 1959, to $49.6 \mu\mu\text{c Cs}^{137}/\text{g K}$ during May of 1960, to $37.4 \mu\mu\text{c Cs}^{137}/\text{g K}$ during December of 1960. This represents a 10% decrease in Cs^{137} content during the first 5 months of 1960 and a 25% decrease during the last 6 months. The same percentage decreases were noted for the other four subjects for these last three dates, but their data were not included in the table since they had not been measured every time on the earlier dates.

Table 5

Cesium-137 potassium-40 ratios in eight control
subjects over a span of 30 months

Subject	7/31/58	11/20/58	3/19/59	6/17/59	9/14/59	12/8/59	5/24/60	12/15/60
C L	57	48.5	59	68.5	60	56.2	54	38.4
I S	51.5	52	59	54.6	54.5	47.4	49	44.6
H. M	71.0	68.5	75.2	83.2	68.2	69.5	64.9	55.9
C M	43	58	76	81	64.2	62	50.5	33.9
B O	47	47.8	54.5	57.2	45.5	42.2	45.4	29.8
B. P	43.5	46.2	53.5	59	54.7	53.5	48.2	31.6
R R	39	39.5	47	46	45.6	45.2	38.2	27.9
O S.	48.4	51	61.8	65.5	64	63.1	46.6	37.3
Average	50	51.4	60.7	64.4	57.1	55	49.6	37.4

A group of 11 male Chicago residents, age 40 to 70 years, measured during the last 3 months of 1960 contained an average of $38.8 \mu\mu\text{c Cs}^{137}/\text{g K}$. This value agrees with the amount found for the controls during December of 1960. The spread in individual values was somewhat greater in that they ranged from $20 \mu\mu\text{c Cs}^{137}/\text{g K}$ to $60 \mu\mu\text{c Cs}^{137}/\text{g K}$ while the values of the younger controls ranged from 27.9 to 55.9--a 2 to 1 spread.

ANGULAR RESPONSE OF SCINTILLATION DETECTORS

Harold A. May

The response of a scintillation counter to gamma rays as a function of the angle of incidence may be useful for several reasons. In biological or environmental studies, extended sources must frequently be counted at small source-detector distances in order to reduce statistical uncertainties or counting time. Calibration sources of identical geometry and composition are required for greatest accuracy but may be difficult to devise; however, calculations to a precision of 25 to 50% are usually possible from a knowledge of the scattering and absorption properties of the matrix in which the radioactivity is located and the spatial response and absolute detection efficiency of the counter at the gamma ray energy to be observed.⁽¹⁾ Similarly, in relating the observed background counting rate within a given massive shield to the gamma rays originating in its walls, be it from cosmic-ray interactions or from its residual activity, the average response of the crystal to a uniform, omnidirectional flux must be known.

Some preliminary experiments have been performed to evaluate the response of a 7-inch diameter by 3.5-inch thick crystal, canned in a double-walled stainless steel container designed to provide a built-in one-half-inch thick mercury shield around the sides of the crystal. The mercury, which may be readily drained out, is separated from the crystal surface by 0.020-inch steel sheet and some 0.075 to 0.100-inch of MgO reflector. The walls are welded to stainless steel flanges at both ends, to which the $\frac{3}{8}$ -inch quartz optical window and the 0.010-inch stainless steel gamma-ray "window" are gasketed and clamped by light steel rings. Since there is a fair amount of absorbing material near the corners of the crystal, its response is not likely to be typical of commercially packaged units.

Gamma-ray spectra of Ra^{226} + daughters, Cs^{137} , and Co^{60} were obtained as the angular position of the source, lying in a plane which included the crystal axis, was changed by 30° intervals between runs. The source distance was maintained at 70 cm; therefore, the full angular divergence of the gamma rays at the crystal was of the order of 14° . Total net counts (energy > 25 kev) and those falling in the photopeak were determined at each angular setting. Data from positions of symmetry about the axis (i.e. 30° and 330° , etc.) were averaged; in no case did the total count of either differ by more than $3\frac{1}{2}\%$ from their mean. The degree of collimation produced by filling the shield with mercury was also tested using Ra^{226} only. The resulting numbers, expressed as a fraction of the count obtained with the source at 0° (in front of the crystal, on axis), are tabulated in Table 6. Figures 11 to 13 present the same data in polar coordinate form.

Table 6						
Source	Cesium 137		Cobalt 60		Radium	
Energy range	25 910 kev (total)	410 910 kev (photopeak)	0 025 1 575 Mev (total)	0 87 1 575 Mev (both peaks)	0 025 1 57 Mev	0 025 1 57 Mev (Hg in housing)
Angle						
0°	1 0	1 0	1 0	1 0	1 0	1 0
30°	0 93	0 89	0 94	0 92	0 92	0 83
60°	0 81	0 72	0 84	0 76	0 80	0 45
90°	0 72	0 70	0 78	0 75	0 70	0 21
120°	0 74	0 62	0 87	0 64	0 71	0 34
150°	0 80	0 79	0 76	0 78	0 75	0 54
180°	0 65	0 60	0 73	0 58	0 61	0 46
I _{avg}		0 73		0 76	0 76	0 43

Figure 11
Angular response, Cs^{137}

Figure 12
Angular response, Co^{60}

Figure 13
Angular response, Ra^{226} and daughters. Total counts

The average response of the crystal to a uniform isotropic flux, relative to that of an isotropic detector with an efficiency equal to the maximum observed experimentally (i.e., at $\theta = 0^\circ$), is given by:

$$I_{\text{avg}} = 1/2 \int_0^{2\pi} F(\theta) \sin \theta \, d\theta \quad .$$

Although $F(\theta)$, a continuous function having the observed values shown in the table at certain discrete angles, is unknown, an approximate value of I_{avg} may be calculated by means of graphical integration. The results of such calculations appear in the last row of Table 6.

The variation in counting rate with angle results from three separate effects. These are:

1. The solid angle subtended by the crystal, and hence the number of gamma rays striking it, is variable. This purely geometric effect may be readily evaluated for a simple right circular cylinder, but would be far more difficult to evaluate for the more complex contour of the crystal container.

2. The number of gamma rays passing through the crystal edges, and therefore traversing less than the average path length within it, also varies, with a resultant change in the interaction ratio. This also could be directly evaluated, in theory, by an extension of available computer-programmed Monte Carlo methods.

3. Interaction of gamma rays with the non-uniform mass of the steel container introduces an angular dependence of the number, energy distribution and directionality actually reaching the crystal.

While the results for cesium and cobalt sources are similar, the energy-dependent differences are readily seen. Minima in the photopeak curves, in the vicinity of 75° and 120° apparently are caused by the thicker steel of the container flanges. Further measurements at lower energies should demonstrate the same general behavior to an even greater extent.

The technical assistance of Paul Hess is gratefully acknowledged.

Reference

1. R. C. McIlhenny and J. McCarthy. Analytical Gamma Spectrometry: Semiempirical Estimations of Response-Curve Slopes. Meeting of Bioassay and Analytical Chemistry Group at Gatlinburg, Tenn., Oct. 1959 TID-7591, p. 40.

THE INDEX OF REFRACTION OF CERTAIN OPTICAL COUPLING MEDIA

Harold A. May

I. Introduction

The physical processes resulting in energy loss within scintillating crystals and the luminescence phenomena leading to emission of light quanta which result may be adequately described by mathematical statements of the appropriate physical laws and verified by direct experiment. The same cannot be said about the ensuing light transfer and collection since the history of each quantum is essentially a random process. Since the light losses, both within the crystal proper and at its boundaries, may be altered by local inhomogeneities or by geometrical factors, a Monte Carlo-type treatment would seem to be required. While such a calculation has been proposed, we are not aware that it has been performed, nor have the necessary experimental techniques been devised to measure directly each variable.

Even though the inevitable non-uniformities in self-absorption and surface reflectivity, as well as geometrical effects, may not be readily accounted for, an approximate but mathematically simple treatment involving average values of these parameters would appear to be useful in predicting the relative performance of large crystals and in indicating what measures should be most fruitful in further improving their resolution capabilities. Several mathematical models of the light transfer process from scintillation site to photocathode which include the major steps suggested above have been proposed.⁽¹⁻⁴⁾ Van Sciver⁽⁴⁾ has shown that the collection efficiency η , defined as the escaping fraction of the light quanta released in the crystal, is given by:

$$\eta = \frac{1 - X}{1 - tX} \quad (1)$$

where $X = (1 - aT/A)$, a = area of crystal surface not covered with reflector, ideally equal to the photocathode area, A = total crystal surface area, and T = transmission at the crystal-window interface. Also, $t = \exp(-\alpha \bar{y})$, i.e., the optical transmission through a medium of absorption coefficient α and average path length $\bar{y}$. The transmission T is averaged over all angles of incidence, assuming a uniform, omnidirectional flux within the crystal,*

*Although the site of the original light flash will usually be nearer the surface through which the penetrating radiation is incident and will shift with incident energy, dependent upon crystal dimensions, the randomizing effects of multiple internal reflections and diffuse scattering from the reflector result in a uniform illumination of the crystal, at least if it is of good quality and of conventional shape.

and may be calculated from the index of refraction of NaI relative to that of the optical coupling media employed. It is approximately equal to $\Omega_c/2\pi$, where Ω_c is the solid angle included in a cone generated by the critical angle θ_c . Hence

$$T = 1 - \cosine(\sin^{-1} \mu_{\text{media}}/\mu_{\text{crystal}}) \quad . \quad (2)$$

A more accurate treatment recognizes the small amount of internal reflection occurring at less than the critical angle, as given by the Fresnel theory.⁽⁵⁾ The effect is small, for, assuming values of μ_D of 1.774 and 1.404 for NaI and silicone oil respectively, the resulting transmission is 0.38 by the approximate formula, versus 0.36 by numerical integration of the Fresnel equations.⁽⁶⁾

Since T and α are wavelength dependent, a rigorous treatment requires a consideration of the crystal emission spectrum, represented by a distribution function $S(\lambda)$. Equation (1) should be replaced by

$$\overline{\eta} = \frac{\int_0^\infty S(\lambda) \eta(\lambda) d\lambda}{\int_0^\infty S(\lambda) d\lambda} \quad (3)$$

In practice, it is probably sufficient to evaluate collection efficiency at the peak of the emission spectrum, namely at around 4100 Å in thallium-activated sodium iodide.

While the alkali halides have been studied extensively, the hygroscopic nature of NaI seems to have prevented any careful measurement of its index over an extended portion of the spectrum. The only readily-available figure, already quoted, was reported by Spangenberg in 1922.⁽⁷⁾ This situation has recently been improved as a result of measurements by Managan and others in the Electronics Division at Argonne National Laboratory.⁽⁸⁾

II. Coupling Media

Values of refractive index of the transparent oils and grease most frequently used for optical coupling were available only at the sodium D line. We here report a series of measurements covering most of the visible region.

An Abbe refractometer was used, Bausch and Lomb model 33-45-58.* This instrument was designed to be used with white light illumination, the image being rendered monochromatic at 5893 Å when properly adjusted by means of compensating prisms inserted into the optical path. These were removed and the prism illuminated by the imaged slits of a Bausch and Lomb monochromator used to select desired lines in the mercury, sodium and hydrogen spectra. The refractometer was not kept at constant temperature since no thermostated water bath was convenient; however, the laboratory temperature was observed to be between 22° and 24° C at all times.

As the refractometer scale is marked directly in refractive index at the D line, the observed reading μ_D must be corrected to give the correct index μ_λ at wavelength λ by the expression

$$\mu_\lambda = \mu_D + \Delta\mu \quad . \quad (4)$$

The correction term $\Delta\mu$ depends upon the region of the index scale being read and upon the wavelength of the incident light, as well as upon certain constants of the particular instrument. Since $\Delta\mu$ is small, it may be calculated for a few assumed values of index μ_s at the particular wavelengths to be used and values for the scale readings actually observed then obtained by interpolation. The correction $\Delta\mu_s$ was calculated from the relationship

$$\Delta\mu_s = \mu_i [\sin \{ B \pm \arcsin (\sin i / \mu_i) \}] - \mu_s \quad (5)$$

where μ_i is the index of refraction of the refractometer prism at the wavelength for which the correction term is being calculated; B, the prism angle of the refractometer; and i, the angle of emergence of light from the prism when in "balance" at the assumed index scale reading μ_s . The positive sign is used if $\mu_s \geq 1.55$, the negative sign otherwise.

Values of μ_i , B and angle i were supplied by the manufacturer. Values of $\Delta\mu_s$ were calculated to six significant figures on an IBM 610 computer, for nine assumed wavelengths and for values of μ_s from 1.3000 to 1.5500 in increments of 0.0500. They ranged in value from +0.04642 at $\mu_s = 1.30$, $\lambda = 4047$ Å to -0.01458 at $\mu_s = 1.30$, $\lambda = 7655$ Å.

Readings could be made to five significant figures, and were usually reproducible to within one or two digits in the last place, except for observations in the extreme blue-violet, where the poor visual acuity limited accuracy. Three to five observations were made and averaged at each point. Table 7 lists the corrected data. DC-703 vacuum pump oil, although not ordinarily used as an optical coupling media, was included because of its high index. Figure 14 permits visual comparison and rough interpolation along the curves.

* This instrument was kindly made available by Mr. Robert Reed of the Remote Control Engineering Division.

Table 7

Index of refraction of selected optical media*

Wavelength, Å	Materials				
	DC-200		QC-2-0057	Mineral Oil	DC-703
	200 centistokes	10 ⁶ centistokes			
4047	1.4151	1.4169	1.4486	1.4782	1.5464
4340	1.4138	1.4149**	1.4401**	1.4718**	1.5391**
4358	1.4132	1.4149	1.4434	1.4738	1.5395
5460	1.4056	1.4060	1.4330	1.4651	1.5238
5785	1.4042	1.4047	1.4318	1.4637	1.5209
5893	1.4043	1.4045	1.4309	1.4633	1.5199
6563	1.4009	1.4014	1.4284	1.4606	1.5150

* T = 23 ± 1° C

** These values fall well below a smoothed curve through the other points, and are presumed to be in error, due perhaps to the very low intensity of the line.

As an independent check on the corrected values, two of the samples were measured by Mr. E. I. Bradshaw of the Sinclair Research Laboratories (SRL), where a B and L Precision Oil Refractometer, capable of measurements to an additional significant place, was available. Certain systematic differences in the two sets of values are apparent, as seen by Table 8.

Table 8

Comparison of measurements of two samples of optical media

DC-200, 10 ⁶ centistokes			
Wavelength	4358 Å	5460 Å	5893 Å
ANL (23° C)	1.4149	1.4060	1.4045
SRL (20° C)	1.4152	1.4074	1.4057
Δ =	-0.0003	-0.0014	-0.0012
DC Grease, type QC-2-0057			
ANL	1.4434	1.4330	1.4309
SRL	1.4435	1.4340	1.4319
Δ =	-0.0001	-0.0010	-0.0010

Figure 14

Index of refraction of selected optical media

The temperature coefficient at the sodium D line is known to be of the order of -0.00037 per degree Centigrade, for most organic oils.⁽⁹⁾ This correction, if assumed valid for the silicones, amounts to -0.00111 and hence, is seen to result in rather good agreement at the longer wavelengths. A cursory literature search revealed no information on how the temperature coefficient changes with wavelengths, hence the significance of the small differences at $4358 \text{ \AA}$ remains unknown. Further measurements at controlled temperatures seem indicated; however, these differences are much too small to affect the optical transmission calculations.

III. Discussion

The refractive index of sodium iodide at $4100 \text{ \AA}$ is 1.855 ; from this and from the interpolated data of Table 7, the transmission of each of the coupling media was obtained, using the approximate relation, Equation (2). The practical effect upon relative light yield was then evaluated from Equation (1) for two typical crystals. Results predicted for a hypothetical medium having an index equal to the geometric mean of NaI and quartz are included since such a medium should give maximum light transfer. Values of $\alpha = 0.005 \text{ cm}^{-1}$ and $\bar{y}$ equaling the mean of crystal diameter and length were used. The results of these calculations are summarized in Table 9.

Table 9

Medium	μ_{4100}	T	$\eta(I)$	$\eta(II)$
DC-200	1.417	0.353	0.555	0.369
QC-2-0057	1.447	0.373	0.570	0.383
Mineral oil	1.477	0.394	0.585	0.396
DC-703	1.545	0.445	0.616	0.427
Optimum	1.65	0.544	0.673	0.478

Crystal I: 4 x 4 inches, one five-inch phototube; hence, $a/A = 0.166$.

Crystal II: 8-inch diameter by 4 inches long, with three 3-inch phototubes; hence, $a/A = 0.0806$.

It is apparent that the increase in light transfer predicted should be readily detectable so that experimental measurements would serve to indicate the validity of the mathematical model. The predicted differences, however, are not great enough to make a significant improvement in photo-multiplier statistics, and hence in overall resolution, except in the case of very low energy gamma and x-rays. If, for instance, one is working with the 14.4-keV gamma-ray of Fe^{57} , even a 10% increase in light output would be quite welcome.

It is a pleasure to acknowledge the assistance of Mr. Ralph Griffith and Mr. E. I. Bradshaw of Sinclair Research Laboratories. Jack Corcoran performed the computer programming.

References

1. P. M. Bird and P. R. J. Burch. The Relative Performance of Large Volume Plastic and Liquid Scintillators. *Phys. Med. Biol.* 2 219 (1958).
2. G. W. Clark, F. Scherb and W. B. Smith. Preparation of Large Plastic Scintillators. *Rev. Sci. Instr.* 28 433 (1957).
3. G. Morton. Recent Developments in the Scintillation Counter Field. *IRE Trans. Nuclear Sci.* NS-3 (4) 132 (1956).
4. W. Van Sciver. Remarks at the Large Crystal Spectrometry Symposium, Cincinnati, Ohio, May 23, 1958. U. S. Atomic Energy Commission Report, Apex-471, p. 28.

5. F. A. Jenkins and H. E. White. Fundamentals of Physical Optics, New York: McGraw-Hill, 1st ed., 1937, Chapter 18, p. 393.
6. Personal communication from Dr. Van Sciver.
7. K. Spangenberg. Z. Kryst. 57 515-518 (1922).
8. W. Managan, S. Baker and R. Adams, to be published.
9. R. Kanthack. Tables of Refractive Indices, Vol. II, Oils, Fats, and Waxes. London, Adam Hilger, 1921.

IMPROVED APPARATUS FOR MEASUREMENT OF THE COSMIC-RAY NEUTRON BACKGROUND

Roger Grismore, O. J. Steingraber,
B. G. Oltman and L. D. Marinelli

Certain measurements undertaken by the present authors to assess the cosmic-ray neutron dose rate to contemporary man have been described in previous reports.⁽¹⁻³⁾ The preliminary neutron flux measurements⁽³⁾ made in room B-110 of building D-203 at Argonne indicated a neutron background several times what would be expected for this latitude from the cosmic-ray neutron studies⁽⁴⁾ of a University of California group. As suspected, this high background has been traced to neutrons produced by two nearby high-energy Van de Graaff accelerators. Using a Hanson and McKibben-type long counter,⁽⁵⁾ it was found that, with the accelerators running, the counting rate varied up to ten times the normal background, and a broad maximum in the flux intensity was found as the long counter was rotated in the horizontal plane through the direction of the larger Van de Graaff. As a result of these findings, the final neutron flux measurements will be made in a location well removed from accelerators and reactors.

Recently, some improvements have been made in the "twin" scintillation fast neutron spectrometer^(6,7) used in the foregoing measurements. A new pair of liquid scintillation cells has been designed and built with somewhat larger dimensions for increased counting efficiency. The cells are open-ended cylinders with $\frac{1}{16}$ -in. thick aluminum walls, Teflon-lined to improve the light reflection. The two photomultiplier tubes which close a cell are sealed by means of O-rings. The design permits seating the photomultipliers so that the active lengths of the cells may be varied up to $2\frac{1}{2}$ in. Although the length of the Teflon liner must be recut for each adjustment of cell length, still the relative ease with which this adjustment may be made greatly simplifies the problem of varying the ratio of the volumes of the two cells for the purpose of amplitude-matching their gamma-ray spectra. In addition, this design facilitates the changing of photomultiplier tubes. The scintillating solutions are also easily interchanged between the cells in order to compensate for natural radioactivity of the containers and cross talk between the photomultipliers. Before insertion of the upper photomultiplier tubes the solutions may be flushed with nitrogen gas for prevention of the oxygen-quenching of scintillators of low fluorescence yield [such as our diphenyloxazole in 1,4-bis (trifluoromethyl) benzene solution]. One further advantage is that, unlike the older design, the new cells may be operated in any desired orientation relative to the horizontal plane.

An additional recent improvement has been the installation of a pulse generator arrangement for frequent checks of the gains throughout the electronic system. The generator is adjusted to deliver a standard pulse which closely resembles the photomultiplier pulses in shape, and

its amplitude is checked by observation with a calibrated oscilloscope and by potentiometer measurement of the pulse-generator charging voltage. A tap has been placed in each photomultiplier-tube housing to permit feeding the standard pulse through a low-capacity connection into the input of the cathode follower fed by the photomultiplier. Thus, the gain stabilities of the pulse-amplifying systems can be checked quickly by observing the responses of the pulse height analyzer as the standard pulse is delivered to individual cathode follower inputs. In the event of a change of gain, the offending circuit can be located easily by measuring the outputs of the successive components of the system involved with a calibrated oscilloscope and comparing the results with those of previous similar measurements. Pulse generator observations of this sort have led to changes in the designs of several of the electronic circuits in the spectrometer with resulting increased gain stability.

The pulse generator is also useful in pinpointing deterioration in a scintillation solution or change in photomultiplier gain through a process of elimination when one of the frequent counting checks with Co^{60} gamma rays indicates a gain loss. Previously, the tests with Co^{60} were the only checks made, and, in the event of change, a great deal of difficulty was encountered in determining the source of trouble. Other operations in which the pulse generator is used include setting and checking the discrimination levels in the photomultiplier-noise-elimination coincidence systems. Likewise, it is used to set and check the balances of the sum circuits where the pairs of pulses from a given cell are added before pulse height analysis.

During our preliminary neutron background measurements with the "twin" scintillation spectrometer, (3) Owen⁽⁸⁾ described a scintillation counter capable of discriminating between neutrons and gamma rays on the basis of the differences in the pulse shapes produced by protons and electrons. It seemed that Owen's method might be practical as an independent means for measuring the neutron background. However, first it was deemed necessary to determine whether the electronic circuitry involved added an indeterminate factor to the known counting efficiency of the scintillator.

To date two different electronic circuits designed to discriminate between neutron - and gamma-ray-produced scintillations have been tested, using a $1\frac{1}{2}$ -in. diameter by $\frac{1}{4}$ - in. thick stilbene crystal mounted on an R.C.A.-type 6810A 14-stage photomultiplier tube as the basic scintillation counter. The first circuit tried was that of M. Forte.⁽⁹⁾ In it, the discrimination pulses which gate the pulse height analyzer for analysis of neutron events are formed by adding negative pulses from the photomultiplier anode to the corresponding positive pulses from the thirteenth dynode. The anode pulses are delay-line clipped so as to eliminate the portions due to the slower components of scintillation decay, while the dynode pulses are handled in the ordinary manner. In operation, the relative amplitudes of the two which are to be combined are adjusted so that after the addition and

subsequent charge integration the resultant pulses are as small as possible for gamma-ray events. These pulses, then, will not gate the pulse height analyzer. However, since proton-recoil scintillations contain a relatively larger proportion of slow component than do electron scintillations, they will produce positive resultant pulses which will gate the pulse height analyzer. The pulses to be analyzed are taken directly from a third element of the phototube.

The limiting factor affecting the above circuit seems to be how completely opposite polarity "overshoot" caused by the reflected pulse from the delay line can be eliminated from the clipped anode pulse since any positive excursion of the latter tends to interfere with the discrimination process. Even limitation of the "overshoot" with good, modern semiconductor diodes leaves something to be desired. Ultimately, it was found impossible to exact the best performance from the Forte circuit without operating the photomultiplier in such a manner as to produce space charge limitation of its current. However, since this method of operation, just by itself, enables excellent discrimination against gamma rays (see below), we found no reason to continue use of the Forte circuit.

The second circuit was that of Broek and Anderson⁽¹⁰⁾ wherein the discrimination pulses which gate the pulse height analyzer to admit neutron events are taken from the last dynode of the photomultiplier. The latter is operated with only a small accelerating potential between the dynode and the anode so that space charge limitation of the anode current occurs with large pulses. Under the proper conditions of operation proton-recoil scintillations will produce positive pulses at the dynode which will gate the pulse height analyzer. However, electron scintillations, in which the high-intensity, fast component of decay is of considerably greater importance, will produce only negative pulses there due to space charge repulsion and will thus be eliminated. The results obtained with this far-simpler circuit were found to be equal, or superior, to those with the Forte circuit in all respects, and so only those obtained with the former will be quoted.

Both discrimination circuits were tested with a Pu-Be neutron source and with monoergic D-D and D-T neutrons from a 250-keV Cockcroft-Walton. The accelerator-produced neutrons proved more satisfactory, owing to the simple, well-known^(3,10) shapes of the proton-recoil pulse-height spectra obtained with them. The losses in proton-recoil counting efficiency due to the action of each discrimination circuit were determined by comparing the pulse-height spectra found using the given circuit with those from the scintillation counter alone without discrimination, the neutron source being surrounded with lead sufficiently thick to effectively suppress all spurious gamma rays. The efficiency of each discrimination circuit in eliminating electron pulses was measured by comparing the spectra obtained in the presence and absence of intense Co^{60} and Ra^{226} gamma radiation.

Based on studies such as these, it appears that for proton recoils below 1 Mev in energy there is insufficient light output from a single scintillation of the given stilbene crystal to allow discrimination between proton and electron scintillations. Nevertheless, with the Broek and Anderson circuit it is possible to determine pulse-height spectra in the proton energy range 1 to 15 Mev with quite high discrimination against electron pulses and with only reasonable losses in the counting efficiency for proton-recoil pulses. However, this large range must be examined in two segments, each of which requires a different setup of the electronic system. The counter can be used in the range 1 to 10 Mev with the voltage between the anode and last dynode of the photomultiplier set at about +2 v and with a 10,000-ohm resistor shunted with a 50-mmfd capacitor serving as the load circuit of the last dynode. Under these conditions electron scintillations from Co^{60} gamma irradiation are at least 99.7 percent eliminated. The counting efficiency of the discrimination circuit for proton-recoil pulses is practically 100 percent at 1 Mev, but it decreases monotonically to about 10 percent at 10 Mev. Just above 10 Mev this efficiency undergoes a minimum and then rises very sharply with increasing energy, a phenomenon which is not well understood, but which serves to impose a practical upper limit on the range of the instrument under these conditions of operation.

By increasing the potential difference between the anode and last dynode to about +5 v, the last-dynode load capacitor to 1500 mmfd, and raising the discrimination level in the pulse-height-analyzer gate circuit to a point where protons of energy less than 3 Mev are eliminated, the minimum in the proton-recoil-pulse counting efficiency curve of the discrimination circuit can be pushed out beyond 15 Mev. The behavior at still higher energies has not been investigated for lack of suitable neutron sources. Under this new set of operating conditions the counting efficiency for proton-recoil pulses is about 25 percent at 15 Mev, again increasing monotonically with decreasing energy. It has not been possible to check the effectiveness with which electron pulses are eliminated at the high end of this range, owing to the lack of an appropriate gamma-ray source.

To summarize, the amplitude and even the shape of the counting efficiency curve of the pulse-shape-discrimination counter depend in a sensitive way upon the values chosen for the components in the electronic circuit. Hence, it will be necessary to determine this efficiency curve precisely with appropriately calibrated neutron sources before contemplating use of the counter for measurements of the cosmic-ray-produced neutron background.

References

1. R. Grismore, L. D. Marinelli, and B. G. Oltman. Radiological Physics Division Semiannual Report, January through June, 1958. ANL-5919, pp. 11-13.
2. R. Grismore, L. D. Marinelli, and B. G. Oltman. Radiological Physics Division Semiannual Report, July through December, 1958. ANL-5967, pp. 24-25.
3. R. Grismore, B. G. Oltman, and L. D. Marinelli. Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049, pp. 6-14.
4. W. N. Hess, H. W. Patterson, R. Wallace, and E. L. Chupp. The Cosmic-Ray Neutron Energy Spectrum, Lawrence Radiation Laboratory, Berkeley, California, Report, UCRL-8628 (February 6, 1959).
5. A. O. Hanson and J. L. McKibben. Phys. Rev. 72 673 (1947).
6. I. B. Berlman and L. D. Marinelli. Rev. Sci. Instr. 27 858 (1956).
7. I. B. Berlman, R. Grismore, and B. G. Oltman. Rev. Sci. Instr. 31 1198 (1960).
8. R. B. Owen. IRE Trans. Nuclear Sci. NS-5 198 (1958).
9. M. Forte. Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958. (Geneva: United Nations, 1958). Vol. 14, p. 300.
10. H. W. Broek and C. E. Anderson, Rev. Sci. Instr. 31 1063 (1960).

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

Roger Grismore and B. G. Oltman

The "twin" scintillation fast neutron detector^(1,2) is the principal spectrometer being used in the studies^(3,4) of the cosmic-ray-produced neutron background being carried out in this Laboratory. The operation of this detector is based on the differential response to neutrons of two scintillating solutions of equal electron but unequal hydrogen content. In order to obtain optimal neutron counting efficiency it is necessary to maximize in one solution and minimize in the other the ratio of protons to electrons (see Appendix). In addition, both the energy resolution and usable energy range of the spectrometer depend upon the light output obtainable from the poorest scintillator of the two solutions. Hence, the fluorescence yields of the solutions under consideration must also be taken into account while attempting to maximize the neutron counting efficiency.

The liquid scintillator solvents hereto employed by the present authors are xylene and 1,4-bis(trifluoromethyl) benzene (fluorinated xylene). Recently our interest in the possibility of improving the spectrometer's performance through the use of a new and different combination of solvents was renewed upon reading of the work of Williams and Hayes at Los Alamos.⁽⁵⁾ As one of the scintillator solvents in their detector they employed hexafluorobenzene, which contains no hydrogen at all and yet is comparable to fluorinated xylene in regard to light yield. While hexafluorobenzene is apparently unavailable, a check of the literature (see references in the Appendix) indicates that it should be possible to improve our combination of solvents with respect to either counting efficiency or fluorescence yield, and perhaps in both respects. However, it is difficult to determine the relative fluorescence yields of all the solutions of interest from the literature since all have not been measured by a single investigator, and conditions have varied greatly between experiments. Hence, an experimental program is in progress to measure the yields from various solutions which seem to hold promise. In this paper are reported the first results on several solvents which have relatively low proton-electron ratios but which might be expected to scintillate well with diphenyloxazole dissolved in them.

As measures of the relative light yields of the solutions we have determined the pulse heights at the peaks just below the Compton edges excited by Co⁶⁰ gamma radiation. The scintillation cell for these tests was made from the lower half of a 20-ml beaker, to the opening of which a quartz window was bonded with R-313.*

*Bonding Agent R-313, Carl H. Biggs Company, Los Angeles, California.

Filling of the cell was accomplished through a small-diameter glass tube fastened into the face opposite the quartz window. During the operation of the cell this tube was closed off with a Teflon stopper. All the outside faces of the cell were coated with Tygon paint* to provide a good diffuse reflecting surface. The quartz window of the cell was optically coupled to the photocathode window of a Dumont type 6292 photomultiplier tube with Dow Corning 200 Fluid of viscosity 12,500 centistokes. Return of the cell to the same position on the photocathode for all runs was assured by the use of a Teflon collar which held the cell and phototube together.

The electronic system used with the photomultiplier tube in the light-yield measurements consisted of a cathode-follower probe feeding a type A-61 amplifier and 256-channel pulse height analyzer. The zero intercept of this system and its gain as a function of input voltage to the cathode-follower probe were determined and frequently checked with a mercury-wetted relay type variable pulse generator, the pulse shape of which closely matched that from the photomultiplier tube. The linearity of the pulse generator amplitude with dial setting had previously been determined to be at least within $\pm 0.3\%$ of the largest pulse height involved over the experimental pulse height range. Long-range stability of the pulse generator amplitude was maintained by frequent potentiometer-checking of the charging voltage, readjusting the latter when necessary. The stability of the photomultiplier tube gain and the reproducibility of the optical contact between scintillation cell and multiplier photocathode were checked by means of a series of runs using a standard diphenyloxazole in xylene solution which were made before and after the regular series of measurements.

The light-yield measurements reported here were all carried out with 4 g/liter of diphenyloxazole dissolved in the solvents and no wave-shifting materials. The scintillation cell was thoroughly washed out before each run, finishing with two rinses with the solvent of the next solution to be tested. After this, the cell and solution were placed in a dry box containing a nitrogen atmosphere. In the dry box very pure nitrogen gas was bubbled through the solution for 35 minutes, and the cell was flushed out with the same gas. The cell was then charged with 7 cc of the bubbled solution, taking great care to make the amount as exact as possible, and was corked up, all before removal from the nitrogen atmosphere. The stated amount of solution was sufficient to bring the liquid level almost but not quite to the top of the cell. Following the insertion of the scintillating solution into the cell, the counting unit was quickly assembled and allowed to stand for 15 minutes for partial dark adaption of the photomultiplier tube. At the end of this time the counter was run under Co^{60} irradiation for a long enough time to obtain several thousand counts per pulse-height-analyzer channel at the peak below the Compton edge so that the position of that peak could be accurately determined. The same Co^{60} source and identical source-counter geometries were

*Tygon Paint, TP-61 White, Plastics and Synthetics Division, U. S. Stoneware, Akron, Ohio.

used in all runs, including the orientation of the photomultiplier tube relative to the earth's magnetic field. The photomultiplier voltage was set at 1025 for all the measurements and was frequently checked with a voltage divider-potentiometer arrangement.

The results of the first series of measurements, which proved to be reproducible to a high degree, are shown in Table 10. Included among the solvents for purposes of comparison are paraxylene and fluorinated xylene, each doubly-distilled. The Compton-peak pulse heights given in the table for the various solutions were corrected by the pulse generator calibration data for the electronic system. The probable errors quoted for these pulse heights are based on the reproducibility of the measurements, the uncertainties of the determinations of the centers of the Compton peaks, and the uncertainty of the pulse-generator calibration of the electronic system.

Table 10

Relative pulse heights from solutions of 4 g/liter diphenyloxazole in various organic solvents under Co^{60} gamma irradiation

Solvent	Chemical formula	Pulse height at Compton peak, arbitrary units
1,4-difluorobenzene	$\text{C}_6\text{H}_4\text{F}_2$	58.6 ± 0.7
1,3-bis(trifluoromethyl) benzene	$\text{C}_6\text{H}_4(\text{CF}_3)_2$	32.3 ± 0.8
α,α,α -trifluorotoluene (practical)	$\text{C}_6\text{H}_5\text{CF}_3$	41 ± 2
1,4-dimethylbenzene (paraxylene)	$\text{C}_6\text{H}_4(\text{CH}_3)_2$	163 ± 2
1,4 bis(trifluoromethyl) benzene (fluorinated xylene)	$\text{C}_6\text{H}_4(\text{CF}_3)_2$	48.1 ± 0.9

The measurements reported in Table 10 for 1,4-difluorobenzene, 1,3-bis(trifluoromethyl) benzene, and α,α,α -trifluorotoluene were all made with the solvents just as they were received from the supplier without further purification. This is probably satisfactory for the first two since these chemicals were obtained from L. Light and Co.* whose products have proved in the past to be of purity sufficiently high for scintillation counter use without further distillation. However, the α,α,α -trifluorotoluene, being of practical grade (and from a different supplier), should probably be tested again after distillation.

*L. Light and Co. Ltd., Colnbrook, Bucks, England.

It will be seen from Table 10 that some improvement in the light yield of the "twin" scintillation spectrometer could already be obtained by substituting 1,4-difluorobenzene for the fluorinated xylene without too great a loss in the proton-electron ratio. The next step in the experimental program will be to study some solvents with relatively high proton-electron ratios.

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, L. D. Marinelli, and B. G. Oltman. Radiological Physics Division Semiannual Report, January through June, 1958. ANL-5919, pp. 11-13.
4. R. Grismore, B. G. Oltman and L. D. Marinelli. Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049, pp. 6-14.
5. D. L. Williams, and F. N. Hayes. Liquid Scintillator Radiation Rate Meters for the Measurement of Gamma and Fast Neutron Rates in Mixed Radiation Fields. U.S. Atomic Energy Commission Report, LA-2375 (March 14, 1960).

APPENDIX: A FIGURE OF MERIT FOR LIQUID SCINTILLATORS
RELEVANT TO THE DETECTION OF NEUTRONS BY THE
"TWIN" SCINTILLATOR METHOD*

L. D. Marinelli

In the "twin" scintillator method, the signal Q due to the neutron component of mixed radiations impinging for a given time t on the detectors can be expressed as:

$$Q = \pi (N_{H'} - N_H) = \pi (N_{H'} - N_H) \pm \Gamma (N_e' - N_e) \quad (1)$$

where

$N_{H'}$ and N_H are the number of protons in solutions 1 and 2 respectively

$N_e' = N_e$ = the number of electrons present in them

π = the number of pulses elicited by the neutron component of the given radiation per hydrogen atom in the solution during time t

Γ = the corresponding number of pulses produced by the photon component per electron in the scintillator.

In the actual measurement the term $\Gamma (N_e' - N_e)$ is not exactly zero, because of the statistical processes involved. The square of statistical counting error ϵ of Q is given by the following expression:

$$\epsilon^2 = \pi (N_{H'} + N_H) + 2 \Gamma N_e \quad (2)$$

and the square of the relative error by the ratio of (2) to (1) squared; namely,

$$\epsilon^2 Q^{-2} = \pi^{-1} (N_{H'} + N_H) (N_{H'} - N_H)^{-2} + 2 \Gamma \pi^{-2} N_e (N_{H'} - N_H)^{-2} \quad (3)$$

Let us consider the proton-electron ratios p' and p in each solution, namely let us put:

$$N_{H'} = p' N_e \text{ and } N_H = p N_e \quad (4)$$

upon substitution in (3) we obtain:

*Appendix to paper by Grismore and Oltman, "Liquid Scintillator Solvent Study for the Purpose of Improving the "Twin" Scintillation Fast Neutron Detector."

$$\epsilon^2 Q^{-2} = \pi^{-1} N_e^{-1} (p' - p)^{-2} \{ p' + p + 2 \Gamma \pi^{-1} \} \quad (5)$$

As a figure of merit we may assume the reciprocal of the relative error for a given experimental condition, namely $Q\epsilon^{-1}$, which is:

$$Q\epsilon^{-1} = \pi^{1/2} N_e^{1/2} (p' + p + 2 \Gamma \pi^{-1})^{-1/2} (p' - p) \quad (6)$$

In this expression π and N_e have obvious significance, namely an increase in neutron flux and scintillator volume will increase the figure of merit and, hence, decrease the relative error; the first of these factors, however, is beyond experimental control and the second is subject to limitations imposed by difficulty of interpretation of the recoil spectrum; the latter increases with the increase in multiple scattering in the scintillator.⁽¹⁾ It is obvious that, other conditions being equal, the best experimental conditions are obtained by maximizing the expression:

$$(p' - p) (p' + p + 2 \Gamma \pi^{-1})^{-1/2} \quad (7)$$

Although the value of this expression is not entirely independent of radiation to be measured, it readily shows that the figures of merit to be maximized are:

$$(p' - p) (p' + p)^{-1/2}$$

where

$$\Gamma = 0 \quad ,$$

namely in the absence of gamma radiation,

and

$$(p' - p)$$

when gamma radiation is relatively so intense that

$$2\Gamma\pi^{-1} \gg p' + p \quad .$$

Values of p for some scintillation solvents (excluding the solute) are shown in Table 11.

Table 11

Compound	Formula	p	Relative pulse height* and references	
pentane	C_5H_{12}	0.286	-	-
hexane	C_6H_{14}	0.28	0.23(3)	0.106(5)
heptane	C_7H_{16}	0.276	0.28(3)	0.13(5)
octane	C_8H_{18}	0.272	(0.35?)	0.16(5)
mineral oil	$C_{12}H_{26}$	0.265	0.26(3)	0.10(5)
hexadecane	$C_{16}H_{34}$	0.261	(0.57?)	0.26(5)
cyclohexane	C_6H_{12}	0.250	0.27(3)	
methyl cyclohexane	C_7H_{14}	0.250	0.31(3)	
dicyclohexyl	$(C_6H_{11})_2$	0.234	0.44(3)	
decalin	$C_{10}H_{18}$	0.231	-	
triethylbenzene	$C_{12}H_{18}$	0.200	0.86(3)	
phenylcyclohexane	$C_{12}H_{20}$	0.183	0.91(3)	1.13(5)
xylene	C_8H_{10}	0.172	1.0(2,3,5,7)	
isopropyl byphenyl	$C_{15}H_{16}$	0.151	0.91(6)	
toluene	C_7H_8	0.150	0.89(3)	0.91(5)
benzene	C_6H_6	0.143	0.75(3)	
stilbene	$C_{14}H_{12}$	0.125	1.39(2)	
0-fluorotoluene	C_7FH_7	0.121	0.66(3)	
fluorobenzene	C_6FH_5	0.114	0.58(3)	
resorcin	C_6H_6	0.111	-	
anthracene	$C_{14}H_{10}$	0.106	1.7(2)	
2,5 difluorotoluene	$C_7F_2H_6$	0.091	-	
α - α - α trifluorotoluene	$C_7F_3H_5$	0.087	0.25(7)	
butadiyne	C_4H_2	0.077	-	
1,4 difluorobenzene	$C_6F_2H_4$	0.069	0.50(3)	0.36(7)
1,2,4 trifluorobenzene	$C_6F_3H_3$	0.0455	-	
fluorinated xylene	$C_8F_6H_4$	0.038	0.30(7)	
hexafluorobenzene	C_6F_6	0.000	0.23(4)	

*Pulse heights referred to xylene + 3.0 g/l. of 2,5 diphenyloxazole or 4 g/l. terphenyl.

A few comments on the value of $\Gamma\pi^{-1}$ are in order for the specific case of atmospheric neutrons at sea level. In the simplified case of thin scintillation solutions, where first collision phenomena predominate and carbon recoils are neglected, the following expressions hold

$$\pi = f_n \sigma_p t \text{ and } \Gamma = f_\gamma \sigma_e t \quad (8)$$

where f_n and f_γ are the neutron and gamma-ray flux respectively and σ_p and σ_e are the neutron-proton and total photon-electron scattering cross sections. For carbon the photoelectric effect is neglected because it is negligible (less than 1%) for gamma-ray energies greater than 100 kev. (8)

Since both f_n , σ_p and f_γ and σ_e are functions of neutron and gamma-ray energy, a strict evaluation of $\Gamma\pi^{-1}$ would require integral expressions involving the spectral distribution of the two radiations. For our present purposes, however, we shall consider average values of $\Gamma\pi^{-1}$ in the energy range 0.1 to 10 Mev, which should be correct within an order of magnitude. The value σ_p/σ_e is a slowly varying function of energy decreasing from about 30 at neutron and photon energies of 10 kev to about 10 at 50 Mev; we shall assume it constant and equal to 20. As to the neutron and gamma-ray fluxes to be encountered, one may assume for the 0.1 to 10 Mev range

$$\begin{aligned} f_n &\simeq 1.0 \text{ neutron cm}^{-2} \text{ min}^{-1} \quad (9) \\ f_{\gamma_1} &\simeq 1.5 \text{ cosmic-ray photons cm}^{-2} \text{ min}^{-1} \\ f_{\gamma_2} &\simeq 100 \text{ terrestrial photons cm}^{-2} \text{ min}^{-1} \end{aligned}$$

The value of f_{γ_1} was obtained from the values of Carmichael (cosmic-ray electron flux) (10) and the photon-electron ratio of about 3.0 in air computed by Poiani (11) and by Richards and Nordheim. (12) The value of f_{γ_2} was obtained by assuming the terrestrial flux at the earth's surface equal to one-half that at the center of an infinite concrete mass containing 1.0×10^{-12} c of radium per gram, (13) which, in turn, is in fair agreement with a background gamma-ray dose of 50 mrad year⁻¹ (14) if the average energy of the terrestrial photon is taken as 1.0 Mev. Substitution of these values in (8) gives:

$$\Gamma\pi^{-1} = \frac{100}{20} = 5 \quad (9)$$

in the region of the terrestrial photon energies (<3 Mev),

and

$$\Gamma\pi^{-1} = \frac{1.5}{20} \simeq 0.07 \quad (9')$$

for energies >3 Mev.

Before these figures can be applied to the estimate of the difference in the light pulse height distributions to be expected with the "twin" scintillators, notice must be taken of

- a) shape of the energy spectrum of background neutrons and gamma rays
- b) the difference in the light emitted by recoil protons and the electrons of the same energy.

As to the item (a), the expressions quoted in the literature are of the form

$$f_{\gamma}(E) = k_{\gamma} E^{-\gamma}$$

and

$$f_n(E) = k_n E^{-n}$$

where γ seems to vary from 1 to 2 and n between 1 and 1.4, according to the energy region, and the k 's are constants. For the purposes of the rough approximation sought here, we shall assume $\gamma \simeq n$ and $f_{\gamma}(E) = 100 f_n(E)$ in the low energy region and $f_{\gamma}(E) \simeq f_n(E)$ in the region above 3 Mev. As to the item (b), the ratio of the relative light efficiencies L_e and L_p we shall take $L_e L_p^{-1} \simeq 2$ to 5,⁽¹⁵⁾ the higher value applying to the lower energies. If to a first approximation we assume the proton and electron pulse height recoil spectrum to be rectangular in shape and of width equal to $L_e E_{\gamma}$ and $L_p E_n$, respectively, then for a particular channel in the pulse height scintillation spectrum we shall have a net neutron contribution:

$$f_n \sigma_p (L_p E_n)^{-1} N_e (p' - p) t = N_e \pi (L_p E_n)^{-1} (p' - p) \quad (10)$$

and a gamma ray contribution in each counter

$$f_{\gamma} \sigma_e (L_e E_{\gamma})^{-1} N_e t = N_e \Gamma (L_e E_{\gamma})^{-1} \quad (11)$$

the ratio of which is

$$\text{recoil proton/electrons} = \pi \Gamma^{-1} (p' - p) (L_e L_p^{-1})$$

The numerical value of this ratio for xylene and fluorinated xylene ($p' - p = 0.134$) for the values of $\pi \Gamma^{-1}$ as in (9) and (9') and for the values of $L_e L_p^{-1}$ mentioned above is of the order of 5 to 13%, a figure close to the one experimentally observed. It is obvious, therefore, that the net neutron signal is likely to be weak and that every effort should be made to raise it by increasing $(p' - p)$ as much as possible. The best practical combination available seems to be pentanehexafluorobenzene for which $p' - p = 0.286$; it remains to be seen whether the fluorescent efficiency of pentane is high enough for practical purposes.

At higher energies, in the absence of terrestrial gamma rays ($E_\gamma > 3$ Mev and $E_n > 6$ Mev), the photon/neutron flux ratio is sharply reduced, and consequently the proton recoils should represent a higher fraction of the light pulses. It is to be anticipated, however, that in the presence of nuclear reactions (n,p),(n, α), etc., the substitution of F for H is to be avoided, and the use of pure hydrocarbons is to be preferred although, in so doing, the value of $p' - p$ will decrease.

References

1. C. D. Schwartz and G. E. Owen. Recoil Detection in Scintillators, in Fast Neutron Physics, Part I. New York: Interscience Publishers, Inc., 1960, pp. 211-246.
2. D. G. Ott. In Liquid Scintillation Counting. New York: Pergamon Press, 1958, pp. 101-107.
3. F. N. Hayes, et al. Nucleonics 13(1) 46 (1955) and U.S. Atomic Energy Commission Report, LA-1639, p. 61.
4. F. N. Hayes, et al. U.S. Atomic Energy Commission Report, LAMS-2455, p. 130.
5. H. Kallman. Nucleonics 8(3) 32 (1951).
6. W. Buck and R. K. Swank. Rev. Sci. Instr. 29 252 (1958).
7. R. Grismore and B. Oltman, this report, p. 45.
8. G. W. Grodstein. X-Ray Attenuation Coefficients from 10 kev to 100 Mev. NBS Circular 583 (1957).
9. W. N. Hess, et al. The Cosmic-Ray Neutron Energy Spectrum, U.S. Atomic Energy Commission Report, UCRL-8628 (1959).
10. H. Carmichael. Energy Spectrum of the Soft Component Near Sea Level. Phys. Rev. 107 1401-1409 (1957).
11. G. Poiani. Sullo Sviluppo Della Componente Fotonica Nell'atmosfera. Nuovo Cimento II(5) 1114-1118 (1955).
12. J. A. Richards, Jr. and L. W. Nordheim. The Energy Distribution in Cosmic-Ray Showers. Phys. Rev. 74 1106-1113 (1948).
13. L. D. Marinelli, et al. Low Level Gamma-Ray Spectrometry: Experimental Requirements and Biomedical Applications, in Advances in Biological and Medical Physics, Volume VIII, ed. Tobias and Lawrence. New York: Academic Press, in Press
14. R. A. Dudley, Natural and Artificial Radiation Background of Man, in Low Level Irradiation, ed. A. M. Brues. AAAS Publication No. 59 (1959). pp. 7-31
15. R. K. Swank. Characteristics of Scintillators in Ann. Rev. Nuclear Sci. 4 111-140 (1954).

CORRELATION OF THE NATURAL RADIOACTIVITY OF THE HUMAN BODY TO THAT OF ITS ENVIRONMENT: UPTAKE AND RETENTION OF Ra^{226} FROM FOOD AND WATER

Henry F. Lucas, Jr.

I. Introduction

Measurements of the natural radium content of man are of great importance in studies to evaluate the effects of small increases in the radiation dose resulting from skeletally deposited radioelements for two reasons: one, our knowledge of long-term radiation effects is largely confined to the effect of radium; two, as a natural radioelement found in our food, water and air, its uptake and retention is a product of a life-long natural environmental condition.

The relative contribution of food and of water to the whole-body content is required for a valid estimate of the body contents of the large populations known to consume water with a high Ra^{226} content, on which direct measurement is not possible. This is important since 35,000 people are known to consume water from municipal systems with from 10 to 37 $\mu\mu\text{g/liter}$ Ra^{226} . An additional 800,000 people consume water with more than 1 $\mu\mu\text{g/liter}$, of which one-half or 400,000 consume water with from 4 to 10 $\mu\mu\text{g/liter}$.⁽¹⁾ These municipal waters were obtained from wells, some of which have been in use for 90 years. The Ra^{226} content of these waters can be compared to the currently accepted MPC for industrial workers of 100 $\mu\mu\text{g/liter}$ and of large populations of 10 $\mu\mu\text{g/liter}$.⁽²⁾

To evaluate the relative contribution of food and of water on the whole-body content of humans, early measurements were made on a relatively small number of living subjects by a considerable extension of the radon breath method of analysis.⁽³⁾ The living subjects measured in this work were tested in two experiments which were designed to determine: (1) the rate of Ra^{226} accretion by humans after reaching adulthood and (2) the relative accretion by two groups of youths whose drinking water contained 8 and 0.03 $\mu\mu\text{g Ra}^{226}/\text{liter}$ respectively.

In the first of these, prisoners at Stateville Penitentiary, Illinois who had previously resided in Chicago or other areas with surface or low radium water sources were chosen. The body contents of new prisoners were compared to those who had been at the prison for up to 22 years, consuming water at 4 $\mu\mu\text{g/liter}$. This experiment demonstrated an increase of body content with duration of exposure. The average body contents after 4 months and 20 years were 1.0 and 2.4×10^{-10} g respectively

Proper interpretation of these and the following measurements requires the evaluation of the rate of radium intake. Direct measurements of radium intake were not possible. It was, however, apparent that for intake at a steady state, excretion must be within a very few percent of intake or a much higher body content would have been found. Thus, excretion rates can be considered equal to intake rates. The accretion of Ra^{226} from water for the 20-year prisoners corresponds to 17 times the daily intake or that in about 50 liters of water. However, prison dietary conditions are not entirely normal, and intake of radium from water was unusually high. The average Ra^{226} intake corresponded to that from food plus that in 3 liters of water. This results, in part, from the extremely frequent occurrence of soups, stews and boiled foods. A normal population might be expected to have a daily intake equal to that from food plus that in 1 to 2 liters of water. The 20-year uptake for this normal group would still correspond to 17 times the daily intake. The body content, however, would only be equal to that in 20 to 30 liters of water.

In the second experiment, the accretion of radium by 8 high school boys from Lockport, Illinois, (average age 17.5 years; $8 \mu\mu\text{g Ra}^{226}/\text{liter}$ drinking water) was compared with that by 7 similar boys from Chicago ($0.03 \mu\mu\text{g Ra}^{226}/\text{liter}$). The average value for each group was 3.7×10^{-10} g and 0.4×10^{-10} g whole body, respectively. From measurements of the rate of excretion it was determined that the whole-body content of the Chicago boys corresponds to 22 times the daily intake of radium, which necessarily came from food. Similarly, the whole-body content of the Lockport boys corresponds to that from food plus that in 40 liters of water or about 45 times the daily intake.

In this present work, measurements are being made of the Ra^{226} content of normal human bone, teeth and soft tissue to determine for a normal population the relative contribution of food and water to the whole body content.

II. Experimental Method

Ashing Procedure. For this study normal, uninvolved bone obtained during ordinary surgical procedures or at autopsy was used. The samples were shipped immediately without any preservative, thereby avoiding possible contamination from the local water source. On receipt the samples were scraped to remove soft tissue, dried at 110°C and stored at -27°C until analysis.

Samples selected for analysis were prepared in one of two ways, depending on the number and type of analyses to be performed. For those on which the Ra^{226} content only was desired, the following procedure was used. The dried, weighed sample was placed in a small uncovered platinum

crucible or dish and ashed at 600°C overnight in an electric muffle furnace lined with Crystalon* to reduce the possibility of the contamination from fire brick. The weighed ash was then transferred to 50-ml centrifuge cones and dissolved in 1.85 cc concentrated nitric acid per gram of bone ash and diluted with distilled water to give a solution having 0.2 g ash/ml. With one exception no residue was observed.

Samples on which both Ra^{226} and Th^{228} were determined were given a preliminary treatment to remove blood residues by boiling 3 times in freshly distilled ethylene diamine and 3 times in distilled water; they were dried at 110°C, then ashed and dissolved as previously described above.

The thorium was extracted into dibutyl phosphate, while the aqueous solution containing the radium was transferred to an emanation flask and processed in the usual manner. No loss of radium or contamination was detectable.

Figure 15

Emanation flask

Measurement. The bone solution was transferred to an emanation flask of the type shown in Figure 15, using 1 N HNO_3 to a final volume of 300 ml. The solution was flushed with a radon-free air, sealed and allowed to stand for at least 7 days.

The sample flask was attached to the transfer system shown in Figure 16 with water traps (B) and radon trap (C) dry-ice cooled. The system, with the exception of the sample flask, was evacuated to a pressure of ca. 1 mm Hg. Aged tank air which had been dried** and passed through a charcoal trap at dry-ice temperatures for radon removal was then used to purge the transfer system and transfer the radon from the flask to the charcoal trap. It is unnecessary to heat the flask since quantitative (99.9%) transfer of the radon was effected in 13 liters of the 30-liter flush routinely used.

The charcoal trap containing the radon sample was evacuated while still at dry ice temperatures with a mechanical pump to about 0.1 mm Hg in 7 min or less and the radon transferred to the alpha scintillation counter

*Cat. No. 204 MC Lindberg Eng. Co. E. H. Sargent and Co.,
4647 West Foster Ave., Chicago, Illinois.

**Linde Molecular Sieve, Type SA, $\frac{1}{16}$ " pellets, Linde Co.,
230 N. Michigan Ave., Chicago, Illinois.

previously described.⁽⁴⁾ This system has a sensitivity of 5.40 cpm/ $\mu\mu$ c radon and a background counting rate of less than 0.1 cpm. Using this system, it was possible for one person to process 16 to 20 radon emanations per day. All radon samples were counted for 7 to 16 hours. Repeated determinations were made if the first two failed to agree within the 0.9 statistical counting error.

Figure 16

Radon collection system. A, Emanation flask; B, Dry-ice-cooled water freeze out traps; C, Dry-ice-cooled, charcoal-filled (7 g) radon trap; D, Flow rator, 0 to 2 l./min; E, Radon-free air input; F, Vacuum.

A system blank of $0.002 \pm 0.002 \mu\mu$ c Rn was found if the rubber tubing used in the Sigma Motor pump to transfer radon from the charcoal trap to the radon counter bottle was replaced at start of sample processing each day with radon-free (aged in sealed jar) tubing. Reagent and distilled-water blanks were found, with some exceptions, to be less than $0.002 \mu\mu$ g, and no corrections have been made except for samples of less than 1 g ash.

The alpha scintillation radon counting system had been calibrated previously with an aliquot from a National Bureau of Standards certified radium solution.⁽³⁾ Additional aliquots were evaporated on bone and, in eight measurements, $99 \pm 2\%$ of the radium was recovered, and thus, no correction for chemical recovery of radium from bone has been applied to the data.

III. Measurements

A. Uptake from food

To evaluate the effect of food on the content of radium in bone, subjects were selected who had lived all their lives in cities having surface water sources or wells with less than $0.1 \mu\mu$ g/liter Ra^{226} . The average concentration of Ra^{226} in bone ash from 42 individuals summarized in Table 12, is $0.0124 \mu\mu$ g/g ash. This corresponds to a whole skeletal content (2600 g ash) of 0.32×10^{-10} g. As has been found by other investigators^(5,6) from a random sampling of non-occupationally exposed humans, no correlation of radium content with age, sex, or illness was observed.

Table 12

Ra²²⁶ content of human bone from food sources*

Sample number	Number of cities of residence	Age, years	Sex	Bone	g ash	Ra ²²⁶ , $\mu\mu\text{g/g ash} \pm E^{**}$
185	4	56	M	rib	1 30	0 005 $\pm$ 0 007
163	2	49	M	rib	7 47	0 005 $\pm$ 0 001
213	2	56	M	rib	3 65	0 006 $\pm$ 0 003
156	2	55	F	rib	2 85	0 006 $\pm$ 0 003
130	3	35	M	rib	6 62	0 006 $\pm$ 0 001
244	2	34	M	rib	5 51	0 007 $\pm$ 0 002
212	3	21	F	rib	2 85	0 007 $\pm$ 0 003
100	1	25	M		3 17	0 008 $\pm$ 0 003
122	1	38	M	rib	6 11	0 009 $\pm$ 0 002
126	1	46	M	rib	9 29	0 009 $\pm$ 0 001
243	1	45	M	rib	3 64	0 009 $\pm$ 0 003
246	4	58	M	rib	3 44	0 009 $\pm$ 0 003
211	3	52	M	rib	2 96	0 010 $\pm$ 0 003
215	1	70	F	joint	6 24	0 010 $\pm$ 0 002
158	2	82	F	rib	1 55	0 010 $\pm$ 0 007
338	1	70	M	tibia	5 83	0 010 $\pm$ 0 001
339	1	65	M	tibia	4 06	0 010 $\pm$ 0 002
152	1	72	F	rib	0 87	0 011 $\pm$ 0 022
286	3	61	M	tibia	7 40	0 011 $\pm$ 0 002
245	2	61	M	rib	6 47	0 011 $\pm$ 0 001
170	5	69	M	rib	4 54	0 011 $\pm$ 0 003
121	3	33	M	rib	5 48	0 011 $\pm$ 0 001
125	2	57	M	rib	7 51	0 012 $\pm$ 0 001
132	1	26	M	rib	4 99	0 012 $\pm$ 0 003
282	3	50	F	femur	9 75	0 013 $\pm$ 0 001
171	1	47	M	rib	6 68	0 013 $\pm$ 0 002
370	1	23	M	femur	7 24	0 013 $\pm$ 0 001
230	2	50	F	vertebra	6 19	0 014 $\pm$ 0 002
142	3	36	M	rib	7 40	0 014 $\pm$ 0 002
209	2	49	M	rib	6 67	0 014 $\pm$ 0 002
168	1	73	F	rib	0 98	0 015 $\pm$ 0 007
103	6	30	M	tibia	12 32	0 016 $\pm$ 0 001
160	1	54	F	rib	3 41	0 016 $\pm$ 0 003
228	1	0	F	vertebra	0 53	0 016 $\pm$ 0 016
216	1	67	F	cort	6 16	0 016 $\pm$ 0 002
129	2	62	M	rib	8 83	0 017 $\pm$ 0 002
135	1	57	M	vertebra	2 92	0 019 $\pm$ 0 003
138	1	39	M	rib	4 84	0 019 $\pm$ 0 003
210	1	46	M	rib	3 41	0 020 $\pm$ 0 003
279	1	3	M	femur	1 68	0 021 $\pm$ 0 006
205	1	0	F	vertebra	2 29	0 022 $\pm$ 0 004
173	1	69	M	rib	1 71	0 027 $\pm$ 0 006
mean $\pm \sigma$						0 0124 $\pm$ 0 005

*Ra²²⁶ content of drinking water equal to or less than 0.1 $\mu\mu\text{g/liter}$ throughout entire lifetime

**90% confidence interval

Walton⁽⁷⁾ demonstrated that radium was distributed to within 15% of the average in each of 11 bones from 11 skeletons tested for variation. This information lends welcome validity to measurements of bone specimens as a means of estimating whole body content. However, as shown in Table 13 this is not always the case.

Table 13
Relative Ra²²⁶ content of human bone

Bone	Sample number					
	41A	41B	42A	42B	43A	43B
	Ra ²²⁶ , $\mu\mu\text{g/g ash} \pm E^*$					
Skull	0.070 $\pm$ 0.004	0.070 $\pm$ 0.004	0.100 $\pm$ 0.009	0.051 $\pm$ 0.002	0.028 $\pm$ 0.003	0.032 $\pm$ 0.003
Tibia	0.019 $\pm$ 0.003	0.014 $\pm$ 0.001	0.030 $\pm$ 0.002	0.025 $\pm$ 0.002	0.030 $\pm$ 0.002	0.026 $\pm$ 0.002
Jaw Bone	0.016 $\pm$ 0.002	-	0.025 $\pm$ 0.002	-	0.027 $\pm$ 0.003	-
Teeth	0.019 $\pm$ 0.003	-	0.026 $\pm$ 0.002	-	0.027 $\pm$ 0.003	-
Rib	0.012 $\pm$ 0.001	-	0.028 $\pm$ 0.002	-	0.030 $\pm$ 0.002	-
Birthplace	Nebraska		Connecticut		Florida	

*90% confidence interval.

In two of these three subjects the radium content is seen to vary significantly from bone to bone. No difficulties were encountered in analysis, and the differences in the duplicate analyses on sample numbers 41 and 42 are statistically significant. Since the radium content of individual bones has been found to vary considerably after single injections,⁽⁸⁾ it is probable that these differences represent changes in the environment. While the city of residence during their early years could not be determined, well waters with high radium content have been found by Barker⁽⁹⁾ in both Nebraska and Connecticut, and it may be presumed that these individuals consumed water of high radium content. These samples were provided by Dr. J. B. Hursh and while they were not a part of his earlier study,⁽⁵⁾ they have nearly identical Ra²²⁶ contents.

In order to extrapolate from bone to the whole-body content, measurements of radium in soft tissue are required. As shown in Table 14, the average radium content of soft tissue ash is 0.8 that of bone ash. This is in agreement with measurements by Radley, Turner and Mayneord⁽¹⁰⁾ and Hursh et al.,⁽¹¹⁾ but only about 3% of that found by Muth et al.⁽¹²⁾ Since the skeleton constitutes 86% of the whole-body ash, only 10 to 15% of the radium will be found in the soft tissues of the body.

The 0.04×10^{-10} g found in soft tissues added to the 0.32×10^{-10} g found for the skeleton gives a whole-body content of 0.36×10^{-10} g. This is in excellent agreement with the 0.4×10^{-10} g found previously by the

radon breath technique.⁽³⁾ Using the previously determined daily intake rate for humans in Chicago, Illinois, of $1.6 \mu\mu\text{g/day}$, this whole body content corresponds to 22 times the daily intake and necessarily comes from food.

Table 14

Ra²²⁶ content of human soft tissue

Sample number	Bone		Soft tissue			Ra ²²⁶ ratio Tissue ash Bone ash
	g ash	Ra ²²⁶ , $\mu\mu\text{g/g ash}$	g wet	g ash	Ra ²²⁶ , $\mu\mu\text{g/g ash}$	
216	6.16	0.016 ± 0.002	101*	0.794	0.020 ± 0.008	1.25 ± 0.42
338	5.83	0.015 ± 0.0015	412**	4.06	0.0069 ± 0.0025	0.66 ± 0.24
339	8.22	0.0129 ± 0.0010	409**	4.06	0.0096 ± 0.0015	0.74 ± 0.18
Weighted average						0.81

*muscle and fat

**muscle

B. Uptake from water

Measurements have been completed on 35 bones from individuals who lived either all their lives, or not less than the last thirty years, in a city with high radium drinking water. The radium content of these samples is summarized in Table 15. The radium content varied from a low of $0.012 \mu\mu\text{g/g ash}$ for a resident of a city with $0.13 \mu\mu\text{g/liter}$ in the drinking water to a high of $0.47 \mu\mu\text{g/g ash}$ in a city with $10 \mu\mu\text{g/liter}$ of water.

The correlation of the radium content of these samples with that in the drinking water is shown in Figure 17. For all data plotted in this figure it was assumed that $0.012 \mu\mu\text{g/g ash}$ was normally obtained from food and has been subtracted from the value found in Table 12. An excellent correlation of radium in bone with that in the water supply is obtained for drinking water with more than $1 \mu\mu\text{g/liter}$. For less than $1 \mu\mu\text{g/liter}$, normal statistical variations from food uptake overlap the uptake from water, and the scatter of points is not statistically significant. Extrapolating to a Standard Man (3000 g ash, whole body), uptake from water equal to that in 50 liters of drinking water would seem to be indicated as a suitable average.

Variations in sources of municipal water or its treatment have significant effects on the level of radium. The value reported for each city is the average of samples from all wells in use over the past 3 or more years. In two cases even this is not a very satisfactory method of

estimating the average radium content of the drinking water. In the first, City No. 13, Table 15, a water-softening system was installed two years ago, which removed 60 to 70% of the radium. In the second, City No. 11, Table 15, all of the wells in the Northeast part of the city have 2 to 4 times the radium content of those in the rest of the city. The radium content of water samples from the distribution system correlates with that of the nearest active well and, until a better estimate can be made, the average of the four samples from the distribution system will be used.

Table 15

Ra²²⁶ content of human bone from cities with high radium drinking water

City number	Drinking water, $\mu\text{g/l.}$	Age	Residence		Sample number	Bone	g ash	Ra ²²⁶ , $\mu\text{g/g ash}$	Mean, $\mu\text{g/g ash}$
			Years	City code					
1	0.13	9	9	KR08-4	280	femur	3.56	0.012 $\pm$ 0.003	-
2	0.2	72	43	2GN1RG1L-1	110	rib	2.46	0.015 $\pm$ 0.004	0.024
	"	36	35	"	109	rib	2.28	0.017 $\pm$ 0.005	
	"	0	-	"	229	vertebra	0.19	0.031 $\pm$ 0.037	
	"	0	-	"	240	"	1.27	0.032 $\pm$ 0.008	
3	0.4	11	11	2N0TS2P3P-5	384	rib	6.51	0.013 $\pm$ 0.002	-
4	0.48	12	12	13R02P-1	386	humerus	1.76	0.033 $\pm$ 0.006	-
5	0.85	75	75	R2TSB2W-2	284	femur	4.44	0.033 $\pm$ 0.005	-
6	1.8	12	12	IN0T160-5	283	"	4.69	0.020 $\pm$ 0.003	-
7	2.5	33	33	5R2P-1	172	"	1.04	0.055 $\pm$ 0.010	0.081
	"	87	63	"	145	"	2.38	0.082 $\pm$ 0.006	
	"	25	25	"	118	vertebra	6.68	0.105 $\pm$ 0.002	
	"	"	"	"	"	"	"	"	
8	3.1	65	65	1N0N36-5	281	talus	3.75	0.052 $\pm$ 0.005	-
9	4.6	64	33	161TT5-1	249	rib	1.97	0.044 $\pm$ 0.005	0.112
	"	68	68	"	175	"	1.30	0.058 $\pm$ 0.011	
	"	79	55	"	226	"	1.83	0.060 $\pm$ 0.007	
	"	65	30	"	275	"	2.04	0.094 $\pm$ 0.007	
	"	69	69	"	120	femur	4.88	0.100 $\pm$ 0.008	
	"	50	50	"	164	rib	2.09	0.118 $\pm$ 0.008	
	"	71	40	"	293	"	1.89	0.130 $\pm$ 0.012	
	"	44	44	"	201	"	2.57	0.147 $\pm$ 0.008	
	"	66	66	"	194	"	3.20	0.170 $\pm$ 0.007	
	"	47	34	"	235	"	2.57	0.200 $\pm$ 0.088	
10	5.0	66	66	TSR5HML2-1	124	iliac	5.10	0.078 $\pm$ 0.003	-
11	5.0	82	80	1R4R51-1	111	tibia	10.98	0.042 $\pm$ 0.002	0.069
	"	48	48	"	112	rib	2.09	0.047 $\pm$ 0.003	
	"	61	61	"	150	"	7.37	0.080 $\pm$ 0.002	
	"	72	55	"	165	"	2.23	0.083 $\pm$ 0.004	
	"	53	53	"	113	"	4.52	0.095 $\pm$ 0.003	
12	8.2	44	44	KNON3M-1	197	rib	1.60	0.056 $\pm$ 0.009	-
13	8.5	84	35	8BS2LG0-1	248	rib	1.87	0.042 $\pm$ 0.005	0.137
	"	54	54	"	169	"	2.23	0.137 $\pm$ 0.014	
	"	58	58	"	272	"	1.95	0.191 $\pm$ 0.016	
14	10.5	54	54	2LL3VN2RG-1	148	rib	4.05	0.472 $\pm$ 0.010	-

In addition, care must be taken to check as often as possible the actual level of radium consumed at work and at home. Home-type water softeners of the ion-exchange type removed approximately 98% of the radium. For instance, Subject No. 117 lived in a house in which both hot

and cold water were softened. The Ra^{226} content of $0.013 \pm 0.002 \mu\mu\text{g/g}$ ash found correlates with the $0.01 \mu\mu\text{g/liter}$ actually consumed and not with the $5 \mu\mu\text{g/liter}$ of the city supply.

Figure 17

Correlation of Ra^{226} in bone and drinking water. Solid circles represent individuals who have lived all their lives in a single city, with constant use of well water with high radium content. Open circles represent older life-long residents of a single city with exposure to high radium content drinking water for more than the last 30 years but not for their entire lives.

To evaluate the effect of age on whole-body content, the stability of the radium content of the water supply must be evaluated over the lifetime of the subjects. Since teeth have a very much smaller rate of turnover than bone, teeth from individuals of varying ages would give a good indication of the level of radium in the drinking water in past times. The radium content of teeth from life-long residents of the city (No. 9, Table 15) with $4.6 \mu\mu\text{g/liter}$ in its drinking water are summarized in Table 16. The average of $0.10 \mu\mu\text{g/g}$ ash is almost identical to the $0.11 \mu\mu\text{g/g}$ ash found for bone from residents from this same city. This may be fortuitous since, as shown in Figure 18, the radium content of both bone and teeth decrease with age.

Table 16

Ra^{226} in human teeth from city with $4.6 \mu\mu\text{g/l}$. Ra^{226} in drinking water

Sample number	Age	g ash	Ra^{226} , $\mu\mu\text{g/g ash} \pm E^*$
236	47	2.05	0.046 ± 0.010
68	57	1.35	0.060 ± 0.006
140	54	0.69	0.060 ± 0.012
137	29	0.93	0.069 ± 0.013
139	33	1.64	0.083 ± 0.007
136	35	1.53	0.085 ± 0.010
134	37	1.30	0.112 ± 0.011
70	26	1.52	0.147 ± 0.020
138	30	1.13	0.264 ± 0.015
Average			0.103

*0.9 statistical counting error.

This apparent decrease of radium content with age correlates with known changes in the water source for this city. Prior to 1896, all water came from springs or shallow artesian wells, which are presumed from current measurements of similar sources to be of low radium content, and it was not until 1922 that all of the water came from deep-well sources. Because of the small number of samples and the rather recent changes in the water source, no conclusion can be made about the effect of age on whole-body content. It would, however, appear that teeth are a sensitive indicator of drinking water radium content in the past and can be used to estimate the radium content of populations on which other methods are not feasible.

Figure 18

Variation in Ra^{226} content of bone and teeth with age and drinking water source.

IV. Summary

Measurement of the skeletal and soft tissue contents have been made on normal humans whose primary source of radium was their food. An average whole-body content of 0.36×10^{-10} g is inferred from these measurements. This corresponds to a whole-body uptake equal to 22 times the daily intake. In agreement with other investigators on non-occupationally exposed humans, no correlation with age was noted.

For lifetime uptake, accretion of radium by bone was found to correlate well with that in the drinking water, and the whole-body content for a standard man was found to correspond to that contained in about 50 liters of water. An attempt was made to evaluate the effect of age on whole-body content but because of the small number of samples and recent changes in water source, no conclusion can be made at this time. Teeth appear to be a useful indicator of the radium content of populations on which other methods are not feasible.

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References

1. H. F. Lucas and D. P. Krause. Preliminary Survey of Ra^{226} and Ra^{228} (MsTh_1) Contents of Drinking Water. *Radiology* 74 114 (1960).
2. National Bureau of Standards Handbook 69, Maximum Permissible Body Burden and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure.
3. A. F. Stehney and H. F. Lucas, Jr. Studies on Radium Content of Humans Arising from Natural Radium of Their Environment. Peaceful Uses of Atomic Energy: Proceedings International Conference, Geneva, August, 1955. New York: United Nations 11 49-54 (1956).
4. H. F. Lucas. Improved Low-Level Alpha Scintillation Counter for Radon. *Rev. Sci. Instr.* 28 680-683 (1957).

5. J. B. Hursh and A. A. Gates. Body Radium Content of Individuals with No Known Occupational Exposure. *Nucleonics* 7 46-59 (1950).
6. R. F. Palmer and F. B. Queen. Normal Abundance of Radium in Cadavers from the Pacific Northwest. *Am. J. Roentgenol.* 79 521-529 (1958).
7. A. Walton, R. Kologrivov and J. L. Kulp. The Concentration and Distribution of Radium in the Normal Human Skeleton. *Health Physics* 1 409-416. (1959).
8. W. P. Norris, T. W. Speckman and P. F. Gustafson. Studies of the Metabolism of Radium in Man. *Am. J. Roentgenol.* 79 785-802 (1955).
9. F. B. Barker. Personal communication.
10. R. C. Turner. Personal communication to L. D. Marinelli.
11. J. B. Hursh, A. Lovaas and E. Biltz. Radium in Bone and Soft Tissues of Man. U. S. Atomic Energy Commission Report, UR-581 (1960).
12. H. Muth, B. Rajewsky, H. J. Hantke and K. Aurand. The Normal Radium Content and the $\text{Ra}^{226}/\text{Ca}$ -Ratio of Various Foods, Drinking Water and Different Organs and Tissues of the Human Body. *Health Physics* 2 239-245 (1960).

CRITIQUE ON THE HALF-LIVES OF LEAD AND RaD IN THE HUMAN BODY

Richard B. Holtzman

I. Introduction

In order to evaluate the possible sources of the stable and radioactive lead, Pb^{210} (RaD), found in the human body it is necessary to know the rate of intake and residence time of lead in the body. For the sake of comparison to the data given in the Report of Committee II of the International Commission on Radiological Protection (ICRP) ⁽¹⁾ and for ease of calculation, a single exponential model is used in which

$$T_b = \frac{0.693 \text{ mC}}{If_w} \quad (1)$$

where

T_b = the biological half-life in days of the element of reference.

m = mass in grams of the organ of reference.

C = concentration in grams per gram of tissue of the element or nuclide of reference.

I = daily intake in grams of the element or nuclide of reference.

f_w = fraction of the daily ingested intake retained by the organ of reference.

In the case of a radioactive nuclide, the effective half-life T_e is

$$T_e = \frac{T_b T_r}{T_b + T_r} \quad (2)$$

where

T_r = radioactive half-life.

In cases where the fraction of the total body content of a material in a given tissue is not known, this fraction may be approximated by calculation as follows: If T_e^x is the effective half-life of the nuclide in tissue "x", $m^x C^x$ is the amount of the nuclide in the tissue, and f_w^x is the fraction of I going to the tissue, then the fraction of the total body content of the nuclide in the organ, f_2^x , at equilibrium, can be calculated from the exponential model ⁽¹⁾ as

$$f_2^x = \frac{m^x C^x}{mC} = \frac{f_w^x T_e^x}{f_w T_e} \quad (3)$$

where the symbols without superscripts refer to the properties in the total body.

These equations make possible the calculation of T_e , T_b , and the various values of f where some, but not all the values, are known from direct measurements. Since lead has been studied extensively, one can determine these parameters for the stable element and apply them to a radioactive isotope of lead, such as RaD.

In order to calculate T_b from Equation (1) it is necessary to determine the values of m , C , I , f_w , and f_2' , where the latter is the fraction of lead going from the blood to the organ of reference. Knowledge of f_2' is often necessary because the fraction of lead going from the alimentary canal to the organ is not always measured directly. The fraction to the organ, f_w , is then the product of the fraction f_1 from the alimentary canal to the blood, and the fraction from the blood to the organ; namely $f_1 f_2'$. In the following an analysis is made of data available in the literature, and certain values of these parameters are chosen before embarking into a calculation of T_b or T_e .

II. Survey and Choice of Metabolic Parameters

m. The masses of the various organs, m , are taken from the "Standard Man" of Lisco⁽²⁾ (also given in Table 8 of the ICRP report⁽¹⁾). However, since the mineral contents of various types of bones in a single individual correlate better with the ash content than with the wet bone weight, the ash content of the skeleton is used here and taken as 2800 g. The ash contents of trabecular (based mainly on data taken from ribs) and cortical bones are assumed to be 21 and 60% of the wet weights, respectively. The use of ash weight, rather than wet weight, is essential since the samples measured by Tipton,⁽³⁾ based on an average ash content of 21%, appear to be heavily weighted towards trabecular bone. Thus, if lead content is proportional to calcium content, much too low a value for the total lead content of the body would result if measurements were based exclusively on wet weight.

C. The concentrations of lead in soft tissues are taken mainly from the data of Tipton et al.⁽³⁾ and Table 7 of the ICRP report⁽¹⁾ as is the lead content of the skeleton as shown in Table 17. The average lead concentration of wet bone is divided by the average percent ash content given by Tipton to obtain the concentration per gram of ash. This, weighted by the number of samples, gives an average value of $33.0 \mu\text{g}$ of lead/g ash, which, in a 7000-g skeleton, is equivalent to $13.2 \mu\text{g/g}$ wet weight, a value twice that used in Table 7 of the ICRP report.⁽¹⁾ The earlier data of Kehoe⁽⁴⁾ shown in Table 18 for the ribs and femurs from 8 unexposed persons, when corrected for the ash fractions of rib and femur, given above, show averaged results identical with those of Tipton et al.⁽³⁾

Table 17

Concentration of lead in the human skeleton, taken from the data of Tipton et al.⁽³⁾

Report No. (ORNL-CF-)	Lead in wet bone, $\mu\text{g/g}$ wet bone	% Ash, avg	C_i Lead in ash, $\mu\text{g/g}$ ash	n_i Number of samples	$C_i n_i$ $\mu\text{g} \times \text{number}$ of samples
56-3-60	4.2	17.6	24.0	23	552
57-11-33	7	21	33.3	31	1036
57-2-3	7.6	15.6	48.7	22	1071
57-2-4	7.4	28	26.5	23	610
		21.1 (Average)		99	3269

$$\text{Average Lead Content} = \frac{\sum_i C_i n_i}{\sum_i n_i} = \frac{3269}{99} = 33.0 \mu\text{g/g ash}$$

Table 18

Concentration of lead in human rib and femur from data of Kehoe⁽⁴⁾

Donor age, years	Concentration of lead, mg/100 g wet	
	Rib	Femur
3	-	2.22
6	1.02	1.14
20	1.11	-
21	0.47	-
64	-	0.80
70	0.39	3.59
75	0.60	2.89
95	0.56	1.36
Average/g wet weight	0.69	2.00

Average/g ash*: Rib: $0.69/0.21 = 32.9 \mu\text{g/g ash}$ Femur: $2.00/0.6 = 33.3 \mu\text{g/g ash}$

* Assume ash contents of 21 and 60% for rib and femur, respectively.

The total body content (mC) based on the above considerations for m and C, as measured in the major tissues, results in 111 mg of lead, as shown in Table 19.

Table 19
Content of lead in various tissues of the human body

Tissue	Fraction of total body weight, ⁽²⁾ %	Weight of tissue, ⁽²⁾ g (m)	Lead concentration,* ⁽¹⁾ $\mu\text{g/g}$ wet weight (C)	Lead content,* mg (mC)	Fraction of total body lead content % (i_2)
Muscle	43	30,000	0.17	5.1	4.6
Skin and subcutaneous tissue	87	6100	0.55	3.4	3.1
Fat	14	10,000	(0.2)	(2.0)	(1.8)
Bone	10	7000 2800 g ash	33 [†]	92.5	83.2
Red marrow	2.1	1500	(0.2)	(0.3)	(0.3)
Yellow marrow	2.1	1500	(0.2)	(0.3)	(0.3)
Blood	7.7	5400	0.3	1.6	1.4
Gastrointestinal tract	2.9	2000	0.3	0.6	0.5
Liver	2.4	1700	2	3.4	3.1
Brain	2.1	1500	(0.4)	(0.6)	(0.5)
Lungs	1.4	1000	0.68	0.7	0.6
Lymphoid tissue	1.0	700	(0.2)	0.1	0.1
Kidneys	0.43	300	1.24	0.3	0.3
Heart	0.43	300	0.2	0.1	0.1
Spleen	0.21	150	0.67	0.1	0.1
Total body	100	70,000	1.58	111.1	100.0

* Data in parentheses are maximum limits given in References 1 and 3.

[†] Bone concentration given in units of $\mu\text{g/g}$ ash. (See text.)

I. At equilibrium, the daily intake acquired through alimentary and respiratory absorption is equal to the daily urinary and fecal excretions. Alimentary intake, as represented by the lead content of the food and feces, based on the data of Kehoe⁽⁴⁾ and Cholak and Bambach⁽⁵⁾ is about 0.30 mg/day. However, an additional 9% should be added to compensate for the removal of lead by absorption from the alimentary canal, a figure based on the total lead balance of one individual for a period of 1.8 years measured by Kehoe et al.⁽⁶⁾ Hence, we shall use the value 0.33 mg/day for the daily alimentary intake.

Absorption of lead into the body from inhalation is more difficult to estimate because of the sharp variability of atmospheric lead content. The U.S. Public Health Service, through its national network of air pollution measuring stations,⁽⁷⁾ found an average lead concentration in air of $1.4 \mu\text{g}/\text{m}^3$ in urban areas, with Los Angeles reporting the highest average, $5.2 \mu\text{g}/\text{m}^3$. Non-urban and suburban stations have reported concentrations of $0.3 \mu\text{g}/\text{m}^3$. These may be compared to the 1941 Cincinnati value of $5.1 \mu\text{g}/\text{m}^3$ reported by Kehoe⁽⁴⁾. The somewhat high average value of $3.0 \mu\text{g}/\text{m}^3$ found in the cities with populations over 2 million,⁽⁷⁾ will be used for the calculations.

If the inhalation rate of $20 \text{ m}^3/\text{day}$ is used for man,⁽¹⁾ about $0.06 \text{ mg}/\text{day}$ of lead is inhaled, a value in fair agreement with the $0.08 \text{ mg}/\text{day}$ estimated by Monier-Williams⁽⁸⁾ in England. In order to estimate the amount of lead actually absorbed into the body from the air, a modified form of Equation 46 in the ICRP report⁽¹⁾ will be used

$$f_a = R(1/3 + 2/3 f_1) f_2' \quad (4)$$

where

f_a = fraction of the material taken into the body by inhalation that arrives at the organ of reference.

R = fraction of material retained in respiratory system from inspired air.

f_1 = fraction of the material taken into the alimentary canal that enters the blood.

f_2' = fraction of the material in the blood that enters the organ of interest.

This equation is based on the assumption that the respiratory tract removes a fraction R of the material from the air, of which one-third of a soluble retained material is absorbed directly into the body [lead is thought to be quite soluble in the environment of the lung⁽⁹⁾] and the remaining two-thirds enters the gut where a fraction f_1 is absorbed. Calculations based on this formula with the ICRP value of $R = 75\%$ show 29% or $0.017 \text{ mg}/\text{day}$ absorbed into the body through this route. This value appears to be somewhat high, as might be expected of the ICRP, an organization which, in the interests of safety, would favor the use of higher values of retention which are likely to be attained in industrial exposures. However, much of the available evidence indicates that under average conditions the retention R is likely to be between 15 and 50% ; thus the literature surveys of Shapiro⁽¹⁰⁾ and Morrow et al.⁽¹¹⁾ show a minimum retention of 15 to 40% in the range of particle sizes 0.1 to 0.5 microns; Jacobs⁽¹²⁾ states that most particles in outdoor air are in the 0.3 - to 1.1 -micron diameter range, with an average of about 0.6 microns. The results of Bale and Shapiro⁽¹³⁾ show 25% retention of radon decay products in human subjects breathing room air of unknown particulate content. More recently Aurand et al.⁽¹⁴⁾ reported similar results (30% retention)

in naturally occurring atmospheric radon daughters. Considering that lead may distribute somewhat differently on dust than do radon daughters, a retention factor of 40% will be used.

Using the above assumptions one obtains an absorption of lead from air in urban areas of 0.009 mg/day. Addition of this value to the amount absorbed from food (that is 0.33 mg/day with an absorption coefficient of 0.08) namely 0.026 mg/day, leads to a value for the total absorption of 0.035 mg/day, which is in excellent agreement with the average observed urinary lead excretion of 0.037 mg/day (see Table 20) based on an average concentration of 0.026 mg/day of urine^(4,5) and an excretion rate of 1.4 liters/day.⁽¹⁾ The daily urinary concentration represents also the total lead excretion from the body since in humans little or no lead is excreted through the gut.⁽⁶⁾ An indirect confirmation that the air contribution to the total absorption is likely to be small can be had by comparing the daily urinary lead output of 0.037 mg from city dwellers breathing a high lead-content air (Germans, French, and Americans) with the 0.031 mg from rural-dwelling Mexican Indians breathing air of low-lead content.⁽⁴⁾

Table 20

Daily lead intake and excretion in humans

Source of data	Sample	Excretion or intake, mg/day	Average excretion or intake, mg/day
Kehoe ⁽⁴⁾	Food, 3 subjects	0.28	0.30
	Fecal, 3 subjects	0.32	
	Average fecal samples, 10 American cities		0.398
Cholak and Bambach ⁽⁵⁾	Feces, 1000 samples	0.270 ± 0.145	0.30*
	Feces, 4 subjects	0.322	
Kehoe ⁽⁴⁾	Urine,** 188 samples	(0.026 mg/l.) 0.036	0.037*
Cholak and Bambach ⁽⁵⁾	Urine,** 1000 samples	(0.027 ± 0.014 mg/l.) 0.038	

*Estimated over-all average

**Assume 1.4 l./day, ICRP⁽¹⁾

Because of the smallness of the inhalation contribution and in order to proceed with the calculations of T_b by means of Equation (1), the total daily absorption will be assumed to be entirely due to alimentary intake, the magnitude of which, I, can be best obtained from the ratio of the daily

urinary output, 0.037 mg, to the fraction of the intake absorbed, $f_1 = 0.08$ (vide infra): this leads to $I = 0.46$ mg/day.

f_1 . The fraction of the ingested lead entering the blood through the alimentary canal, f_1 , neglecting the poorly known inhalation value, is the ratio of urinary to fecal contents: $f_1 = 0.037/0.33 = 0.11$. However, by accounting for the atmospheric lead contribution of 0.009 mg/day, we obtain $f_1 = 0.08$, in good agreement with Kehoe et al.⁽⁶⁾

In order to maintain maximum consistency with the ICRP report⁽¹⁾ ($f_1 = 0.08$ and $I = 0.4$ mg/day), we will use $f_1 = 0.08$ and the previously estimated $I = 0.46$ mg/day.

f_2' . Because of lack of data on human subjects, the fraction of lead entering the organ from the blood will be taken from the data of Hamilton⁽¹⁵⁾ and Durbin et al.⁽¹⁶⁾ in which Pb^{203} was injected into rats, and the distribution in the tissues was determined after 1 day. The various values of f_w and f_2' in the rat are given in Table 21.

Table 21

Values of f_w , f_1 and $f_2'^*$

Alimentary canal to	
Blood	$f_1 = 0.08^*$
Bone	$f_w = 0.022$
Liver	$f_w = 0.0064$
Kidney	$f_w = 0.013$
Blood to	
Bone	$f_2' = 0.28$
Liver	$f_2' = 0.08$
Kidney	$f_2' = 0.14$

* All data, except f_1 , obtained in rats.^(15,16)

Data on f_1 from Kehoe et al.⁽⁶⁾ (Human subject).

III. Calculation of Half-Lives and Distributions of Lead and RaD in the Body

The results of calculations of T_b and f_2 for lead, and the derived T_e and f_2 for RaD (assuming $T_r = 7100$ days) are shown in Table 22. These calculations using Equations (1), (2), and (3) were made for each of four organs, total body, skeleton, liver, and kidneys and are based on: (a) the values of the "metabolic" parameters m , C , I , and f_w as selected above,

Table 22
Half-lives and f_2 of lead and RaD in the human body and selected tissues

Tissue	Source of data	m (mass of tissue), g	c (conc. of Pb), $\mu\text{g/g}$ wet weight(a)	I (daily ingestion rate), mg/day	f_w (fraction of ingested Pb to tissue)(a)	$T_b(\text{Pb})$, days (Eq. 1)	$T_e(\text{RaD})$, days ($T_r = 7100$ Eqs. 1 and 2)	f_2 (calculated fraction of total Pb in tissue, Eq. 3)	f_2 (measured fraction of total Pb in tissue)(1,3)	f_2^* (calculated fraction of total RaD in tissue, Eq. 3)	f_2^* (measured fraction of total RaD in tissue, this laboratory)
Total body	This work	7×10^4	1.59	0.46	0.08	2085	1612	1	1	1	1
	ICRP										
	Calc. Eq.(1)	7×10^4	1.1	0.4	0.08	1670	1350	1	1	1	1
	Tabulated	7×10^4	1.1	0.4	0.08	1460	1200	1	1	1	1
	Sultz and Hursh(17)										
	Given	125 mg		0.087		990	870	1	1	1	1
	Revised	125 mg		0.037		2240	1700	1	1	1	1
Skeleton	This work	2800 (ash)	33 (ash)	0.46	0.022(b)	6350	3350	0.84	0.83	0.57	0.63
	ICRP										
	Calc. Eq.(1)	7000	6.6	0.4	0.02(b)	3650	2400	0.55	-	0.44	-
	Tabulated	7000	1	0.4	0.02(b)	3650	2400	0.7	-	0.55	-
Liver	This work	1700	2	0.46	0.0064(b)	800	719	0.031	0.031	0.036	0.017
	ICRP										
	Calc. Eq.(1)	1700	2	0.4	0.0064(b)	920	830	0.044	-	0.049	-
	Tabulated	1700	2	0.4	0.0064(b)	1947	1500	0.1	-	0.1	-
Kidney	This work	300	1.24	0.46	0.011(b)	59.8	59.7	0.0039	0.0036	0.0051	-
	ICRP										
	Calc. Eq.(1)	300	0.14	0.4	0.01(b)	7.3	7.3	6.4×10^{-4}	-	7.5×10^{-4}	-
	Tabulated	300	0.14	0.4	0.01(b)	531	495	0.05	-	0.06	-

^aConcentration given "per g ash" under skeleton, this work as noted.

^bThe data for f in the skeleton, liver and kidneys are the products of the fraction of the lead intake absorbed into the body, f_w and f_2 the fraction going from the blood to the tissue taken from the data on rats.(15,16)

(b) values of these parameters stated in Table 12 of the ICRP report, and (c) the estimates of Sultz and Hursh.⁽¹⁷⁾ Also in Table 20, the values of T_b and T_e and f_2 explicitly stated in Table 22 of ICRP report are included for comparison.

Total Body. Under this section, Table 22, it is apparent that the value of T_b for lead stated in Table 12 of the ICRP report (1460 days) is inconsistent with Equation (1) and the values of m , C , I , and f tabulated therein; apparently it should have been 1670 days instead. We are unable to ascertain whether this discrepancy is the result of typographical or other error. The values of the concentration, C , and the intake, I , used in this paper lead to an increase in half-life, T_b , to 2085 days, 45% greater than the tabulated ICRP value.

The value of T_e of RaD, evaluated by means of Equation (2) becomes 1612 days on the basis of our assumptions as contrasted to 1200 days as tabulated in the ICRP report.

Also presented under "Total body" are the calculations of lead excretion from the human taken from the studies of Sultz and Hursh⁽¹⁷⁾ on Po^{210} content of the urine of miners exposed to radon. In their work they assumed a much greater urinary lead excretion (0.087 mg/day) than was observed elsewhere;^(4,5) revision of their estimate to our value of 0.037 mg/day increases T_b to about 2000 days, as estimated in this paper. They also estimated T_b to be 840 days from the data of Kehoe et al. (p.75),⁽⁶⁾ based on the decrease of lead content in the body of one subject over a period of 1.2 years after the discontinuance of lead administration in excess of the normal intake. From the source paper we have derived values of 750 to 1200 days, depending on the points chosen, the higher values resulting from data taken at later times. This indicates an increase of T_b with time, which appears to approach the calculated long-term value of about 2000 days, and it raises the question as to whether the single exponential function of excretion postulated for lead in the body might better be replaced by a power function. However, because of the uncertainties involved in these calculations [estimation of the factors m , C , I , and f in Equation (3)] and the lack of rigorous data necessary to determine the power-function parameters, it is felt that little would be gained by such a procedure at this time.

Skeleton. The skeletal lead concentration stated in Table 12 of the ICRP report, namely $C = 1 \mu\text{g/g}$ wet weight, is probably a misprint since the value of T_b stated therein is consistent with Equation (1) and a skeletal lead concentration of $6.6 \mu\text{g/g}$ wet weight as given in Table 7 of that report. However, according to our estimate, the value of $C = 33 \mu\text{g/g}$ ash should be used, resulting in a half-life T_b of 6350 days.

The fraction of the total body lead, f_2 , residing in the skeleton given in Table 12 of the ICRP report, 0.7, is apparently derived from the concentration of lead in the various organs listed in Table 7 of that report.

However, this fraction becomes 0.55 when calculated with the aid of Equation (3) from the data in Table 12 of the report, except for the skeletal concentration which should be $6.6 \mu\text{g/g}$ instead of $1 \mu\text{g/g}$.

Our choice of $33 \mu\text{g/g}$ ash as the typical concentration in human bone, when used in Equation (3), leads to a value of $f_2 = 0.84$, and this value is in good agreement with our calculation of $f_2 = 0.83$ from the experimental data gathered in Table 19 of this paper.

The half-life T_e of RaD, based on the T_b of skeletal lead and Equation (2) is 3350 days. The fraction of total body RaD in the skeleton, as calculated in this paper, is 0.57, in fair agreement with the measured value of 0.63 in Table 23 of this paper if consideration is given to the large errors in some of the measurements and the relatively drastic assumptions used.

Table 23

Distribution of RaD in various human tissues*

Tissue	Fraction of body weight of organ % (1)	Sample Number	Concentration of RaD, $\mu\mu\text{c/g}$ **	Fraction of total activity, %
Liver	2.4	270L	0.013 ± 0.002	1.7
		271L	0.0041 ± 0.0017	
		269L	0.018 ± 0.002	
		292L	0.0093 ± 0.0021	
		Average	0.011 ± 0.003	
Muscle	43	269M	0.0022 ± 0.0006	17
		292M	0.0010 ± 0.007	
		Average	0.006 ± 0.004	
Bone (rib)				
Ash	4.0	292B	0.238 ± 0.029	63
Total	10	268B	0.232 ± 0.021	
		Average	0.235 ± 0.036	
Other tissues	45		$0.006^\dagger$	18

* Data taken in this laboratory

** Concentration is given per gram of wet tissue, except bone which is given per gram of ash.

† Other tissues assumed to have same average concentrations as muscle.

Liver. The half-life for lead in the liver (1947 d), as given in Table 12 of the ICRP report, is also inconsistent with Equation (1) and the values of the pertinent parameters as calculated from Table 7 therein. The half-life of 800 days in the present paper differs from the revised ICRP value of 920 days only because of the 15% larger value of the daily intake, I , assumed in this paper.

The fraction of the total body lead in the liver, 0.1, given in the ICRP report, Table 12, is at variance with the value of about 0.05, which we have obtained by using the data in Table 7 of the same report. The value calculated from the ICRP results is 0.044 (using $T_b = 920$ days), which is somewhat greater than 0.031, the calculated (Table 22) and measured (Table 19) value of this paper.

The half-life of RaD in the liver, T_e , is 719 days as determined from the above T_b of 800 days and Equation (2) which may be compared to the revised ICRP half-life, T_e , of 830 days based on a T_b of 920 days. The fraction f_2 of the total body RaD in this organ is given as 0.1 in Table 12 of the ICRP report; however, a calculation based on the 920-day lead half-life in the liver, a 1670-day half-life in the body and Equation (3), leads to $f_2 = 0.049$. The value of 0.036 calculated from the parameters chosen in this paper is at variance with the measured value of 0.017 in Table 23. However, as discussed under "Skeleton", this disagreement cannot be considered too serious with the scant data available.

Kidneys. The half-life of lead in the kidneys, $T_b = 60$ days, based on the data selected in this report, is about 10% of that tabulated in Table 12 of the ICRP report. A recalculation of T_b using the given parameters, m , C , I , and f stated in Table 12 of the latter report brings the half-life to 7.3 days. The difference between the half-life determined in this report and the revised ICRP value is due mainly to the choice of the $1.24 \mu\text{g/g}$ concentration given in Table 7 of the latter report as opposed to the value of $0.14 \mu\text{g/g}$ used in Table 12 of that report.

The fraction of total body lead in the kidneys $f_2 = 0.05$ given in Table 12 of the ICRP report, is an order of magnitude larger than given in Table 19 of this paper, whereas that calculated from the revised ICRP results ($T_b = 7.3$ days) and Equation (3) is 6.4×10^{-4} . On the other hand the value of f_2 calculated in this paper (0.0039) agrees well with the experimental value of 0.0036 (Table 19 this paper).

The half-life of RaD in the body, T_e , is 60 days calculated from Equation (2) and a T_b for lead of 60 days.

The fraction of total body RaD in the kidneys is given as 0.06 in Table 12 of the ICRP report, but the calculated value using the tabulated data in that table and a T_b of 7.3 days is 7.5×10^{-4} . On the other hand, use

of the data estimated in this paper and Equation (3) gives a value of 0.0036; no experimental values are available for comparison.

IV. Conclusion

The use of the higher skeletal lead content which we have derived from the literature and, consequently, the greater total body content results in half-lives of lead and RaD in the body and skeleton longer than those previously estimated.^(1, 17) As a result, the internal consistency of the data is improved and the calculated fractions of total body lead in the various organs agree with those measured by Tipton et al.⁽³⁾ This consistency indicates also that the f_2^I values obtained from measurements on rats may apply to the lead metabolism in humans. The information on the RaD distribution is in reasonable, but somewhat poorer agreement, due mostly to insufficient experimental data in the human. The chemical identity of stable lead and RaD makes the information about the former useful in metabolic estimates concerning the latter whenever direct measurements are not available.

Definition of Symbols Used in this Paper

- T_b = the biological half-life in days of the element or nuclide of reference.
 T_e = effective half-life in days of the radionuclide in the organ of reference.
 T_e^x = effective half-life in days of the radionuclide of reference in organ X.
 T_r = radioactive half-life in days of the nuclide of reference.
 m = mass in grams of the organ of reference.
 C = concentration in grams or curies per gram of tissue of the element or nuclide of reference.
 I = daily intake in grams (or curies) of the element or nuclide of reference.
 f_w = fraction of that taken into the body by ingestion that is retained in the organ of reference.
 f_1 = fraction of the ingested material that enters the blood.
 f_2 = fraction of total body content of the element or radionuclide in the organ of reference.
 f_2^I = fraction of the material in the blood that enters the organ of reference.
 f_2^x = fraction of the total body content of the material of interest that is in organ X.
 m^x = mass in grams of organ X.

- C^x = concentration in grams (or curies) per gram of tissue of the element or nuclide in organ X.
- f_w^x = fraction of element or radionuclide taken into the body by ingestion that is retained in organ X.
- f_a = fraction of the material taken into the body by inhalation that arrives at the organ of reference.
- R = fraction of the material retained in the respiratory system from inspired air.

References

1. Report of Committee II, International Commission on Radiological Protection. Health Phys. 3 1-390 (1960).
2. H. Lisco. Argonne National Laboratory Quarterly Report, Biological and Medical Divisions. ANL-4253 (1949) pp. 96-101.
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-57-2-4, ORNL-CF-57-11-33, ORNL-CF-58-10-15 and ORNL-CF-59-8-106.
4. R. A. Kehoe. Occupational Med. 3 156 (1947).
5. J. Cholak and K. Bambach. J. Ind. Hyg. Toxicol. 25, 47 (1943).
6. R. A. Kehoe, J. Cholak, D. M. Hubbard, K. Bambach and R. R. McNary. J. Ind. Hyg. Toxicol. 25, 71-79 (1943).
7. Public Health Service, U.S. Department of Health, Education and Welfare. Air Pollution Measurements of the National Air Sampling Network, 1958.
8. G. W. Monier-Williams. Trace Elements in Food. New York: John Wiley and Sons, Inc., 1949 pp. 64-106.
9. L. S. Goodman and A. Gilman. The Pharmacological Basis of Therapeutics, 2nd ed. New York: The Macmillan Company, 1955. pp. 1002-1016.
10. J. Shapiro. The University of Rochester Atomic Energy Project Report, UR-298.
11. P. E. Morrow, E. Mehrhof, L. J. Cassarett, and D. A. Morken. The University of Rochester Atomic Energy Project Report, UR-504.
12. M. B. Jacobs. The Chemical Analysis of Air Pollutants. New York: Interscience Publishers, Inc., 1960. pp. 76-77.

13. W. F. Bale and J. V. Shapiro. Proceedings of the International Conference on Peaceful Uses of Atomic Energy, Geneva, 1955. New York: United Nations, 1956. Vol. 13, pp. 233-236.
14. K. Aurand, W. Jacobi, H. Muth and A. Schraub. Strahlentherapie 112 262 (1960).
15. J. G. Hamilton. University of California Radiation Laboratory Medical and Health Physics Quarterly Report, UCRL-1282 (1951) p.32.
16. P. W. Durbin, K. G. Scott and J. G. Hamilton. University of California Pub. Pharmacol. 3 1 (1957).
17. M. Sultzzer and J. B. Hursh. The University of Rochester Atomic Energy Project Report. UR-266 (1953).

A COMPARISON OF OBSERVED PLUME RISES WITH VALUES OBTAINED FROM WELL-KNOWN FORMULAS

Harry Moses and Gordon H. Strom*

I. Introduction

The height to which a stack effluent will rise in the atmosphere is of importance in calculating the resulting concentrations at ground level. There are at least eight theoretical or empirical formulas one may use,⁽¹⁻¹¹⁾ but these yield results which may differ by as much as an order of magnitude. Up to the present time reliable measurements under varying stack and meteorological conditions, adequate to test these formulations, have been few. This paper compares the predictions of six such formulas with data obtained with an experimental stack operated by the Meteorology Group at the Argonne National Laboratory.⁽¹²⁾ The formulas tested are those of: 1) Holland, 2) Davidson and Bryant, 3) Sutton, 4) Scorer, 5) Bosanquet, Carey, and Halton, and 6) an additional one by Bosanquet.

II. Equipment and Technique

Smoke plumes are obtained by injecting fog-oil droplets into the airstream supplied to the 111-foot Meteorology Experimental Stack by a 4500-CFM, $7\frac{1}{2}$ -hp blower. Both flow rate and fog-oil concentration can be controlled by louvered dampers. A continuous record of the flow rate is obtained by a Hagan Ring Balance pressure differential recorder, which measures the pressure drop across an orifice plate.

The effluent air-smoke temperature is measured by two pencil-type copper-constantan thermocouples inserted into the stack, one at 31 feet above the base, and the other $\frac{1}{2}$ foot from the top. Once each minute the temperature near the top of the stack is recorded for 40 seconds and that at 31 feet for 20 seconds, alternately, on a Minneapolis-Honeywell self-balancing potentiometer recorder.

Speed and Crown Graphic cameras using 4 x 5-inch film photograph the plume; one located 40 feet from the foot of the stack points vertically; the other 1000 feet from the plume photographs its side. The camera aimed vertically is equipped with a 12.7-cm f4.7 Kodak Ektar lens; the more distant camera has a 38-cm f5.6 Graflex Tele-Optar lens, both using Kodak Tri-X film. Sixteen exposures per film (one every 15 seconds) are made to observe the plume for about 4 minutes.

*Resident Research Associate, Argonne National Laboratory, summers 1955-58 and 1960; Professor of Aeronautics, New York University, New York, New York.

The associated meteorological equipment used for this study has been described elsewhere.⁽¹³⁾ Briefly, wind speed and direction are measured by Aerovanes, while temperature and stability are measured by aspirated copper constantan thermocouples. These instruments (except for the ones nearest the ground) are mounted at four logarithmically spaced levels on the 150-foot Meteorology Tower located 225 feet from the Meteorology Experimental Stack. The Esterline-Angus recorders for wind and Minneapolis-Honeywell Elektronik Potentiometers for temperature are located in the Meteorology Building.

III. Analysis of the Data

From the photographs obtained by the two cameras, it is possible to obtain the position in space of the top and bottom of the plume to a maximum distance of about 300 feet downwind from the stack. The center line of the plume, located midway between the observed top and bottom positions, is compared with the predictions of the plume rise formulas. Samples of the data are presented graphically in Figure 19.

Figure 19

Heights of plume rise

Wind speed values used in this study are obtained by plotting five readings (from 79 inches to 150 feet) on log-log paper and interpolating the 120-foot wind speed. Values of stability are obtained by measurements of the temperature difference between 144 and 5.5 feet above ground.

Meteorological and stack parameters for each run are shown in Table 24.

Table 24
Meteorological and stack parameters

Run No	Date	Beginning Time of Run CST	Meteorological Parameters			Stack Parameters	
			Wind Speed (MPS)	Lapse Rate °C/100M	Air Temp (°K)	Stack Effluent Vel (MPS)	Effluent Temp (°K)
15	8/4/55	1806-1/2	3 94	+ 2 1	304 0	8 03	320 0
18	8/9/55	1750	1 00	+ 2 8	297 9	12 25	314 7
19	8/9/55	1801-1/2	1 00	+ 3 6	298 2	10 53	316 3
20	8/9/55	1826	1 61	+ 4 7	297 6	12 53	311 9
26	8/10/55	1816	1 39	+ 0 5	298 2	6 49	326 9
27	8/10/55	1822	1 25	+ 0 5	298 2	4 83	320 8
28	8/10/55	1835-1/2	1 39	+ 2 4	297 8	7 65	315 2
30	8/12/55	0940-1/2	5 92	- 1 2	297 6	12 65	309 7
33	8/12/55	1001-1/2	4 47	- 1 4	297 5	4 73	330 2
37	8/16/55	1751	0 98	+ 3 6	301 8	9 63	320 0
38	8/16/55	1818	1 12	+ 5 0	301 6	9 65	321 0
39	8/16/55	1824	1 12	+ 5 9	301 4	6 71	326 0
42	8/18/55	1529-1/2	3 40	- 0 9	304 7	12 68	319 7
45	8/18/55	1622-1/2	2 19	+ 0 0	304 0	12 90	321 9
49	8/18/55	1728-1/2	2 21	+ 1 7	303 4	8 15	330 0
51	8/18/55	1806-1/2	2 26	+ 4 3	302 8	8 27	327 0
52	8/19/55	1031	1 98	- 1 2	303 5	13 18	326 0
62	8/22/55	1043-1/2	6 50	- 1 4	299 7	10 33	323 0
99	9/7/55	1053-1/2	5 50	- 0 9	292 0	12 94	313 0
101	9/7/55	1103	7 02	- 1 4	291 5	10 54	314 7
102	9/7/55	1108-1/2	6 17	- 1 7	291 4	7 70	319 7
103	9/7/55	1114	7 30	- 1 4	291 5	7 70	319 7
104	9/7/55	1119-1/2	6 17	- 1 4	291 5	4 87	327 4
106	9/8/55	1525	6 35	- 0 4	292 9	13 00	308 0
107	9/8/55	1530-1/2	5 94	- 0 4	292 8	10 78	310 0
108	9/8/55	1536-1/2	5 50	- 0 4	292 6	7 98	317 0
111	9/12/55	0542-1/2	3 67	+12 3	280 1	13 00	297 4
112	9/12/55	0552	3 95	+11 9	281 0	12 94	295 2
113	9/12/55	0602	4 47	+13 00	281 4	12 97	295 8
115	9/12/55	0655-1/2	3 42	+ 6 4	282 1	13 03	299 1
116	9/12/55	0706	2 83	+ 5 2	282 7	13 07	300 8
117	9/12/55	0712-1/2	2 07	+ 3 6	283 2	13 08	301 3
118	9/12/55	0725	1 29	+ 0 5	282 2	13 16	304 1
119	9/13/55	1259-1/2	5 59	- 0 7	291 4	12 94	309 0
122	9/13/55	1315-1/2	5 36	- 0 5	291 8	12 83	308 0
123	9/13/55	1320	5 36	- 0 5	291 9	12 83	308 0

IV. The Plume Rise Equations

The equations used to calculate the heights of rise according to each of the six techniques are presented in detail below. All of the theoretical formulas have shortcomings since simplifying assumptions were necessary in their derivation. Some of these are pointed out; however, a detailed critique of each of the formulations is beyond the scope of this paper.

The very interesting technique for calculating the height of rise of a bent-over plume given by Priestley⁽⁶⁾ is not included because his equations are specific to a zero temperature gradient. Too few measurements in this study were made under this condition.

A. The Davidson-Bryant Technique

Using data obtained from Bryant's wind tunnel experiments,⁽¹⁴⁾ Davidson derived the following empirical expression⁽⁴⁾

$$\Delta h = d \left(\frac{V_s}{u} \right)^{1.4} \left(1 + \frac{\Delta T}{T} \right) \quad (1)$$

where

Δh = rise of the plume above the stack in meters

d = inside stack diameter in meters

V_s = stack velocity in meters per second

u = wind speed in meters per second

ΔT = temperature excess of effluent gas over ambient temperature ($^{\circ}\text{K}$)

T = ambient air temperature ($^{\circ}\text{K}$) .

This expression, like that of Holland, does not consider stability, and the calculated values are too low.⁽²⁾ Nevertheless, it has been widely used, probably because it is simple to apply.

B. The Holland Technique

Holland⁽²⁾ presented the following empirical expression for calculating the height above the stack, Δh , to which a plume will rise

$$\Delta h = \frac{1.5 V_s d + 4 \times 10^{-5} Q_H}{u} \quad (2)$$

where

V_s = stack velocity in meters per second

d = inside stack diameter in meters

Q_H = heat emission rate in calories per second

and

u = horizontal wind speed in meters per second.

This formula is a modification of one presented by Rupp, Beall, Bornwasser, and Johnson⁽³⁾ based on wind tunnel tests, and it was used to predict the behavior of the Oak Ridge X-10 reactor cooling air exhaust. The formula by Rupp et al., contained a momentum term only. By employing height of rise data from three stacks, two operated by Oak Ridge and a third by the Tennessee Valley Authority, Holland was able to introduce an empirical buoyancy term, $\frac{4 \times 10^{-5} Q_H}{u}$, and thus reduce the scatter in his data. Since the height of plume rise depends on atmospheric stability as well as on the factors in his equation, Holland suggests that stability can be taken into account by adding to the mean height 10 to 20 per cent in lapse conditions and subtracting a like amount in inversions. This arbitrary correction was applied to the calculations used to plot the Holland curves in Figures 20 to 23.

C. The Scorer Technique

This method is discussed in greater detail than the others since it has been extended somewhat by the authors to provide an equation giving the height of rise as a function of distance from the stack. Only the technique for calculating the height at which leveling off occurs is presented by Scorer.

He assigns three phases to the rising plume:

Phase I. In this phase the plume is nearly vertical and the upward velocity exceeds the horizontal wind velocity. Phase I ends and Phase II begins when the vertical velocity approximately equals the horizontal wind velocity.

Figure 20

Histogram of difference between observed and calculated height - 30 meters

Figure 21

Histogram of difference between observed and calculated height - 60 meters

Figure 22

Scatter diagram of observed vs
calculated height - 30 meters

Figure 23

Scatter diagram of observed vs
calculated height - 60 meters

Phase II. This is the transition stage in which the plume bends over and becomes horizontal. Phase II ends when the vertical velocity drops to a value equal to the turbulent velocities of the atmospheric eddies. Scorer assumes that under neutral or unstable conditions this occurs when the vertical velocity is approximately equal to $\frac{1}{10}$ of the horizontal wind velocity. Under stable conditions the eddy velocities may fall to $\frac{1}{100}$ of the wind speed. However, the formulas do not apply under stable conditions.⁽¹⁰⁾

Phase III. In Phase III buoyancy and momentum cease to contribute to the plume rise. The plume is now in a passive stage, and the treatments of Sutton,⁽¹⁵⁾ Roberts,⁽¹⁶⁾ or Frenkiel⁽¹⁷⁾ may be used to describe its subsequent behavior.

Scorer discusses three plume types. These are plumes in which 1) vertical momentum dominates Phases I and II, 2) vertical momentum dominates Phase I but buoyancy is the more effective in Phase II, and 3) buoyancy is active in both Phases I and II. In the experiments used in this report only the first two types were observed.

1. The momentum-dominated plume

Where vertical momentum effects are dominant, the height above the stack to which a plume will rise is given by

$$\Delta h = 2.5 r_0 \left\{ \frac{V_s}{u} \left(\lambda^{-1/2} - \mu^{-1/2} + 2\mu^{-1} \right) - 2 \right\} \quad (3)$$

where

r_0 = stack radius in meters

V_s = effluent exit velocity in meters per second

u = wind speed in meters per second

μ = a proportionality constant. The second phase begins when w , the vertical velocity of the effluent, is equal to μu . μ is assumed equal to 1.

λ = a proportionality constant. The third phase begins when $\lambda = \lambda u$. λ is assumed equal to 0.1.

It is supposed that in a bent-over (horizontal) section of unit length, vertical momentum is conserved, so we may write

$$\begin{aligned} wr^2 &= w_1 r_1^2 = K = \text{constant, and consequently} \\ wy^2 &= w_1 y_1^2 = K_1 = \text{constant} \end{aligned} \quad (4)$$

since $y = 2.25r$ as determined from laboratory measurements. The symbol r represents an effective plume radius, y is the height above a virtual origin associated with the bent-over portion of the plume, and the subscript 1 refers to the level at which the bend-over occurs.

Since $w = \frac{dy}{dt}$ where t is time, we have from Equation (4)

$$y^2 \frac{dy}{dt} = K_1$$

and

$$\int_{y_1}^y y^2 dy = K_1 \int_{t_1}^t dt \quad ;$$

therefore

$$\frac{1}{3} (y^3 - y_1^3) = K_1 (t - t_1) = \frac{K_1}{u} (x - x_1)$$

since $x = ut$ where x is the distance downwind.

Solving for y

$$y = \left[\frac{3 K_1}{u} (x - x_1) + y_1^3 \right]^{1/3} . \quad (5)$$

In Phase I, because momentum is conserved, Scorer shows that

$$wz = V_s z_0 = K_2 = \text{constant} \quad (6)$$

where z is the height above a virtual origin located in the stack and the subscript 0 refers to conditions at the top of the stack. It is to be noted that the origin for the bent-over position of the plume is different from the origin of the vertical jet.

Since $w = \frac{dz}{dt}$, we have from Equation (6)

$$z \frac{dz}{dt} = K_2$$

and

$$\int_{z_0}^z z dz = K_2 \int_0^t dt$$

$$\frac{1}{2} (z^2 - z_0^2) = K_2 t$$

and

$$z^2 = 2 K_2 t + z_0^2 \quad .$$

At the point of the bend-over

$$z_1^2 = 2 K_2 t_1 + z_0^2$$

and since $x = ut$

$$z_1^2 = 2 K_2 \frac{x_1}{u} + z_0^2 \quad .$$

But since $K_2 = w_1 z_1$ and $w_1 = \mu u$

$$z_1^2 = 2 z_1 \frac{\mu u x_1}{u} + z_0^2 = 2 z_1 x_1 + z_0^2 \approx 2 z_1 x_1$$

Therefore,

$$z_1 = 2 x_1$$

Substituting for x_1 , in Equation (5), we have

$$y = \left[\frac{3 K_1}{u} \left(x - \frac{z_1}{2} \right) + y_1^3 \right]^{1/3}$$

Scorer shows that

$$z_1 = \frac{5 r_0 V_s}{u} \quad ,$$

and

$$y_1 = \frac{2.5 r_0 V_s}{u}$$

and since

$$K_1 = w_1 y_1^2 = \mu u y_1^2 \approx u y_1^2$$

we have

$$y = \left[\frac{r_0^2 V_s^2}{u^2} \left(18.75 x - 31.25 \frac{r_0 V_s}{u} \right) \right]^{1/3}$$

Since y refers to the cap of the plume, it is necessary to calculate the height of the center line for comparison with our measurements.

Under the assumption that $y = 2.25r$, the height of the center line (h_{C1}) above the virtual origin for the bent-over plume is therefore

$$h_{C1} = y - r = y - \frac{y}{2.25} = 0.555y$$

$$h_{C1} = 0.555 \left[\frac{r_0^2 V_s^2}{u^2} \left(18.75 x - 31.25 \frac{r_0 V_s}{u} \right) \right]^{1/3} \quad (7)$$

To obtain the height of the center line above the virtual origin in the stack ($j h_{C1}$) we must calculate the height of rise before bend-over, $z_1 - z_0$, and the height during bend-over, $h_{C1} - y_1$. Therefore, the equation for the center line of the plume as a function of distance from the stack is given by

$$j h_{C1} = (h_{C1} - y_1) + (z_1 - z_0)$$

$$= \left[\frac{r_0^2 V_s^2}{u^2} \left(3.22 x - 5.34 \frac{r_0 V_s}{u} \right) \right]^{1/3} + \frac{2.5 r_0 V_s}{u} - 5 r_0 \quad (8)$$

This equation was used to calculate the height of the non-buoyant plume center line at 30 and 60 meters.

2. The plume with buoyancy active in Phase II

When buoyant effects dominate the bend-over phase the height of rise (h) is given by

$$h = \frac{0.58F}{\lambda^2 u^3} + 2.5 r_0 \left[\frac{V_s}{u} \left(-\mu^{-1/2} + 2\mu^{-1} \right) - 2 \right] \quad (9)$$

where F is the so-called flux of buoyancy (to use Scorer's term) given by

$$g \pi r_0^2 V_s \left(\frac{\Delta T}{T} \right)_0$$

To determine whether momentum or buoyancy effects dominate, Equations (3) and (9) are compared - actually, only the first terms need be compared since the other terms are identical. If

$$\frac{0.58F}{\lambda^2 u^3} > \frac{2.5 r_0 V_s}{\lambda^{1/2} u} \quad \text{or more simply} \quad \left(\frac{\Delta T}{T} \right)_0 > 1.4 \frac{\lambda^{3/2} u^2}{r_0} \quad (10)$$

buoyancy predominates and Equation (9) is used, otherwise Equation (3) is appropriate for calculating the total rise of the top of the plume.

The plume rise as a function of distance from the stack when buoyancy is dominant during the bend-over stage was derived using the following assumptions given by Scorer⁽⁹⁾

1. The total buoyancy in a section of unit length bounded by vertical planes remains constant, i.e.
 $y_1^2 B_1 = y^2 B$ where $B = \frac{\Delta T}{T}$. T is the temperature of the air and ΔT the difference between the temperature of the effluent and air.
2. $r = y/2.25$.
3. $w = 0.8(g B r)^{1/2}$.

The constants 0.8 and 2.25 in the last two equations were determined empirically from laboratory models.

With these assumptions it can be shown that in the buoyant plume

$$w y^{1/2} = K = \text{constant} \quad .$$

Since $w = \frac{dy}{dt}$, the differential equation is easily solved, and, by an analysis similar to that of the non-buoyant plume, we obtain the following equation relating the height of rise of the plume's center line as a function of distance

$$B h_{c1} = \left\{ 2.58 r_0 \left[\left(\frac{\Delta T}{T} \right)_0 \frac{V_s}{u^3} \right]^{1/2} \left(x - 2.5 \frac{V_s r_0}{u} \right) + 1.62 \left(\frac{V_s r_0}{u} \right)^{3/2} \right\}^{2/3} + 2.5 \frac{V_s r_0}{u} - 5 r_0 \quad . \quad (11)$$

This equation was used to calculate the rise of the buoyant plume center line at points 30 and 60 meters downwind from the stack.

In using Scorer's formulae for calculating the height of the plume's center line, first the buoyancy criterion of Equation (10) was applied. If

$$\left(\frac{\Delta T}{T} \right)_0 > \frac{1.4 \lambda^{3/2} u^2}{r_0}$$

then the height given by Equation (11) was compared with the height given by

$$\Delta h = \frac{1.25}{2.25} \left(\frac{0.58F}{\lambda^2 u^3} \right) + 2.5 r_0 \left[\frac{V_s}{u} \left(-\mu^{-1/2} + 2\mu^{-1} \right) - 2 \right] \quad (9a)$$

The smaller value was chosen as the plume height at 30 meters and similarly at 60 meters. If

$$\left(\frac{\Delta T}{T} \right)_0 < \frac{1.4 \lambda^{3/2} u^2}{r_0} \quad ,$$

the calculated height given by Equation (8) for the non-buoyant plume was compared with that of the following equation

$$\Delta h = 2.5 r_0 \left[\frac{V_s}{u} \left(\frac{1.25}{2.25} \lambda^{-1/2} - \mu^{-1/2} + 2\mu^{-1} \right) - 2 \right] \quad (3a)$$

and again the smaller value was chosen. Equations (3a) and (9a) are modifications of Equations (3) and (9) respectively, and give the heights of the center line instead of the plume top.

D. The Sutton Technique

Sutton⁽¹⁾ presented the following theoretical expressions for calculating the height of plume rise:

$$w_1^3 = w_s^3 \quad (12)$$

$$z = \frac{3}{2} \frac{w_1^3}{u^3} [\cot \psi \csc \psi - \ln(\cot \psi + \csc \psi)] \quad (13)$$

where

$$w_1^3 = \frac{7}{3} \left(\frac{g \Delta T r_0^2 V_s}{TC} \right) \quad (14)$$

from which it follows that

$$x = \frac{w_1^3}{u^3} \left(\frac{\cot^2 \psi - 2}{\sin \psi} + 2 \right) \quad (15)$$

The origin of the cartesian coordinate system is taken at the top of the stack with:

- x = distance downwind in meters
- s = distance along the plume axis in meters
- w = vertical velocity in meters per second
- z = vertical distance in meters
- ψ = angle made by the tangent to the plume axis with the x axis
- g = acceleration of gravity assumed to be 9.8 meters/sec^2
- ΔT = temperature difference between the stack gas and the atmosphere ($^{\circ}\text{K}$)
- r_0 = internal radius of the stack in meters
- V_s = stack exit velocity in meters per second
- T = ambient air temperature ($^{\circ}\text{K}$)
- C = Sutton's diffusion parameter in $(\text{meters})^{n/2}$ where n is another diffusion parameter
- u = horizontal wind in meters per second.

For the computations, C and n were assigned the values shown in Table 25 based on the recommendations of "Meteorology and Atomic Energy"⁽¹⁸⁾ for a level of 110 feet.

Table 25
Selected Sutton parameters

	Stability	$C(\text{meters})^{n/2}$	n
Large lapse	$\frac{\partial T}{\partial z} \leq -1^{\circ}\text{C}/100 \text{ meters}$	.20	.20
Isothermal or small lapse	$0 \geq \frac{\partial T}{\partial z} > -1^{\circ}\text{C}/100 \text{ meters}$	.11	.25
Moderate inversion	$0 < \frac{\partial T}{\partial z} < 2^{\circ}\text{C}/100 \text{ meters}$	.07	.33
Strong inversion	$\frac{\partial T}{\partial z} \geq 2^{\circ}\text{C}/100 \text{ meters}$	.06	.50

Sutton derives the expression for the radius of the plume, r , as:

$$r = 1.5C z^{(2-n)/2} \quad (16)$$

A virtual origin was assumed at a distance, z_0 , below stack top for which r equalled r_0 , the stack radius. The distance of virtual origin below the stack top (z_0) was calculated from the equation

$$z_0 = \left(\frac{r_0}{1.5C} \right)^{2/(2-n)} \quad (17)$$

If the theoretical plume were plotted from the virtual origin, the plume axis would cross the level corresponding to the stack top, the zero level, a distance downwind from the stack. This, of course, was unreal. The entire theoretical plume was translated upwind so that it crossed the top of the stack. This correction still presents inherent difficulties since its inclination at the stack top differs from the virtual inclination of the actual plume. Nevertheless, this correction provides a better fit for the data.

E. The Bosanquet-Carey-Halton Formula

In this technique the plume rise due to momentum and that due to buoyancy are computed separately and then added to obtain the total rise. The maximum rise due to momentum effects, $z_{\max}$, is given by

$$z_{\max} = \frac{4.77}{1 + 0.43 \frac{u}{V_s}} \cdot \frac{\sqrt{Q V_s}}{u} \quad (18)$$

and the height at any distance by

$$h_V = z_{\max} \left(1 - 0.8 \frac{z_{\max}}{x} \right) \quad (19)$$

where

$$x > \text{about } 2 z_{\max}$$

and where Q is the volumetric emission rate, x is the distance downwind, V_s the stack velocity, and u the wind velocity. Any consistent system of units may be used.

The rise due to buoyancy h_B at a distance x may be obtained from the equations

$$x = 3.57 \frac{\sqrt{Q V_s}}{u} X \quad (20)$$

and

$$h_B = \frac{6.37g Q(T_g - T_1)}{u^3 T_1} Z \quad (21)$$

if

$$u^2 > \frac{(T_g - T_1)g}{T} \sqrt{\frac{Q}{V_s}}$$

where T_g is the temperature of the stack gases, T_1 the temperature at which the density of the flux gases equals that of the atmosphere, and g is the acceleration of gravity. A graph showing the functional relation between the non-dimensional parameters X and Z is presented in Reference 5. The total rise above the chimney top (h_T) is given as

$$h_T = h_V + h_B \quad (22)$$

Bosanquet, Carey, and Halton point out that Equations (19) through (22) are derived for an adiabatic atmosphere only but suggest that the equations may be used close to the stack "without serious error" in a non-adiabatic atmosphere.

F. The Bosanquet (1957) Formula

Bosanquet⁽⁷⁾ published another technique for calculating the heights of rise of a smoke plume which follows the same general lines as that of his earlier paper⁽⁵⁾ with Carey and Halton. Bosanquet claims to have simplified the computation.

In comparing the Argonne experimental data with the 1957 Bosanquet technique, the following formula was used

$$\Delta h = AU \left\{ f_I(X) + f_{II}(X_0) - \frac{0.615 X_0^{1/2}}{\left[\left(\frac{V_s}{U} \right)^2 + 0.57 \right]^{1/2}} \right\} \quad (23)$$

where

$$A = \frac{gQ\theta}{2\pi\Delta^2T_1U^4}$$

$$X = \frac{t + t_0}{A} = \frac{L}{UA} + \frac{t_0}{A}$$

$$t_0 = \frac{4}{3} \frac{V_s T_1}{g\theta}$$

$$X_0 = \frac{t_0}{A}$$

Tables for $f_I(X)$ and $f_{II}(X_0)$ are given in Bosanquet's paper. The other symbols have the following meaning: g is the acceleration of gravity, Q is the volumetric flow rate, θ is the temperature excess of the effluent over the temperature at which it would have the same density as that of the atmosphere, T_1 is the absolute temperature of the effluent at which it has the same density as that of the atmosphere. (In this study it is sufficiently accurate to assume $T_1 =$ air temperature $+8^\circ$ since the variations in effluent density were small.) U is wind speed; Δ is a dilution coefficient; t is time in seconds; and L is distance from the stack. Any self-consistent system of units may be used. Bosanquet assumes a value of 0.13 for Δ . The inflow per unit area of the plume is the product of Δ and the wind speed. For a plume oriented longitudinally to the wind having a half-angle of 12° he shows that the inflow per unit area is 0.106 U . For a plume oriented transversely to the wind he shows that the inflow is 0.159 U . A value for Δ of 0.13 representing a compromise between 0.159 and 0.106 is therefore suggested by Bosanquet. In this study calculations were carried out for values of Δ equal to 0.09, 0.13, and 0.17.

V. Discussion of Results

Since the plumes were photographed over a maximum length of only about 100 meters, one could not be certain of the final maximum height obtained. However, since the buoyancy of these plumes was relatively small (see Table 24), it appears that most of the rise had already occurred by the time the plumes reached the second check point 60 meters from the stack. In support of this assertion is the fact that the average slope of the center line between 30 and 60 meters was less than 5° in 27 of the 31 plumes measured to 60 meters. Four of these even showed a slight negative slope, probably because of atmospheric downdrafts.

In this study the observed heights at both 30 and 60 meters are compared with either the maximum heights as given by the Davidson-Bryant and Holland equation, or the height at 30 and 60 meters as given

by the other formulations. Tables 26 and 27 summarize the measured and computed heights. Blanks occur in the Scorer columns because calculations for stable conditions were omitted. It is of interest to note that all Scorer calculations for 60 meters and all but one for 30 meters represent the plume as being in the passive stage, Phase III, further support for the observations of the previous paragraph.

Table 26
Summary of measured and computed heights in meters
30 meters

Run No.	Observed Height	Davidson-Bryant	Holland	Scorer	Sutton	Bosanquet-Carey-Halton	Bosanquet - 1957		
							$\Delta = .09$	$\Delta = .13$	$\Delta = .17$
15	3.7	1.3	1.3	-	5.3	3.0	4.6	3.1	2.4
18	9.3	15.7	6.8	-	25.9	11.0	34.9	22.0	19.3
19	12.2	12.7	5.9	-	25.3	11.9	30.1	21.7	16.7
20	4.7	8.2	4.4	-	15.9	9.4	22.2	15.2	11.4
26	9.9	4.2	3.1	-	18.1	8.9	14.4	10.1	6.8
27	3.2	3.2	2.5	-	16.9	7.6	12.0	8.2	6.2
28	3.3	5.1	3.1	-	16.7	8.4	15.9	11.0	8.3
30	6.9	1.3	2.0	2.2	3.0	3.1	4.6	3.2	2.4
33	1.3	0.5	1.2	0.5	4.0	1.6	2.1	1.4	1.1
37	8.3	11.5	5.5	-	25.1	12.0	28.2	20.4	14.7
38	10.7	9.6	4.9	-	22.7	11.6	25.1	16.4	13.7
39	3.6	5.9	3.4	-	21.8	10.9	18.2	13.1	10.0
42	6.5	2.9	2.8	4.6	6.7	5.4	9.3	6.4	4.8
45	6.1	5.6	3.9	-	12.4	8.0	16.4	11.1	8.4
49	4.7	3.0	2.6	-	12.0	6.1	10.4	7.1	5.3
51	4.8	2.9	2.2	-	11.6	5.9	10.1	6.9	5.2
52	4.1	6.8	6.0	8.4	11.5	9.1	19.1	13.2	9.9
62	3.8	0.9	1.8	1.3	3.1	2.3	3.3	2.3	1.7
99	2.6	1.6	2.1	2.5	4.6	3.5	5.4	3.7	2.8
101	4.2	0.8	1.7	1.2	2.9	2.1	3.0	2.1	1.6
102	6.7	0.7	1.5	0.8	3.2	1.8	2.5	1.7	1.3
103	1.6	0.5	1.3	0.5	2.7	1.4	1.9	1.3	1.0
104	3.6	0.4	1.0	0.1	3.0	1.0	1.4	0.9	0.7
106	1.2	1.3	1.7	2.0	3.4	3.0	4.4	3.1	2.3
107	2.0	1.1	1.6	1.7	3.6	2.6	3.9	2.6	2.0
108	2.0	0.8	1.3	1.1	4.0	2.1	3.0	2.0	1.5
111	2.2	2.8	2.2	-	7.4	5.2	8.9	6.1	4.6
112	3.6	2.5	2.0	-	6.3	4.8	7.9	5.4	4.1
113	3.9	2.1	1.8	-	5.5	4.3	6.9	4.7	3.6
115	3.2	3.1	2.4	-	7.9	5.6	9.7	6.6	5.0
116	4.2	4.0	2.8	-	10.0	6.6	12.3	8.4	6.3
117	5.9	6.2	3.7	-	13.8	8.5	17.9	12.2	9.2
118	10.4	12.4	6.6	-	22.6	11.6	29.4	18.4	16.1
119	4.6	1.5	2.0	2.4	4.2	3.4	5.2	3.6	2.7
122	6.1	1.6	2.0	2.6	4.2	3.5	5.4	3.7	2.8
123	5.4	1.6	2.0	2.6	4.2	3.5	5.4	3.7	2.8

Table 27
Summary of measured and computed heights in meters
60 meters

Run No.	Observed Height	Davidson-Bryant	Holland	Scorer	Sutton	Bosanquet-Carey-Halton	Bosanquet - 1957		
							$\Delta = .09$	$\Delta = .13$	$\Delta = .17$
15	3.9	1.3	1.3	-	9.3	3.2	4.6	3.1	2.4
18	11.7	15.7	6.8	-	43.9	20.2	37.6	23.3	20.2
20	6.3	8.2	4.4	-	27.0	12.5	22.5	15.3	11.5
26	13.1	4.2	3.1	-	30.5	11.6	16.6	11.2	7.5
27	5.2	3.2	2.5	-	28.5	9.6	13.5	9.0	6.7
33	1.4	0.5	1.2	0.5	6.7	1.6	2.2	1.5	1.1
37	10.2	11.5	5.5	-	42.5	18.7	31.4	21.9	15.7
38	16.9	9.6	4.9	-	38.4	16.6	27.5	17.7	14.4
39	5.3	5.9	3.4	-	37.0	14.7	21.4	14.7	11.0
42	5.7	2.9	2.8	4.6	11.2	6.1	9.3	6.4	4.8
49	6.6	3.0	2.6	-	20.3	7.2	10.8	7.3	5.4
51	7.1	2.9	2.2	-	19.9	6.9	10.5	7.1	5.3
52	2.6	6.8	6.0	9.1	18.9	11.6	19.7	13.5	10.1
62	6.4	0.9	1.8	1.3	5.3	2.4	3.3	2.3	1.7
99	3.8	1.6	2.1	2.5	7.7	3.7	5.4	3.7	2.8
101	7.9	0.8	1.7	1.2	4.9	2.2	3.1	2.1	1.6
102	9.0	0.7	1.5	0.8	5.4	1.8	2.5	1.7	1.3
103	2.2	0.5	1.3	0.5	4.5	1.5	1.9	1.3	1.0
104	6.8	0.4	1.0	0.1	5.0	1.1	1.4	0.9	0.7
106	1.7	1.3	1.7	2.0	5.8	3.1	4.4	3.1	2.3
107	2.3	1.1	1.6	1.7	6.2	2.8	3.9	2.7	2.0
108	3.1	0.8	1.3	1.1	6.8	2.2	3.0	2.0	1.5
111	2.1	2.8	2.2	-	12.8	5.8	8.9	6.1	4.6
112	4.9	2.5	2.0	-	10.9	5.3	7.9	5.4	4.1
113	5.8	2.1	1.8	-	9.5	4.6	6.9	4.7	3.6
115	4.4	3.1	2.4	-	13.7	6.3	9.7	6.6	5.0
116	4.0	4.0	2.8	-	17.1	7.7	12.4	8.4	6.4
117	6.6	6.2	3.7	-	23.5	10.6	18.2	12.3	9.3
119	6.7	1.5	2.0	2.4	7.1	3.6	5.2	3.6	2.7
122	7.7	1.6	2.0	2.6	7.2	3.8	5.4	3.7	2.8
123	7.9	1.6	2.0	2.6	7.2	3.8	5.4	3.7	2.8

A. Comparison of Observed Values with Range of Calculated Values

In Figures 24 and 25 the observed heights are arranged in increasing order and compared with the highest and lowest calculated values. At 30 meters 75 per cent, and at 60 meters 74 per cent of the measured heights fall within the range of calculated values. The Sutton values represent the maxima in all cases except one, while the minimum values are divided between the Davidson-Bryant, Holland, Scorer and Bosanquet computations. In all probability atmospheric eddies are responsible for the measured heights falling outside of the computed range.

Figure 24

Comparison of observed height with maximum
and minimum calculated heights

Figure 25

Comparison of observed height with maximum
and minimum calculated heights

B. Comparison by Histograms

Histograms showing the frequency distribution of the differences between calculated and observed values are shown in Figures 20 and 21. The mean difference is shown by the dashed line and the standard deviation of the difference about the mean is indicated by arrows. From the position of the dashed mean difference line with respect to the zero line, one can tell at a glance whether the calculated values generally tend to be too low or too high. At 30 meters, the Davidson-Bryant and the Holland values were somewhat low and the others (not counting Scorer) were too high. At 60 meters the Bosanquet 1957 formula with a diffusion coefficient of 0.17, as well as those of Davidson-Bryant and Holland, reads somewhat low. Holland⁽²⁾ also found the Davidson-Bryant values too low when applied to his data.

Information on how frequently large errors occur or how often the formula predicts the height within some specified range may also be determined from these diagrams. The data of Table 28, showing for each technique the per cent of calculated values falling within 1.5 and within 4.5 meters of the observed values, are based on the histograms. It is of interest to note that the Davidson-Bryant formula predicted the height to within 4.5 meters in well over 80 per cent of the runs. This was true of three of the other techniques for the 30 meters distance, but of only the Davidson-Bryant technique at 60 meters. The Sutton values were the least satisfactory.

Table 28

Per cent of calculated values within indicated
height of measured values

Author	At 30 Meters		At 60 Meters	
	± 1.5 Meters	± 4.5 Meters	± 1.5 Meters	± 4.5 Meters
Davidson-Bryant	42	89	29	84
Holland	33	86	19	61
Scorer	31	87	33	60
Sutton	11	50	13	42
Bosanquet-Carey-Halton	47	86	45	74
Bosanquet (1957)				
$\Delta = .09$	25	50	19	55
$\Delta = .13$	31	64	42	71
$\Delta = .17$	31	75	32	61

C. Comparison by Scatter Diagrams

The observed and calculated plume rises are further compared in the scatter diagrams drawn for each of the formulas based on the measurements taken at 30 and 60 meters (Figures 22 and 23). The four categories of stability used in this paper are denoted by separate symbols as indicated in the legend of these figures. Also shown is a line through the origin of each graph inclined 45° to the horizontal. The scatter of points about this line is a measure of the validity of each formula since, if a particular expression were perfect, all of the points would lie along the line. The root mean squares of the deviations about this line are shown in Tables 29 and 30 which summarize the results of this study. The reader should remember that when points lie above this line the calculated values are too low; when the points lie below the line the formula values are too high.

Also appearing on each scatter diagram is the least squares regression line with the formula value as the independent variable. Because the scatter of points about this line is smaller than that about any other line, it may be used as a practical tool to improve the accuracy of the calculated plume rises. The degree of improvement may be observed by comparing the standard error of estimate values (root mean square of the deviations about the regression line) with the values of the root mean square of the deviations about the 45° line. (See Table 29.)

An inspection of the data in the scatter diagrams discloses that the correlation between calculated and observed values appears better during stable conditions than during neutral or unstable conditions. This is rather severe on the Scorer technique since his values, in accordance with his suggestion,⁽¹⁰⁾ were calculated only for neutral and unstable conditions. Not only his, but all of the other techniques, correlated poorly under these conditions. The graphs show that there is a considerable scatter of points and that the scatter at 60 meters exceeds that at 30 meters. The scatter of all of the points is due in part, no doubt, to the brevity of the 4-minute observing period. Evidence for this is the variation in observed plume height in consecutive runs only a few minutes apart. Eddies with time scales greater than 4 minutes would influence the plume height more than some of the variables that appear in the formulas. The increased scatter at 60 meters is probably the result of the longer time in which the eddies had a chance to act.

VI. Conclusions

From the information contained in Tables 28 to 30 summarizing the results of this study, a suitable formula for calculating the height of plume rise may be selected. There is no one formula which is outstanding in all respects.

Table 29

Statistical summary of calculated and observed plume heights
30 meters

Parameter Units are in Meters	Observed Height	Davidson- Bryant	Holland	Scorer	Sutton	Bosanquet- Carey- Halton	Bosanquet - 1957		
							$\Delta = .09$	$\Delta = .13$	$\Delta = .17$
Mean height	5.0	4.1	2.9	2.1	10.3	5.9	11.5	7.9	6.1
Standard deviation	2.8	4.0	1.6	2.0	7.7	3.5	9.2	6.2	5.0
Calculated minus observed values									
Average Difference		-1.0	-2.2	-1.8	5.3	0.8	6.5	2.8	1.1
Maximum Difference		6.3	6.8	5.9	18.2	7.3	25.6	12.6	10.0
Correlation coefficient (calculated vs observed values)		0.71	0.69	0.26	0.66	0.64	0.69	0.69	0.69
Standard deviation of differences between calcu- lated and observed values		2.7	2.0	2.4	6.1	2.7	7.5	4.7	3.6
Root mean square of differences (root mean square devia- tion of observed heights about 45° line in Figures 6a to 6h)		2.9	2.9	2.9	8.0	2.7	9.8	5.4	3.7
Standard error of estimate (root mean square devia- tion of observed heights about regression line)		1.9	2.0	1.9	2.1	2.1	2.0	2.0	2.0
Number of runs used for Scorer = 16; Observed mean heights for Scorer = 3.9; Standard deviation of observed values = 2.0; Number of runs used for all other calculations = 36.									

The nature of the problem would influence the selection of the formula. For example, if there were interest in keeping to a minimum the average difference between the calculated and observed heights, one would select the formulas of Bosanquet-Carey-Halton or the Bosanquet (1957) with $\Delta = 0.17$. If a minimum root mean square difference between calculated and observed values were the desired condition, then one would choose the formulae of Bosanquet-Carey-Halton, Davidson-Bryant, or Holland. As a matter of practical consideration, the computation time is least for the Bryant-Davidson or Holland formulas.

Scorer's technique may be as valid as any of the others, but it must be compared with the same set of data to determine its effectiveness. The formulas of Sutton or Bosanquet, (1957), $\Delta = 0.09$, are not recommended. For many purposes the Davidson-Bryant, Holland, Bosanquet-Carey-Halton, or the remaining Bosanquet (1957) formulas would appear to be equally satisfactory.

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References

1. O. G. Sutton. The Dispersion of Hot Gases in the Atmosphere. J. Meteorol. 7 (5) 307-310 (1950).
2. J. Z. Holland. A Meteorological Survey of the Oak Ridge Area. U. S. Atomic Energy Commission Report, ORO-99 (1953).
3. A. F. Rupp, S. E. Beall, L. P. Bornwasser, and D. F. Johnson. Dilution of Stack Gases in Cross Winds. U. S. Atomic Energy Commission Report, AECD-1811 (1948).
4. W. F. Davidson. The Dispersion and Spreading of Gases and Dust from Chimneys. Trans. Conf. on Industrial Wastes, 14th Ann. Meeting, Ind. Hygiene Found. Am., Nov. 18, 1949. pp 38-55.
5. C. H. Bosanquet, W. F. Carey and E. M. Halton. Dust Deposition from Chimney Stacks. Proc. Inst. Mech. Engs. 162 355-367 (1950).
6. C. H. B. Priestley. A Working Theory of the Bent-Over Plume of Hot Gas. Quart. J. Roy. Meteorol. Soc. 81 139-143 (1956).

7. C. H. Bosanquet. The Rise of a Hot Waste Gas Plume. J. Inst. of Fuel. 30 (197) 322-328 (1957).
8. R. S. Scorer. Natural Aerodynamics. London: The Pergamon Press, 1958.
9. R. S. Scorer. The Behavior of Chimney Plumes. Int. J. Air Poll. 1 (3) 198-220 (1959).
10. R. S. Scorer. The Rise of Bent-Over Hot Plumes. Advances in Geophys. 6 399-411 (1959).
11. F. Wippermann and W. Klug. Schornsteinmindestenhoeen, Ihre Bestimmung aus Gesetzmassigkeiten der turbulenten Diffusion in der Atmosphere. Verein Deutscher Ingenieure, Technische Hochschule, Darmstadt, West Germany, July 1960.
12. R. D. M. Clark. Photographic Techniques for Measuring Diffusion Parameters. Fourth AEC Air Cleaning Conf, held at Argonne National Laboratory. U. S. Atomic Energy Commission Report, TID-7513, Pt. I (1955). pp. 186-200.
13. H. Moses and J. H. Willett. Five-Year Climatological Summary, July 1949-June 1954, Argonne National Laboratory, DuPage County, Illinois, ANL-5592 (1957).
14. L. W. Bryant. The Effects of Velocity and Temperature of Discharge on Shape of Smoke Plumes from a Funnel or Chimney in a Wind Tunnel. National Physical Laboratory (Great Britain), 1949.
15. O. G. Sutton. Micrometeorology. New York: McGraw-Hill Book Co., Inc., 1953.
16. O. F. T. Roberts. The Theoretical Scattering of Smoke in a Turbulent Atmosphere. Proc. Roy. Soc. A 104 640-654 (1923).
17. F. N. Frenkiel. Turbulent Diffusion. Advances in Applied Mechanics 3 (1953).
18. Meteorology and Atomic Energy, U. S. Weather Bureau. U. S. Atomic Energy Commission Report, AECU-3066 (July 1955).

THE ARGONNE METEOROLOGICAL MODEL TOWING TANK*

Paul Frenzen

Extension of the experimental studies of Lagrangian autocorrelations in grid-produced turbulence^(1,2) to more strongly stratified fluids revealed that under these conditions the basic assumptions of the "Batchelor corrections"⁽²⁾ relative to the character of the turbulent decay process no longer were appropriate. Instead of an approximately homogeneous field in which the fluctuation energy ($\overline{u^2}$) decreased linearly with time, flow patterns featuring two-dimensional (x,z) jets were observed. Moreover, indications of brief periods of accelerated decay that abruptly terminated in very slow rates disclosed certain limitations upon the use of the Batchelor decay corrections which must be observed in stable regimes. As a result of a detailed program of experiment and analysis, it has been found possible to predict the onset of these phenomena in terms of critical values of a non-dimensional shear-stability parameter, the internal Froude number.

The experimental program consisted of eleven separate runs, one in which the density stratification was in neutral equilibrium (isothermal) and ten with stable stratifications ranging from $0.09^\circ\text{C cm}^{-1}$ to $0.50^\circ\text{C cm}^{-1}$. Each run resulted in about fifteen photographs of one-second-long tracer displacements taken from the side of the test section; the first ten were at two-second intervals while the last five were spaced four seconds apart. Thus, the evolution of the field of flow was followed for about forty seconds subsequent to the passage of the grid. All experiments in this series employed the $\frac{3}{8}$ -inch bar, 2-inch mesh rectangular grid driven at 25 cm per second (grid Reynolds number 1.4×10^4), the same combination as that used in the earlier Lagrangian trials.

Quantitative velocity data were obtained by measuring horizontal (x) displacements of tracer droplets with digitized coordinate read-out equipment, choosing for measurement only those traces that lay nearest the points of a ten by ten grid superimposed upon each photograph. By averaging the as many as ten measurements obtained at each of the ten levels of the analysis grid, successive vertical profiles of the developing horizontal flow were obtained. These showed the steady growth of jet-like velocity discontinuities during the early portion of each of the stably-stratified experiments, a fairly steady jet maximum eventually being established very near the middle

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level. The jets appeared earlier and grew to greater strengths as the fluid stability was increased from one experiment to the next; in the neutral case the jet was not observed at all.

Plots of the root-mean-square velocity fluctuation ($\underline{u}$) against time in log-log coordinates determine the rates of energy decay. In the neutrally stratified case, the more or less familiar first period of decay ($\underline{u} \propto t^{-0.5}$) was observed to prevail throughout the nearly forty-second record of the experiment. This is in direct support of the earlier assumption that the Batchelor decay corrections, which presuppose this rate of decay, may be applied to data obtained in neutrally stratified fluids. However, the unexpectedly different results of stably stratified experiments suggest that the extension of these relations to data from stable regimes is permissible only when certain restrictions upon the duration of the periods of observation are obeyed.

With a relatively weak stable vertical temperature gradient ($0.09^\circ\text{C cm}^{-1}$) the first period decay rate was seen to prevail for only twelve or thirteen seconds after grid passage. During the next fourteen seconds the rate increased to $\underline{u} \propto t^{-0.77}$. Similar tendencies for the appearance of periods of accelerated decay with rates near $t^{-0.7}$ were observed in six of the ten stable cases, three of these being especially well marked. It is of interest to note that just such an "intermediate period" of more rapid decay has been predicted,⁽³⁾ the theory calling for $\underline{u} \propto t^{-5/7}$. That this rate has not often been experimentally observed is perhaps due to the fact that initial grid Reynolds numbers in wind tunnels are usually not sufficiently large.

The current observations (which, it should be remembered, include only that component of turbulence directed parallel to the mean flow) suggest that the presence of stable density stratifications favors the occurrence of the intermediate period. In view of the assumptions involved in the theoretical treatment, (namely that the similarity of the spectral shape is preserved during decay mainly by viscous dissipation in the high frequencies and principally by transfer between eddies of different sizes in the low frequencies, cf. Reference 3) this may be the result of an interference with the transfer of energy across the middle range of the turbulence spectrum. Further speculation in this regard leads to a picture of turbulence in stratified media in which eddies larger than some median size, decoupled to some degree from the higher frequency range by the presence of the stable stratification, tend to establish (through the agency of the transfer process) a state of equilibrium with the very largest eddies. Since in stable stratifications the larger eddies may include gravity waves, the possibility exists that some components of a turbulent field under these conditions might experience a net gain in energy at the waves' expense.

During the latter stages of each of these experiments in stable fluids, the fields of turbulence were observed to degenerate abruptly into a regime characterized by greatly reduced rates of decay ($\underline{u} \propto t^{-0.25}$). The time of this collapse varied inversely with the stability of the fluid, that is, it occurred successively earlier in the more stable cases. This stage should not be confused with the "final" or "viscous" period of turbulent decay, one for which both theory and experiment have established a decay rate of $\underline{u} \propto t^{-5/8}$. In fact, since these rates of decay are so small, it would appear that this terminal regime is no longer turbulent at all in the usual sense. On the other hand, neither has it yet become truly laminar.

A number of workers,⁽⁴⁻⁸⁾ the list including both theoreticians and experimentalists, have found intimate connections between jet phenomena in stable fluids and a critical ratio of inertial and gravitational forces, namely

$$Fi^2 = \frac{\bar{\rho} \underline{u}^2}{\ell} / \bar{\rho} g \left(\frac{\Delta \rho}{\bar{\rho}} \right) = \frac{u^2 \bar{\rho}}{g \ell \Delta \rho}$$

where Fi is termed "the internal Froude number." With these precedents, a bulk form of this parameter was calculated for each photograph in the current series by averaging the point-to-point vertical velocity differences over the entire analysis grid; the result was employed as the vertical shear factor in the Fi expression above.

It was found that this form of Fi decayed with time as one might expect in turbulence in the lee of a grid, the decay proceeding in accordance with power-law relationships of the form

$$Fi = at^{-b} ,$$

Both the intercepts, a , and the negative slopes, b , were found to decrease in a fairly regular and predictable manner with increasing stability (Figure 26). Further, by noting that $Fi \propto u^2/\ell$ and assuming the first period decay relationships for representative velocity and length scales (i.e., $\underline{u} \propto t^{-0.5}$ and $\ell \propto t^{+0.5}$), it becomes possible to derive the limiting decay rate for negligibly small stable lapse rates: $b \rightarrow 0.75$ as $d\theta/dz \rightarrow 0$. (In the isothermal experiment of this series, the ratio u^2/ℓ was found to decay as $t^{-0.77}$.)

With the aid of the foregoing results, the Fi decay data from all of the stable experiments were interpolated to construct the single nomogram of Figure 27. The practical significance of this diagram to the towing-tank diffusion studies becomes apparent when one assembles the values of the internal Froude number which obtained when the rates of turbulent decay increased to $t^{-0.7}$ and again when the fields collapsed into the terminal regime of very slow decay. The intermediate period of accelerated decay was found to begin when Fi approached unity, six scattered values ranging from $Fi = 1.3$ to 0.7 .

Figure 26

Observed relations between slopes and intercepts of F_i decay curves and the vertical temperature gradient. (Increased scatter in data of set II is at least partly due to malfunction of digitized coordinate read-out equipment.)

Figure 27

Empirically derived relations between the decay of the internal Froude number and the vertical temperature gradient; grid Reynolds number = 1.4×10^4 . (Nomogram constructed by graphical interpolation of Figure 26.)

More definitely, the turbulent field was observed to degenerate to the terminal state when the internal Froude number had fallen to a narrow range centered around $Fi = 0.6 (\pm 0.04)$. Hence, these limits shown upon the nomogram of Figure 27 predict the limiting times beyond which the first period decay relations (and therefore, the Batchelor decay corrections) can no longer be applied over a range of fluid stability conditions.

Of considerable interest in themselves and certainly worthy of further study, these results have immediate application in the design of Lagrangian autocorrelation experiments in the stable range.

References

1. P. Frenzen. Radiological Physics Division Semiannual Report, July through December 1956, ANL-5679, pp. 95-100; Radiological Physics Division Semiannual Report, January through June 1957, ANL-5755, pp. 77-82; Radiological Physics Division Semiannual Report, January through June 1958, ANL-5919, pp. 113-124; Radiological Physics Division Semiannual Report, July through December 1958, ANL-5967, pp. 183-190; Radiological Physics Division Semiannual Report, July through December 1959, ANL-6104, pp. 100-108.
2. P. Frenzen. Radiological Physics Division Semiannual Report, July through December 1957, ANL-5829, pp. 218-227.
3. T. Von Karman and C. C. Lin. On the Statistical Theory of Isotropic Turbulence. *Advances in Appl. Mech.* II 2-19 (1951).
4. C. G. Rossby. On the Vertical and Horizontal Concentration of Momentum in Air and Ocean Currents. *Tellus* 3 15-27 (1951).
5. A. Craya. Critical Regimes of Flows with Density Stratification. *Tellus* 3 28-42 (1951).
6. R. R. Long. The Motion of Fluids with Density Stratification. *J. Geophys. Research* 64 2151-63 (1959).
7. C. S. Yih. Effect of Density Variation on Fluid Flow. *J. Geophys. Research* 64 2219-23 (1959).
8. W. R. Deblor. Stratified Flow into a Line Sink. *Proc. Am. Soc. Civil Engrs.* 85 51-65 (July 1960).

SOIL TEMPERATURE STUDIES

James E. Carson*

Introduction

During the past few years, the effects of meteorological parameters on the thermal conditions in the soil have been studied by determining the diurnal and annual cycles, the extremes, and the day-to-day and hour-to-hour changes in soil temperature.⁽¹⁻³⁾ From these analyses it became possible to calculate the amount of heat stored within the soil during the course of the annual heating cycle. Also, very detailed information on the soil's response to weather elements was obtained through the use of case studies. This paper, which represents an extension of the previous work, deals primarily with the rate of exchange of heat energy between the ground and the atmosphere, and the analyses presented are based on recent laboratory determinations of hitherto unavailable measurements of moisture and heat parameters for the Argonne site.

Since the atmosphere is heated by the sun primarily by way of the soil surface,⁽⁴⁾ any investigation into the heat economy of the atmosphere, either on a local (such as the diurnal heat exchange and the forecasting of minimum temperatures and frost) or on a global scale (weather systems and the general circulation), must consider the thermal properties of the underlying medium. Several investigations of the large-scale effect of the heat exchange between the air and soil have appeared in recent literature.⁽⁵⁻⁹⁾

The surface layer of soil acts as a heat reservoir - absorbing part of the excess heat of midday and summer, and returning it during the night and winter to the atmosphere. This storage of energy lessens the temperature extremes of the atmosphere and acts as a climatic stabilizer. The heat-storage capacity of the soil is an important factor in the modification of air masses invading a region.⁽⁶⁾

During the day, when the soil surface is usually much warmer than the air above, a steep lapse rate is formed in the atmosphere. This unstable condition leads to air convection, turbulence, and gustiness of the wind, and through these motions, to the removal of water vapor from the air-soil interface. At night, however, the cool soil surface creates a stable lapse rate and may bring about a major pollution problem, due to the lack of turbulence. On the other hand, if the air is sufficiently cooler than the

*Resident Research Associate, Argonne National Laboratory, summers of 1957, 1958, 1959, and 1960. Assistant Professor of Physics at Iowa State University, Ames, Iowa.

soil, it may be possible for enough heat to flow upward to prevent or inhibit the formation of a nocturnal inversion and thus reduce the concentration of pollutants near the ground. Rider⁽¹⁰⁾ has shown that no inversion formed above a plot of bare soil on a clear night, while an inversion did form over an adjacent grass field. He attributes the lack of an inversion over the bare soil to the return of the heat stored in the soil during the day. This example underlines the importance of the grass cover to the microclimate. Kuhn et al.⁽¹¹⁾ have given evidence that local "hot spots" in the surface thermal patterns are closely related to the place of formation of tornadoes. Sanders^(12,13) has shown that, with the arrival of a cloud sheet at night, the heat stored in the soil during the day is often sufficient to evaporate a water fog if the soil at the 2-inch depth is only 2°F warmer than the air above, or to remove frost. Petterssen and Calabrese⁽⁸⁾ have recently pointed out the role of local heat sources, such as the Great Lakes in winter, on the motion and development of large weather systems.

Heat Storage in the Soil

The amount of heat energy stored (or lost) in a unit area of soil during a given time interval can be calculated if the temperature distribution within the soil at the beginning and end of the period and the heat capacity of the soil as functions of depth are known. The heat stored in a unit area of soil, ΔQ (cal/cm²), is

$$\Delta Q = \int_0^{\infty} \rho s \Delta T dz = \int_0^{\infty} C \Delta T dz \quad (1)$$

where ρ is the soil density, s is the specific heat of the soil, C is the volumetric heat capacity ($C = \rho s$), and z increases with depth. The lower limit of integration is the soil surface, $z = 0$; the upper limit in practice is such a depth z that the temperature change during the time interval is zero. This technique of determining the heat flux at the earth's surface is known as the temperature-integral method.

A quantity of considerable interest to meteorologists and climatologists is the amount of heat energy stored in the soil during the summer and then returned to the atmosphere during the cooler season; this quantity can be computed from the above formula. Figure 28 is a plot of the warmest and coolest decades at each of the seven soil temperature measuring levels for each of the three calendar years used in this study (1953, 1954, and 1955) on a logarithmic depth scale. This ordinate scale was chosen since the depth at which the annual cycle of soil temperature is reduced to essentially zero can be determined by the intersection of the curves representing the highest and lowest temperatures. This level at the Argonne site is found at 11 to 12 meters.

Figure 28

Extremes of soil temperature at indicated depths
based on 10-day averages

In order to compute the heat storage per unit area from Equation (1), allowance must be made for the exponential decrease of the annual temperature range of soil temperature with depth. A plot of the logarithm of the annual temperature range on a linear depth scale was used to estimate the annual range at depths between observational levels. The heat storage is given by the area enclosed by two extreme temperature profiles and the ground surface ($z = 0$), multiplied by the heat capacity per unit volume of the soil. Since the temperature change with seasons had not reached zero at the deepest level measured, it was necessary to extrapolate the curve downward and add this area to the region determined by direct measurements.

The soil at Argonne is yellow clay of glacial origin with a cover of pasture grass. Kersten⁽¹⁴⁾ found that the volumetric heat capacity of silty clay loam varied from 0.44 cal/cm³/deg C at low moisture contents to 0.70 for nearly saturated soil. Pasquill⁽¹⁵⁾ found an average value of 0.56 cal/cm³/deg C for clay pasture soil in England. Ingersoll, Zobel ' and Ingersoll⁽¹⁶⁾ for a sandy clay with 15 per cent moisture content reported a value of 0.59 cal/cm³/deg C.

In a previous report of this series,⁽³⁾ the amount of heat absorbed in the Argonne soil during the warmer half of 1955 was computed using Equation (1), the observed temperature values, and an assumed value of volumetric heat capacity. The storage in the soil in 1955, using Pasquill's value for the volumetric heat capacity, 0.56, was found to be 4,190 cal/cm². Rather than using an assumed value of heat capacity, it was deemed desirable to undertake a program for determining the thermal capacity of the soil at Argonne.

Measurements of Argonne Soil Samples

Three determinations of the soil moisture, bulk density, and the specific heat of the soil under widely differing soil moisture conditions have been made. The first sample was made on September 4, 1959 after a prolonged hot, dry period. August 1959 was the second hottest August on record in Chicago, averaging 6.0°F warmer than normal. After a heavy rain on August 3rd, only small amounts of rain fell during the month prior to the date of the soil sample, as seen in Table 31. As a result of the light rainfall, high temperature, and strong sunshine, the top soil layer was extremely dry.

Table 31

Daily rainfall totals at Argonne National Laboratory
in the period preceding soil samples.

Sample A		Sample B	
August 3, 1959	1.38 inches	October 2, 1959	0.02 inches
August 7, 1959	0.20	October 4, 1959	0.36
August 15, 1959	0.23	October 5, 1959	0.39
August 16, 1959	0.16	October 6, 1959	1.06
August 26, 1959	0.06	October 8, 1959	0.55
August 28, 1959	0.03	October 14, 1959	0.14
		October 23, 1959	0.43
		October 24, 1959	0.43
		October 26, 1959	0.10
Date of Sample: September 4, 1959		Date of Sample: October 27, 1959	

The second sample was taken on October 27, 1959, after the soil moisture content was recharged from the heavy rains of late September and October. The daily rainfall totals in October prior to this sample are also given in Table 31. The third sample was taken from under a deep blanket of snow on February 19, 1960. A storm on the 9 and 10th of February 1960, which began as snow and changed to freezing rain and then to sleet, left a total of 0.94 inches of precipitation and a snow cover of 7 inches. The air temperature remained below the freezing point until the day of the sample, except for a short period on the 16th, when the air temperature reached a high of 34.2°F. The snow cover gradually decreased to a depth of 4 inches by the date of the sample, due to ablation and compaction. The top several inches of the soil were observed to be frozen when the sample was obtained. It is clear that this and the second sample were taken when the soil moisture was near the maximum possible. The very dry sample taken on September 4, 1959, was analyzed by the author; the remaining two samples were analyzed by the Testing Service Corporation of Lombard, Illinois.

A power-driven auger was used to drill a borehole 3 feet in diameter to a depth of about 10 feet. Two soil samples at each of 6 depths were then collected for each determination. Thin-walled stainless steel cylinders (about 15 inches long and $2\frac{1}{4}$ inches inside diameter) were used to obtain the first sample at each depth. These cylinders were driven horizontally into the soil with a hammer. The weight and volume of these relatively undisturbed samples were then used to determine the bulk density of the moist soil. After the sample was removed from the tube and oven dried at 110°C, the moisture content and dry density, ρ_d , of the sample were computed. The second sample at each level, used to measure the specific heat and moisture content of the soil, was collected from chunks of soil at the proper depth and kept in tightly-sealed cans until analyzed.

The specific heat of the soil samples collected on September 4th was measured by a calorimetric technique. Each sample was carefully weighed and then chilled for about 72 hours in a refrigerator to about 36°F. The contents of the cans were then carefully placed in water slightly above room temperature and the contents well stirred; the method of mixtures was then used to determine the specific heat of the sample of moist soil. The specific heat of the other two sets was obtained using a different procedure. The soil samples were first oven-dried, powdered, and then placed in thin-walled copper containers. The specific heat of the dry soil in the containers was then measured in a precision calorimeter. This technique is in accordance with ASTM C 351-54T specifications and is certainly more accurate than the method used on the first set. The results of these determinations are given in Table 32.

Several difficulties, in addition to those normally associated with measurement of specific heat, are encountered in attempting to measure soil properties. One difficulty is the destructive nature of the test which

precludes repeated observations in the same location. It is impossible to replace the soil exactly and it is therefore necessary to take each set of samples at a different location. The presence of a borehole also will change the moisture-temperature distributions in the surrounding soil for many years. Hence, samples should not be obtained near permanent soil-temperature-measuring sites. The boreholes used in this report were located approximately 50 feet to the east of the Thermohms used to measure soil temperature. Another problem associated with soils is the lack of horizontal homogeneity of the soil; this condition is pronounced in a glacial soil such as is present at Argonne. Thus, the samples used in this study may or may not be representative of the soil profile at the temperature-measuring site. Another unavoidable difficulty is the change in compaction of the soil when the stainless steel cylinders are driven into the soil. Considerable force is necessary to insert and remove these tubes from the clay soil. This, of course, changes the density of the sample as compared to the undisturbed values. The samples were quickly placed in tight containers to keep the loss of soil moisture, due to evaporation, to a minimum.

Table 32

Thermal properties of the soil

Depth, inches	Moisture content, % dry weight					Dry specific heat, cal/g/deg C				Moist specific heat, cal/g/deg C			
	B	C	D	E	F	A*	D	E	F	A	D*	E*	F*
4	10.6	26.8	22.6	27.4	22.6	0.219	0.205	0.216	0.206	0.294	0.352	0.352	0.385
8	9.9	18.7	19.3	22.1	21.7	0.191	0.198	0.210	0.208	0.264	0.328	0.349	0.353
20	17.8	23.8	22.8	27.5	28.0	0.224	0.211	0.211	0.210	0.341	0.357	0.383	0.396
36	14.3	12.6	14.4	19.9	16.9	0.207	0.202	0.203	0.232	0.306	0.302	0.343	0.335
69	16.4	16.0	15.8	16.4	16.9	0.214	0.208	0.200	0.218	0.325	0.316	0.331	0.313
108	15.5	14.5	15.1	17.6	17.3	0.215	0.216	0.201	0.194	0.321	0.319	0.313	0.321

Depth, inches	Dry bulk density g/cm ³			Moist bulk density, g/cm ³			Volumetric heat capacity, cal/cm ³ /deg C			
	B**	C	E	B	C	E	B	C***	E	F†
4	1.28	1.45	1.52	1.42	1.83	1.87	0.417	0.644	0.658	0.672
8	1.51	1.67	1.59	1.66	1.98	1.94	0.438	0.649	0.677	0.679
20	1.55	1.48	1.47	1.83	1.83	1.88	0.624	0.653	0.720	0.722
36	1.77	1.78	1.82	2.02	2.00	2.12	0.618	0.604	0.727	0.677
69	1.80	1.70	1.68	2.10	1.97	1.97	0.683	0.622	0.652	0.620
108	1.82	1.82	1.82	2.10	2.09	2.14	0.674	0.699	0.670	0.681

Sample Date
 A September 4, 1959, can
 B September 4, 1959, tube
 C October 27, 1959, tube
 D October 27, 1959, can
 E February 19, 1960, tube
 F February 19, 1960, can

*Computed from the formula $s = s_d + W/1 + W$ where s and s_d are the specific heats of wet and dried soil samples and W is the moisture content of the soil

**Compiled from the formula $\rho = \rho_d (1 + W)$ where ρ_d and ρ are the dry and moist bulk densities

***The specific heat of sample C was not measured. The specific heat of dry soil for sample D was used in the calculation.

†The bulk density of sample F was not measured. The value listed in this column was calculated from the dry bulk density values from sample E and the moisture and specific heat values of sample F.

The specific heat of moist soil, s , depends upon the specific heat of the dry matter, s_d , and the moisture content. Repeated experiments have shown that most mineral soils, such as gravel, clay, silt, and sand have approximately the same dry specific heat. Most of the values fall in the range of 0.18 to 0.22 cal/g/deg.⁽¹⁴⁾ The value of the specific heat of moist soils is given by

$$s = \frac{s_d + W}{1 + W} \quad (2)$$

where W is the moisture content of the soil (expressed as the mass of water per unit mass of the dry soil). This formula follows since the specific heat of water, by definition, is unity.

The bulk density of natural soil depends upon the dry density and the moisture content. The dry density depends upon the nature of the soil material and the degree of compaction. The bulk density of moist soil is given by the relation

$$\rho = \rho_d (1 + W) \quad (3)$$

where ρ_d is the dry density and W is the moisture content, as before. As can be seen from these equations, the value of the volumetric heat capacity of moist soil, C , is therefore

$$C = \rho s = \rho_d (s_d + W) \quad (4)$$

Thus, the problem of measuring the heat capacity of soil is essentially that of measuring its moisture content.

The results of these calculations are included in Table 32. There is some scatter in the values of dry specific heat, but the points are in agreement with the findings of other investigators.

The value of the volumetric heat capacity assumed in the author's 1959 report,⁽³⁾ 0.56 cal/cm³/deg C, is obviously too low. A more realistic set of values for the volumetric heat capacity profile, namely, 0.64 from surface to 20 cm, 0.66 from 20 cm to 10 feet, and 0.68 from 10 feet to the depth where the annual temperature cycle is completely damped out, gives a value of the annual heat storage of 4,980 cal/cm².

The use of ten-day average temperatures, rather than extreme daily values, slightly lowers the area enclosed in Figure 28 and thus, produces a lower value of the heat storage. The computed value also does not include the amount of heat energy needed in spring to melt the frozen soil and released in the fall when the soil freezes. If it is assumed that the soil at

Argonne freezes to a depth of about 25 cm (the depth of intersection of the 0°C isotherm and the left temperature profile of Figure 28) and has a typical winter soil-moisture content, an additional heat exchange of 400 to 500 cal/cm² is indicated.

This calculation of the heat stored under a unit area of soil from the time of the minimum in winter to the maximum temperature profile in summer gives a value considerably greater than the values given by Kuhl⁽¹⁷⁾ and Schubert.⁽¹⁸⁾ Kuhl stated that the heat storage at latitudes above 35° is only 1500 to 2500 cal/cm². For a sandy soil at Eberswalde, near Berlin, Schubert found that the annual heat storage in grass-covered soil was 1850 cal/cm².

Heat Flow Method

Another estimate of the amount of heat stored within the soil may be obtained by applying the theory of heat flow into a semi-infinite, homogeneous medium. The rate at which heat energy is conducted through a unit area of soil is

$$S = - \lambda \frac{\partial T}{\partial z} \quad (5)$$

where λ is the thermal conductivity of the soil, z is depth, and T is temperature. The equation can be integrated with respect to time to obtain the amount of heat passing through the soil during the interval, if λ and T as functions of time and depth are known.

The annual temperature cycle at the soil surface can be approximated by a sinusoidal wave

$$T(0,t) = \bar{T} + \Delta T_0 \sin \left(\frac{2\pi}{\tau} t + \Phi \right) \quad (6)$$

where $T(0,t)$ is the temperature of the soil surface at time t , $\bar{T}$ is the average annual temperature, ΔT_0 is the amplitude of the annual cycle at $z = 0$, τ is the period of the cycle (one year) and Φ is the phase angle of the temperature wave. If one assumes that the soil is a semi-infinite, homogeneous medium, the temperature at any depth z and time t is

$$T(z,t) = \bar{T} + T_0 (\exp - z \sqrt{\pi/\tau\kappa}) \sin \left(\frac{2\pi}{\tau} t + \Phi - z \sqrt{\pi/\tau\kappa} \right) \quad (7)$$

where $\kappa = \lambda/C$ is the thermal diffusivity of the soil. The rate of phase shift and amplitude decrease are functions of the period of the wave and the thermal diffusivity of the soil.

Substituting Equation (6) into (5) at the soil surface and integrating over one-half of a temperature cycle, one obtains the flow of heat per cycle through the surface per unit area⁽¹⁶⁾

$$\Delta Q = \Delta T_0 \sqrt{\frac{2\tau}{\pi}} C \sqrt{\kappa} = \Delta T_0 \sqrt{\frac{2\tau C \lambda}{\pi}} \quad (8)$$

This equation shows that the heat storage per unit area is proportional to the square root of the product of the period and the conductive capacity of the medium. Using representative values for κ and C ($0.005 \text{ cm}^2/\text{sec}$ and $0.64 \text{ cal/cm}^3/\text{deg C}$ respectively) and the observed 1955 1-cm amplitude, 13.9°C , this equation gives an annual heat storage of only $2,830 \text{ cal/cm}^2$. Since the amplitude of the temperature wave at 1 cm is less than that at the surface, this computation somewhat underestimates the heat storage.

In deriving Equation (8), it was assumed that heat is transported in the soil by molecular conduction only, and that the soil is a homogeneous, semi-infinite medium. Since these assumptions are not valid in the soil, one cannot expect this calculation to give the same value for the heat storage as the more exact Equation (1).

Annual Heat Exchange Cycles

Equation (1) has been used to estimate the annual and diurnal cycles of heat exchange in the soil at Argonne. In order to make this calculation, it was necessary to assume reasonable values of the volumetric heat capacity since adequate soil moisture data are not available for the period of record. The values of heat capacity listed in Table 32 are close to the extreme limits of this parameter since they were obtained under conditions of very dry or very wet soil. With these values and the rainfall record, a representative value of C was estimated each month for each of the levels where the soil temperature was measured. It was further assumed that the values of C at 10 and 29 feet were constant through the year and equal to 0.70 and $0.71 \text{ cal/cm}^3/\text{deg C}$ respectively. It was also necessary to assume a linear variation of temperature change and heat capacity between levels.

The programs for these calculations were prepared by J. S. Dick of the Applied Mathematics Division for the GEORGE electronic computer. Two slightly different techniques were used to estimate the annual cycle of heat storage in the soil. In the first program, the value of ΔT used was the change of average monthly temperature from one month to the next at each of the seven levels. Figure 29 is a plot of the vertical temperature profiles, based on monthly averages, for each month in 1953. The data for the 29-foot level, which varied from 50.2°F in July and August, to a high of 52.5°F in March, were omitted for clarity. The slope of these profiles determines the direction and magnitude of the vertical heat flux. The net amount of heat entering or leaving the deeper soil layers during a month

is given by the area enclosed by the two temperature profiles, corrected for the heat capacity of the various soil layers.

Figure 29

Soil temperature profiles for 1953 based on monthly averages.

The accuracy of this calculation depends, of course, on the accuracy of the heat capacity values used. It is quite possible that these values are in error by as much as 20 per cent. The results of these calculations are given in Tables 33 and 34. The data in the first table show the amount of heat energy gained or lost by the soil per unit area per day.

Table 33

Average daily value of the heat gained
or lost by the soil profile.*

Month	Year			Month	Year		
	1953	1954	1955		1953	1954	1955
January	-23.4	-30.1	-25.2	July	+29.9	+31.5	+31.6
February	-17.5	-17.2	-18.0	August	+18.1	+18.5	+23.8
March	- 2.5	- 4.5	- 6.8	September	+ 3.5	+ 3.0	+ 0.1
April	+10.5	+12.6	+16.1	October	-14.0	-13.0	-17.9
May	+26.2	+25.2	+32.6	November	-25.9	-33.7	-34.4
June	+38.8	+38.1	+27.6	December	-30.7	-33.9	-34.8

*Data based on the change of average monthly soil temperature, cal/cm²/day.

Table 34

Heat gains and losses from the soil during the course of a year.*

	Average monthly			Last day of month		
	1953	1954	1955	1953	1954	1955
Gains	+3886.2	+3869.3	+4003.8	+4165.1	+4111.1	+4177.3
Losses	-3455.2	-4020.2	-4164.9	-3806.6	-4142.5	-4236.6
Net	+ 431.0	- 150.9	- 161.1	+ 358.5	- 31.4	- 59.3

*cal/cm²

The mean monthly temperature can be considered as typical of conditions in midmonth. Thus, for example, the area in Figure 29 bounded by the temperature profiles for May and June and the soil surface represents the heat gained by the soil per unit area between mid-May and mid-June. Thus the data in Table 33 for a given month represent the average

amount of heat gained or lost by a unit area per day for the period ending in the middle of the month listed. This table shows that the maximum heat exchange in the soil comes before the extreme temperatures are reached, as predicted by heat flux theory.

The interval from the middle of one month to the middle of the next is not the most convenient one for determining heat storage in the soil since most climatological summaries use the calendar month as the unit of time. The amount of heat gained or lost by a unit area of soil in a given month can be computed by means of Equation (1), the temperature change from the last day of the preceding month to the last day of the current month, and assumed heat capacity values. The average temperature on the last day of each month was used to compute ΔT at the seven levels. The results are listed in Tables 34 and 35 and Figure 30. The data in Table 35 show the amount of heat gained or lost per day during the calendar month. It can be seen that there is little heat exchange between air and the soil in March and September. The vertical temperature gradients in the soil during these months were near zero (Figure 29). However, the temperature of the air and upper soil layers is changing rapidly during these months. The maximum fluxes to and from the soil come at or just before the time of maximum and minimum insolation. The data in Table 35 show that abnormally warm and cold months are reflected in the soil heat flux pattern.

Table 35

Average daily value of the heat gained
or lost by the soil profile.*

Month	Year		
	1953	1954	1955
January	-17.4	-23.3	-20.3
February	-14.5	- 6.2	- 9.7
March	+ 4.9	- 7.4	- 4.4
April	+15.1	+34.4	+33.3
May	+39.4	+22.6	+31.4
June	+40.0	+44.6	+28.9
July	+22.1	+23.2	+32.8
August	+14.5	+10.3	+10.2
September	- 9.5	- 4.6	- 8.0
October	-20.3	-29.8	-24.6
November	-30.2	-31.3	-39.9
December	-33.5	-32.7	-31.8

*Data based on the change of soil
temperature on the last day of
each month, cal/cm²/day

Figure 30

Annual cycle of heat flux in soil. Data are based on three-year period, 1953-1955 and represent the change of heat content in the soil from the surface to 29 ft during the month indicated.

Figure 30 shows the annual march of soil heat exchange. This figure is based on three-year averages for each month. This curve is quite similar to the one shown by Pearce and Gold⁽¹⁹⁾ for the soil heat flux at Ottawa, Canada, measured with a heat meter.

Another estimate of the amplitude of the annual heat storage is given in Table 34. This table is based on the data from Tables 33 and 35 and represents the total heat gained (or lost) by the soil during the six months of heat gains (or losses). These values underestimate the amplitude of the annual heat exchange because of the averaging process. The heat storage values indicated in this table are about 20 per cent smaller than that determined in the first calculation (4,980 cal/cm²). Table 34 also shows that the heat gains and losses computed using the data for the last day of the month are larger than those computed from monthly averages.

The net accumulation during a given year would seem to indicate that the soil became warmer in 1953 and cooler in 1954 and 1955. Since the net values are small, a more probable explanation is the unrepresentativeness of assumed heat capacity values.

Diurnal Heat Exchange Cycles

The magnitude of the daily heat exchange can also be estimated from Equation (1). For this calculation, the average daily range of soil temperature at 1, 10, 20, and 50 cm for each month of the three-year period was used to estimate the daily change in temperatures. The values of C used to prepare Tables 33 and 35 were also used in this calculation. The results of this calculation are given in Table 36.

Table 36

Amplitude of the diurnal cycle of heat
exchange between air and soil.*

January	11.7	July	90.4
February	15.4	August	77.7
March	45.9	September	68.5
April	74.6	October	58.4
May	86.9	November	41.4
June	85.2	December	15.6

*cal/cm²

The data in Table 36 appear at first to be too large. For example, Suomi, Portman, and others who measured the daily exchange of heat between the soil and atmosphere on clear days in late summer during the "Project Great Plains" at O'Neill, Nebraska, reported values of only 60 to 70 cal/cm².⁽²⁰⁾ However, two processes are in operation to create the observed daily temperature range in the soil; the daily cycle of net radiation and air mass changes. The data in Table 36 indicate that reduction of the diurnal heat exchange on cloudy days without an air mass change is more than balanced by the much larger heat flows occurring on days with strong air mass advection.

The vertical temperature profiles in the soil during the course of a typical sunny day in summer (June 27, 1955) are shown in Figure 31. The strong temperature gradient observed in the upper soil was directed downwards during the day. During the night, the soil surface cooled rapidly and the gradient was upwards above 50 cm. The strong temperature gradient from 50 to 100 cm indicates a strong flux of heat into the deeper soil levels during the entire day.

Figure 31

Vertical temperature profiles in soil during the course of a summer day, July 27, 1955.

The daily cycle of heat exchange between the air and soil was computed from Equation (1), the observed average monthly hour-to-hour temperature changes, and assumed values of C . Figure 32 shows the results of this calculation for alternate months in 1953. The heat flux patterns are quite similar to those of the net radiation. The maximum flux into the soil is reached at or just before noon (the time of maximum net and solar radiation) but several hours before the air and soil-surface temperature maxima. The flux is negative from about 1600 CST and remains negative for several hours after sunrise. The heating part of the cycle is thus much shorter and more intense than the cooling. The shapes of the curves compare quite favorably with actual measured values, such as those reported by Lettau.⁽²⁰⁾

Figure 32

Daily cycle of heat flux in the soil. Data based on average values of the one-hour temperature change for the hour ending as indicated and on assumed values of soil heat capacity. Curves smoothed slightly.

Summary

The amount of heat stored in the soil during various time intervals has been computed by the temperature-integral method. These estimates are not very accurate since it was necessary to use values of heat capacity of the soil based on estimated values of soil moisture. Measurements of the heat capacity of the ground were made for very wet and very dry conditions. These values plus the rainfall record were used to estimate the soil moisture, which in turn determines the heat capacity.

Calculations of the heat stored in the ground in summer and returned to the atmosphere in winter, as well as estimates of the annual and diurnal cycles of heat exchange between soil and the atmosphere, were made. The results of these calculations agree fairly well with the theory. On a daily basis, the march of heat flux in the soil is quite similar to that of net radiation.

References

1. J. E. Carson and H. Moses. Soil Temperature Studies. Radiological Physics Division Semiannual Report, July through December, 1957. ANL-5829, pp. 187-199.
2. J. E. Carson and H. Moses. Soil Temperature Studies. Radiological Physics Division Semiannual Report, July through December, 1958. ANL-5967, pp. 210-218.
3. J. E. Carson. Soil Temperature Studies. Radiological Physics Division Semiannual Report, January through June, 1959. ANL-6049, pp. 55-68.
4. H. R. Byers. General Meteorology, 3rd. ed. New York; McGraw-Hill Book Co., Inc., 1959.
5. R. Geiger. The Climate Near the Ground, 2nd rev. printing, translation by Milroy N. Stewart, et al., Cambridge: Harvard University Press, 1957.
6. C. H. B. Priestley. The Evolution of Energy Gain by the Atmosphere Through Contact with the Ground or Ocean. Sci. Rept. No. 2. Contract AF 19(604)-2179, Univ. of Chicago, 1957.
7. C. H. B. Priestley. Turbulent Transfer in the Lower Atmosphere. Chicago; University of Chicago Press, 1959.
8. S. Petterssen and P. A. Calabrese. On Some Weather Influences Due to Warming of the Air by the Great Lakes in Winter. J. Meteorol. 16 646-652 (1959).
9. M. I. Budyko. The Heat Balance of the Earth's Surface. Leningrad, 1956.

10. N. E. Rider. A Note on the Physics of Soil Temperature. *Weather* 12 241-246 (1957).
11. P. M. Kuhn, G. L. Darkow, and V. E. Suomi. A Mesoscale Investigation of Pre-Tornado Thermal Environments. *Bull. Am. Meteorol. Soc.* 39, 224-228 (1958).
12. W. E. Saunders. The Clearance of Water Fog Following the Arrival of a Cloud Sheet During the Night. *Meteorol. Mag.* 89 8-10 (1960).
13. W. E. Saunders. The Clearance of Frost Following the Arrival of a Cloud Sheet During the Night. *Meteorol. Mag.* 89 45-51 (1960).
14. M. S. Kersten. Thermal Properties of Soils. University of Minnesota Engr. Expt. Sta. Bull. 28 (1949).
15. F. Pasquill. Eddy Diffusion of Water Vapour and Heat Near the Ground. *Proc. Roy. Soc. (London)* A198 116-140 (1949).
16. L. R. Ingersoll, O. J. Zobel, and A. C. Ingersoll. Heat Conduction, rev. ed. Madison: University of Wisconsin Press, 1954.
17. W. Kuhl. Der Jahrliche Gang der Bodentemperatur in Verschiedenen Klimaten. *Ger. Beit. Geophys.* 8 499-564 (1907).
18. J. Schubert. Der Warmeaustausch im Festen Erdboden. Berlin: Springer, 1904.
19. D. C. Pearce and L. W. Gold. Observations of Ground Temperature and Heat Flow at Ottawa, Canada. *J. Geophys. Research* 64 1293-1298 (1959).
20. H. H. Lettau. Computation of Heat Budget Constituents of the Earth/Air Interface. Exploring the Atmosphere's First Mile ed H. H. Lettau and B. Davidson. London: Pergamon Press, 1957, pp. 305-327.

PUBLICATIONS

July through December 1960

- Berlman, I. B. Energy Transfer in a Solution of PPO in Xylene. J. Chem. Phys. 33 1124-1127 (October 1960).
- Berlman, I. B., R. Grismore and B. Oltman. An Improved "Twin" Scintillation Fast Neutron Detector. Rev. Sci. Instr. 31 1198-1200 (November 1960).
- Berlman, I. B. Luminescence in a Scintillation Solution Excited by Alpha and Beta Particles and Related Studies in Quenching. J. Chem. Phys. 34 216 (1961).
- Grismore, R. Radiation, in The American Peoples Encyclopedia Yearbook, ed. R. C. Hunt. Chicago: Spencer Pres, Inc., 1960, pp. 843-845.
- *Gustafson, P. F. Assessment of Radiation Dose Due to Fallout. Radiology 75 282-288 (August 1960).
- Gustafson, P. F. Fallout Transport as Deduced from Measurement of Gamma-Emitting Radioactivity in Air. Health Physics 4 201 (December 1960) (Abstract).
- Gustafson, P. F., S. S. Brar and M. Kerrigan. Appearance of a Spring Maximum in Nuclear Test Debris in 1960. Science 133 460-461 (1961).
- Rowland, R. E. The Deposition and Removal of Radium in Bone by a Long-Term Exchange Process. Clinical Orthopaedics 17 146-153 (Summer 1960).

* Division of Biological and Medical Research.

PAPERS TO BE PUBLISHED

- Berlman, I. B. Transient Dimer Formation by 2, 5-diphenyloxazole. J. Chem. Phys. (Note)
- Berlman, I., and A. Weinreb. Fluorescence Spectrum and Decay Time of Naphthalene. Molecular Physics.
- *Gustafson, P. F., M. Kerrigan and S. S. Brar. Airborne Fallout Radioactivity. Science.
- Marinelli, L. D. Dosimetrie. Chapter III-1, in Handbook of Medical Radiology, Vol. 11. Heidelberg: Springer-Verlag.
- Miller, C. E. Sensitivity of Scintillation Scanners. Symposium on Radioactivity in Man, Vanderbilt Univ., Nashville, Tenn., April 1960, ed. G. Meneely. Springfield, Illinois: C. C. Thomas.
- Moses, H., and G. H. Strom. A Comparison of Observed Plume Rises with Values Obtained from Well-known Formulas. J. Air Pollution Assoc., and Proceedings of Air Pollution Control Association.
- Moses, H., F. C. Kulhanek and K. W. Clarke. The Argonne Meteorological Data Processing System. Bull. Am. Meteorological Soc.
- Moses, H. Meteorological Research at the Argonne National Laboratory. (Abstract)
- Moses, H. Meteorological Standards for Reactor Site Selection. (Abstract)
- Rowland, R. E. Microscopic Metabolism of Ra^{226} in Canine Bone and its Bearing Upon the Radiation Dosimetry of Internally Deposited Alkaline Earths. Radiation Research.

* Division of Biological and Medical Research.