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A DECAY-TIME STUDY OF AN IRRADIATED SCINTILLATING SOLUTION PART I

I. B. Berlman and M. Zarr*

Introduction

The luminescent efficiency of a scintillating solution decreases with radiation dose. This study examines this effect in terms of quenching of the excited solvent and scintillator molecules by the substances produced in the solution by the radiation. The quenching produced by the radiation products is expressed empirically from the results of luminescent decay-time studies.

Apparatus

The decay-time of luminescence produced in control and irradiated solutions was measured with the apparatus built by R. K. Swank et al., and described in Reference 1. In this apparatus a pulse of electrons is made to impinge on a luminescent sample for a tenth of a millimicrosecond. The decay time of the resulting luminescent pulse is then accurately measured. A feature of the decay-time apparatus is that the measured decay time is a measure of the slowest step in the scintillation process. For xylene-diphenyloxazole (PPO) solutions with scintillator concentrations less than 2 g/l the slowest step is the energy transfer process from the solvent to the solute, while for solutions with concentrations greater than 3 g/l the transfer time becomes very short, and the slowest step is the decay time of the excited PPO molecule itself.

Special wafer-like quartz cells were made to hold the xylene-PPO solutions. Each cell has a 3/16-inch hole in the center of the top surface through which the electron beam impinges on the solution. The hole is covered with an aluminum foil 0.00025 inches thick to prevent atmospheric oxygen from diffusing into the solution. The sample solutions are 2 mm thick. Under certain conditions this thickness is still excessive because energy absorption and re-emission can take place repeatedly in that distance. If these processes are possible, then the measured lifetime of light emission will appear too long. To reduce the effect of absorption and re-emission, the vertical dimension of the solution is reduced by partially filling the cell and the horizontal dimension is limited to 1/16 inch by positioning a hollow stainless steel cylinder in the hole of the cell.

*Student Aide, Summer 1958.

Theory

T_0 , the mean scintillation lifetime of double-distilled and nitro-generated xylene, can be expressed as:

$$T_0 = \frac{1}{p_e + p_q} \quad (1)$$

where p_e is the probability/sec that an excited xylene molecule will emit fluorescent light and p_q is the probability/sec that the excited molecule will be de-excited without the emission of any fluorescence; p_q includes internal and self-quenching possibilities. Furthermore, it is assumed that, in the process of measuring lifetimes of control and irradiated solutions, the electrons in the testing pulse do not form sufficient quencher products to affect the measurement appreciably.

Upon irradiation of xylene, it is postulated that relatively stable quencher molecules are produced which increase the probability of quenching by an amount p'_q . Hence, we may write for the mean lifetime of irradiated xylene the expression.

$$T'_0 = \frac{1}{p_e + p_q + p'_q} \quad (2)$$

In this case the quencher molecule is one of the radiation products. Therefore,

$$\frac{T_0}{T'_0} = \frac{p_e + p_q + p'_q}{p_e + p_q} = 1 + \frac{p'_q}{p_e + p_q} \quad (3)$$

It is assumed that the radiation products do not transfer excitation energy to xylene or PPO. The validity of this assumption is now being tested. Consider now the case of PPO-xylene solution where the PPO concentration is very small (≤ 1 g/l) and let T_2 be its mean lifetime. We may then write:

$$T_2 = \frac{1}{p_e + p_q + p_t}, \text{ and } \frac{T_0}{T_2} = 1 + \frac{p_t}{p_e + p_q} \quad (4)$$

where p_t is the probability/sec of energy transfer from an excited xylene molecule to a PPO molecule. If T'_2 is the mean lifetime of irradiated xylene to which a small amount of PPO has been added, then

$$T'_2 = \frac{1}{p_e + p_q + p'_q + p_t}$$

and

$$\frac{T_0}{T_2'} = \frac{P_e + P_q + P_q' + P_t}{P_e + P_q} = 1 + \frac{P_q'}{P_e + P_q} + \frac{P_t}{P_e + P_q} \quad (5)$$

For PPO concentrations greater than 3 g/l the measured scintillation lifetime is that of PPO alone because the transfer time is shorter than excitation lifetime of PPO. Let T_3 and T_3' be the mean lifetimes of an un-irradiated and an irradiated solution respectively, each containing the same large concentration of solute. In the latter case if the solute is added after the irradiation, then

$$T_3 = \frac{1}{P_e + P_i + P_q} \quad \text{and} \quad T_3' = \frac{1}{P_e + P_i + P_q + P_q'}$$

where P_e , P_i , P_q and P_q' are the probabilities/sec for emission of an excited PPO molecule, for internal quenching, for self-quenching, and for radiation product's quenching respectively. It follows,

$$\frac{T_3}{T_3'} = \frac{P_e + P_i + P_q + P_q'}{P_e + P_i + P_q} = \frac{1 + \frac{P_q}{P_e + P_i} + \frac{P_q'}{P_e + P_i}}{1 + \frac{P_q}{P_e + P_i}} \quad (6)$$

Results

The decay times for double-distilled, nitrogenated, (nonirradiated) xylene, and varying amounts of PPO are shown in Table 1. The indicated uncertainties are due to the variation in results obtained from many measurements. It is seen in column 2 that at PPO concentrations equal to greater than 2 g/l the measured decay time approaches a constant value, presumably that of PPO alone. The values in column 3 are computed from equation 4 and the energy transfer times shown in column 4 are computed by dividing T_0 by the successive values in column 2. Since the energy transfer time approaches the mean lifetime of an excited PPO molecule at large solute concentrations, it is impossible, therefore, to compute the transfer time under these conditions; hence, no tabulation is shown in column 3 at these concentrations. At very low concentrations it is noted in column 4 that the energy transfer time becomes longer than the mean lifetime of an excited xylene molecule at room temperature. The transfer time at low concentrations is being studied more thoroughly.

The decay times of irradiated xylene are compiled in Table 2. These solutions do not contain PPO. The values in column 3 are obtained from equation 3 and the values in the column 4 are obtained by dividing the data

Table 1

Concentration of nonirradiated PPO (g/l)	T ($\times 10^9$ sec)	$\frac{P_t}{p_e + p_q}$	$\frac{1}{p_t} (\times 10^9 \text{ sec})$
0	20.2 ± 0.7		
1/8	10.6	0.91	22.2 ± 0.7
1/4	7.73 ± 0.55	1.61 ± 0.3	12.5 ± 3.5
1/2	5.4 ± 0.1	2.74 ± 0.21	7.4 ± 0.9
1	4.25 ± 0.03	3.75 ± 0.2	5.4 ± 0.5
2	3.1		
4	2.74		
6	2.67		

in column 3 by the dose in units of 10^9 ergs/cm³. It is of interest to note that p_q' increases with dose although the uncertainty is too large to ascertain whether the increase is linear with dose (see column 3).

Table 2

Irradiated xylene

Radiation dose (ergs/cm ²)	T ($\times 10^9$ sec)	$\frac{p_q'}{p_e + p_q}$	$\left(\frac{p_q'}{p_e + p_q} \right) \text{ unit dose}^*$
0	20.2 ± 0.7	-	-
4.36×10^9	6.25	2.23 ± 0.11	0.51 ± 0.03
4.99×10^9	4.85	3.16 ± 0.15	0.63 ± 0.03
6.72×10^9	3.79 ± 0.02	4.33 ± 0.18	0.64 ± 0.03
6.78×10^9	5.24 ± 0.2	2.86 ± 0.28	0.42 ± 0.05

*Unit dose = 10^9 ergs/cm³

The decay times for irradiated xylene plus 0.5 g/l PPO are shown in Table 3. In this experiment first the xylene was irradiated and the PPO was added afterwards. The values in column 4 were obtained by means of equation 5. From these values, the transfer probability ratios shown in Table 1, column 3, were subtracted to obtain the data in column 5. The values in column 5 are divided by their respective doses in units of 10^9 ergs/cm³ to obtain the values in column 6. It is interesting to note that the sample which had received a dose of 1.67×10^9 ergs/cm³ has a value of $p_q'/(p_e + p_q)$ which is in the range of values shown in the last column in Table 2. This result would indicate that the quenching molecules act independently of the energy transfer.

Table 3
Irradiated xylene plus PPO

Conc. PPO (g/l)	Rad. dose (ergs/cm ³)	T (x 10 ⁹ sec)	$\frac{P_q'}{P_e + P_q} + \frac{P_i}{P_e + P_q}$	$\frac{P_q'}{P_e + P_q}$	$\left(\frac{P_q'}{P_e + P_q}\right) / \text{unit dose}$
1/2	3.4 x 10 ⁸	4.95	3.08 ± 0.15	0.34 ± 0.34	1.0 ± .1
1/2	1.67 x 10 ⁹	4.31	3.09 ± 0.16	0.95 ± 0.37	0.57 ± .22

The decay times for irradiated xylene and 4 g/l PPO are shown in Table 4. These measurements give the decay time of PPO in quenched and unquenched solutions. Unless otherwise stated, the PPO was added after the radiation. The values in column 4 were obtained from equation 6 where the self-quenching, P_q , is assumed to be very small at 4 g/l and is, therefore, neglected.

Table 4
Irradiated xylene plus PPO (at larger concentrations)

Conc. PPO (g/l)	Rad. dose (ergs/cm ³)	T (x 10 ⁹ sec)	$\frac{P_q'}{P_e + P_i}$	$\left(\frac{P_q'}{P_e + P_i}\right) / \text{unit dose}$
4	0	2.74	0	0
4	2.83 x 10 ⁹	2.39	0.15	0.053
4	4.40 x 10 ⁹	2.28	0.20	0.046
4*	2.83 x 10 ⁹	2.73	0.004	0.001

*PPO added before irradiation

It is interesting to note that the quenching probability of the PPO by the radiation products is about one tenth that of xylene (column 5). This is interpreted to mean that the G value for the production of quenchers of PPO is approximately a tenth that of xylene. Another point of interest is the result that there seems to be negligible quenching of the PPO when xylene and PPO are irradiated together as a solution.

In conclusion we may state that large radiation doses produce quencher molecules which affect the scintillation process by competing for the excess energy possessed by excited xylene and PPO molecules.

Thanks are due to Robert K. Swank for stimulating discussions, Eugene Mroz for operating the decay-time apparatus, to Warren Buck and Dr. Arye Weinreb for the use of their Beckman DK-2, and to Patricia Walsh for aid in making the radiation exposures.

Reference

1. R. K. Swank. Physics Division Summary Report, ANL-5754, p. 56 (1957)

LIGHT FLUX FROM CYLINDRICAL DISTRIBUTED SOURCES AND APPLICATIONS TO SCINTILLATING SOLUTION STUDIES

W. R. Anderson* and I. B. Berlman

I. Introduction

In the present study the liquid scintillating solution which is excited by external gamma radiation is contained in a small inverted right cylindrical cup, described elsewhere,⁽¹⁾ capped with a quartz window in contact with the multiplier phototube. If the gamma source is sufficiently distant to produce a collimated beam at the cup, then the light source can be considered uniformly distributed through the solution, if its dimensions are small compared to the mean free path of the γ -ray. The fraction of the total light flux emitted reaching the window is dependent, among other things, upon the absorption of the solvent and the absorption of the interior surface at each reflection. In the calculations herein, all factors other than these two are disregarded, appearing as variants between predicted and experimental results. Two cases lend themselves to direct treatment: (a) solution absorbent in a given degree, but surfaces perfect specular reflectors, and (b) solution absorbent in a given degree, and surfaces totally absorbent.

II. Cylinder with 100% Specularly-reflecting Surfaces

The question of how far a given photon travels through the solution before reaching the window, provided it is not absorbed in the process, is in this instance easily answered. Since reflection by a vertical surface does not change the angle a ray makes with a horizontal plane, a photon which starts out in any direction inclined below the horizontal continues the travel at that angle until it reaches the plane of the photocathode. The distance which the photon has travelled is equal to that between the point of origin and the plane of the window, along a line extended in the initial direction of travel; this is pictured for a simple case in Fig. 1 (a). A photon which starts out in any direction inclined above the horizontal travels a distance equal to that between the point of origin and the plane of the window of an image cylinder atop the real cylinder, along a line extended in the initial direction of travel; this is pictured for a simple case in Fig. 1 (b).

A simple equivalent would be an imaginary cylinder twice as high, from which the same total light is emitted, but all in directions below the horizontal. The equivalents of the two rays originating at a single point in Fig. 1 (a) and (b) are pictured in Fig. 1 (c) originated at the two equivalent points in the imaginary cylinder.

*Resident Research Associate from the University of Illinois, Chicago, Summer 1958.

FIGURE 1
EQUIVALENT RAYS

Let H be the height of the real cylinder and R its radius, let z be the vertical distance between point of origin of light and window, the latter being the plane $z = 0$, and let θ be the angle made with the vertical by the ray. Since, as was shown, it is the original direction of the light that is important, then, from Fig. 2, the light reaching the window from a unit volume at P in an increment of solid angle $d\Omega$ subtended by a circular element of radius r centered on a vertical axis through P and of width dr is

$$dE = \frac{E_0}{2\pi R^2 H} \frac{d\Omega}{2\pi} e^{-az \sec \theta}$$

where E_0 is the total light flux emitted in the cylinder and a is the light absorption coefficient of the material through which the light passes, it being assumed that Beer's law holds for the material. It should be remembered that, although in the plane of the window, the annulus-shaped area element may lie partly or wholly outside the real window.

FIGURE 2
ELEMENT OF INTEGRATION

The size of the solid angle can be written as

$$d\Omega = 2\pi \sin \theta d\theta$$

The light from unit volume at P reaching the window in $d\Omega$ is then

$$dE = \frac{E_0}{2\pi R^2 H} \sin \theta d\theta e^{-az \sec \theta}$$

and the total reaching the window from unit volume at P is

$$\frac{E_0}{2\pi R^2 H} \int_0^{\pi/2} e^{-az \sec \theta} \sin \theta d\theta$$

which is independent of the horizontal coordinates of P . The total received at the window from a horizontal section of scintillator of thickness dz is

$$\begin{aligned} & \pi R^2 dz \frac{E_0}{2\pi R^2 H} \int_0^{\pi/2} e^{-az \sec \theta} \sin \theta d\theta \\ &= \frac{E_0}{2H} dz \int_0^{\pi/2} e^{-az \sec \theta} \sin \theta d\theta \end{aligned}$$

The total received from the entire cylinder is

$$E = \frac{E_0}{2H} \int_0^{2H} \int_0^{\pi/2} e^{-az \sec \theta} \sin \theta d\theta dz$$

and the fraction T of the light reaching the window is

$$T = \frac{E}{E_0} = \frac{1}{2H} \int_0^{2H} \int_0^{\pi/2} e^{-az \sec \theta} \sin \theta d\theta dz$$

The equation may be rewritten

$$T = \int_0^{\pi/2} \frac{\sin \theta}{2H} \int_0^{2H} e^{-(a \sec \theta) z} dz d\theta$$

for initial integration over z. The equation then becomes

$$\begin{aligned} T &= \int_0^{\pi/2} \frac{\sin \theta}{2H} \left(\frac{1}{a \sec \theta} - \frac{e^{-2aH \sec \theta}}{a \sec \theta} \right) d\theta \\ &= \int_0^{\pi/2} \frac{\sin \theta \cos \theta d\theta}{2aH} - \int_0^{\pi/2} \frac{e^{-2aH \sec \theta} \sin \theta \cos \theta d\theta}{2aH} \\ &= \frac{1}{4aH} - \int_0^{\pi/2} \frac{e^{-2aH \sec \theta} \sin \theta \cos \theta d\theta}{2aH} \end{aligned}$$

In the remaining integral term, let $x = \sec \theta$

then $dx = \sec \theta \tan \theta d\theta$

and $d\theta = \frac{dx}{\sec \theta \tan \theta}$

and $\sin \theta \cos \theta d\theta = \frac{\sin \theta \cos \theta dx}{\sec \theta \tan \theta} = \frac{dx}{\sec^3 \theta} = \frac{dx}{x^3}$

$$\text{Then } \int_0^{\pi/2} \frac{e^{-2aH \sec \theta} \sin \theta \cos \theta d\theta}{2aH} = \int_1^{\infty} \frac{e^{-2aHx} dx}{2aHx^3} = \frac{1}{2aH} \int_1^{\infty} \frac{e^{-2aHx} dx}{x^3}.$$

Therefore,

$$T = \frac{1}{4aH} - \frac{1}{2aH} \int_1^{\infty} \frac{e^{-(2aH)x} dx}{x^3}.$$

The integral of the form of that in the second term has been evaluated by Mian and Chapman⁽²⁾ for various values in the range $2aH = 0$ to 6 . In the first column of Table 5 are given the values of $2aH$, in the second column their results, and in the last column the values of T calculated for each. These results for T are plotted in Fig. 3 and a working curve obtained therefrom. For values of $2aH$ greater than 6 , the $(2aH)^{-1}$ curve may be used.

Table 5

Calculation of T

$2aH$	$\int_1^{\infty} e^{-2aHx} x^{-3} dx$	$\frac{1}{2aH}$	$\frac{1}{2aH} \int_1^{\infty} e^{-2aHx} x^{-3} dx$	$\frac{1}{4aH}$	T
0	0.5000				1.00
0.01	0.4903	100	49.03	50	0.97
0.05	0.4549	20	9.098	10	0.902
0.1	0.4163	10	4.163	5	0.837
0.25	0.3247	4	1.299	2	0.701
0.5	0.2216	2	0.4432	1	0.5568
1.0	0.1097	1	0.1097	0.5000	0.3903
1.5	.0567	2/3	0.0378	.3333	0.2955
2.0	.0301	1/2	0.0150	.2500	0.2350
2.5	.0163	2/5	0.0065	0.2000	0.1935
3.0	.0089	1/3	0.0030	.1667	0.1637
4.0	.0028	1/4	0.0007	.1250	0.1243
5.0	0.0009	1/5	0.0002	0.1000	0.0998
6.0	0.0003	1/6	0.00005	0.08333	0.08328

If experimental results are in terms of the spectrophotometric quantity, absorbance A , defined as $\log_{10} (1/T)$ where T is the fraction of light transmitted through the given distance, the light absorption coefficient a is found through the relationship

FIGURE 3
FRACTION TRANSMITTED AS A FUNCTION OF ABSORBANCE
AND CYLINDER HEIGHT

$$a = A_1/M$$

where A_1 is the absorbance per unit distance

and

$$1/M = \log_e 10 \doteq 2.303$$

Since specular reflectors with 100% reflectivity in the visible range drop off in the ultraviolet, the expression has not been found directly applicable to simple scintillating solutions. However, it might be found useful in comparisons or when wave-shifters are employed.

III. Cylinder with Totally-absorbing Surfaces - Derivation of A General Expression

In this instance only that light headed initially towards some point on the window has a chance to reach

the window. Let the coordinates of a point P at which light is being emitted be z and ρ , where z is its vertical distance from the window and ρ is its horizontal distance from the axis of the cylinder. Let the coordinates of a window element be r and θ , (and $z = 0$), where r is its distance from the cylinder axis and θ is the angle between r and the plane of ρ and z . See Fig. 4. The area of the window element is then $r d\theta dr$.

The element of solid angle subtended by the window element is

$$d\Omega = \frac{r d\theta dr}{b^2} \cos \phi = \frac{r d\theta dr}{b^2} \frac{z}{b} = \frac{zr d\theta dr}{b^3}$$

where b is the distance from P to the window element, and ϕ is the angle between b and z .

If E_1 is the light originating in unit volume at P, then the light from unit volume at P reaching the window in this element of solid angle is

FIGURE 4
ELEMENT OF INTEGRATION ON WINDOW

$$dE = E_1 \frac{d\Omega}{4\pi} e^{-ab} = \frac{E_1}{4\pi} e^{-ab} \frac{zr d\theta dr}{b^3} = \frac{E_1 zr dr}{4\pi} \frac{e^{-ab}}{b^3} d\theta$$

and the light passing through a complete circular window element of area $2\pi r dr$ centered on the cylinder axis and including the element $r d\theta dr$ is

$$\frac{E_1 zr dr}{4\pi} \int_0^{2\pi} \frac{e^{-ab}}{b^3} d\theta$$

The light passing through the entire window is then

$$\frac{E_1 Z}{4\pi} \int_0^R \int_0^{2\pi} \frac{r e^{-ab}}{b^3} d\theta dr$$

FIGURE 5
ELEMENT OF INTEGRATION IN SOURCE

Consider a circular ring element, as in Fig. 5, centered on the cylinder axis, with volume $2\pi\rho d\rho dz$, and including the point P.

Since all points in this ring bear exactly the same geometric relationship to the window, the total light reaching the window from the ring element is

$$2\pi\rho d\rho dz \frac{E_1 Z}{4\pi} \int_0^R \int_0^{2\pi} \frac{r e^{-ab}}{b^3} d\theta dr$$

From a complete section of the cylinder of thickness dz the light reaching the window is

$$\frac{E_1 Z dz}{2} \int_0^R \int_0^R \int_0^{2\pi} \frac{\rho r e^{-ab}}{b^3} d\theta dr d\rho$$

and the total from the entire cylinder is

$$E = \frac{E_1}{2} \int_0^H \int_0^R \int_0^R \int_0^{2\pi} \frac{z\rho r e^{-ab}}{b^3} d\theta dr d\rho dz$$

Therefore, the fraction T of emitted light transmitted to the window is

$$T = \frac{E}{\pi R^2 H E_1} = \frac{1}{2 \pi R^2 H} \int_0^H \int_0^R \int_0^R \int_0^{2\pi} \frac{z \rho r e^{-ab}}{b^3} d\theta dr d\rho dz .$$

The distance b is a function of θ , r , and ρ , and its expression is determined as follows:

$$c^2 = \rho^2 + r^2 - 2\rho r \cos \theta$$

$$b^2 = Z^2 + c^2 = Z^2 + \rho^2 + r^2 - 2\rho r \cos \theta$$

$$b = (Z^2 + \rho^2 + r^2 - 2\rho r \cos \theta)^{1/2} .$$

After this substitution, as well as that for a in terms of the absorbance per unit distance, A_1 , discussed in Part II, the expression for the fraction transmitted becomes

$$T = \frac{1}{2 \pi R^2 H} \int_0^H \int_0^R \int_0^R \int_0^{2\pi} \frac{z \rho r \exp \left(-\frac{A_1}{M} (Z^2 + \rho^2 + r^2 - 2\rho r \cos \theta)^{1/2} \right)}{(Z^2 + \rho^2 + r^2 - 2\rho r \cos \theta)^{3/2}} d\theta dr d\rho dz .$$

Since this expression does not lend itself to solution in closed form it must be numerically integrated for particular combinations of R , H , and A_1 .

IV. Cylinder with Totally-absorbing Surfaces - Numerical Solution for a Specific Scintillating Solution Container

At Argonne the IBM 704 computer was used in solving the expression for a cylinder of radius 1.430 cm and height 2.921 cm for $A_1 = 0, 0.1, 0.3, 0.6$, and 2, and from the results the working curves of Fig. 7 were prepared. Analyst and programmer were E. H. Bareiss and B. Garbow, respectively. The expression was evaluated by a trapezoidal method in which each dimension was broken into the same number, N , of equal intervals and sequential summations carried out. The value of a function at the midpoint of an interval was taken as the average value of the function in the interval. The mental picture is that of the window being divided into N^2 annulus sectors, (N sectors of radius R and angular width $2\pi/N$, each subdivided into N concentric annulus sectors of radial breadth R/N), flux through each of these being contributed by each of N^2 hollow cylindrical sections (N solid cylindrical sections of radius R and height H/N , each subdivided into N concentric hollow cylindrical sections of radial thickness R/N). The element of summation is the fraction of light reaching a given annulus sector of the window of that emitted by a given hollow cylindrical section of the source. The number of contributions to the sum is thus seen to be $N^2 \times N^2$, or N^4 , as would

be true generally with quadruple integrals evaluated in this fashion. The greater the N the greater the chance that at the midpoint of an interval the value of the function approximates its average value in the interval, but unhappily machine time increases exponentially with N . In such cases, the usual procedure is to continue to increase N until further increases do not change the results appreciably.

Evaluations were made for $N = 4, 8, 12$, and 16 , and although convergence had not resulted, it was felt that the estimated convergence values determined by extrapolation, as indicated in Fig. 6, were sufficiently accurate for present purposes. Computations for larger values of N can conveniently be carried out in the future if warranted. In Table 6 are listed the estimated convergence values of T for the five values of A_1 . These have been plotted in Fig. 7 and a working curve drawn therefrom. For large values of A_1 , the $1/A_1$ plot is useful.

FIGURE 6
ESTIMATION OF CONVERGENCE VALUES FOR COMPUTER RESULTS

Table 6

Computer convergence
values estimated

A_1 (cm^{-1})	T
0	0.145
0.1	0.109
0.3	0.070
0.6	0.042
2.0	0.012
(∞)	(0)

FIGURE 7
FRACTION TRANSMITTED IN BLACK CYLINDER OF RADIUS 1.430 CM AND HEIGHT 2.921 CM

V. Application to Studies of Radiation Damage in Liquid Scintillating Solutions

The application of a heavy dose of radiation to the solvent used in a liquid scintillating solution results in a large measurable drop of the light output. The radiation apparently produces irradiation products, which may (1) have a greater light absorbance, A , than the unchanged solvent molecules, (2) quench excited undamaged solvent molecules, and (3) quench excited scintillator molecules.

The results in Parts III and IV may be used to predict to what fraction the light output will drop using a black container, provided only (1) above is operative. This predicted fraction is the fraction calculated for the irradiated solution divided by the fraction calculated for the solution before irradiation. The amount by which the experimental result disagrees with that predicted is theoretically a measure of (2) and (3).

Since absorbance, A , is wavelength dependent, and furthermore, both the scintillator and multiplier phototube have wavelength-dependent characteristics, the fraction calculated must represent a weighted average over the wavelength range of scintillator emission. The method is illustrated in the following real case, except that for simplification the range is broken into far fewer intervals than needed, and no accuracy is intended:

Source of information	Wavelength range ($m\mu$)	320 -380	380 -440	440 -500	Total
<u>Irradiated sample</u>					
Spectrophotometer	Absolute absorbance of irradiated solvent per cm (A_1)	2.30	0.03	0	
Spectrophotometer	Relative spectra distribution of photons emitted by scintillator	59	38	3	
Graph T vs A_1	x Fraction of light reaching photomultiplier (1)	0.011	0.130	0.145	
	= Relative No. of photons reaching photomultiplier	0.65	1.94	0.41	
Calibration	x Relative photon response of photomultiplier	21	40	39	
	= Expected relative output	14	198	17	229
<u>Unirradiated reference</u>					
(as per calculation analogous to that above, based on the absorbance of the unirradiated solvent)					
					348
<u>Predicted output fraction</u>		$229/348 = 0.66 = 66\%$			

VI. Experimental Results

So far, results have been obtained for only two samples, and these results have large uncertainties. Both consisted of 2 g diphenyloxazole (PPO) per liter of doubly-distilled p-xylene maintained in an oxygen-free nitrogen atmosphere, irradiated approximately 16 hours with gamma radiation of approximately 10^6 r per hour, the scintillator being added after the irradiation. The PPO emission spectrum at very low concentrations was furnished by W. L. Buck of Argonne National Laboratory and was assumed to be the true emission spectrum of PPO. Experimentally, however, a concentration of 2 g/l of PPO was used. At this concentration there is appreciable absorption at low wavelengths and re-emission at longer wavelengths by the PPO, completely modifying the original emission spectrum. Therefore, in presenting the results obtained by using the true emission of PPO and not correcting for a wavelength shift due to absorption and re-emission, it is felt that the results give too low a value. To circumvent the effect of absorption and re-emission, the emission spectrum of a thin sample, 0.5 mm, of a 2 g/l PPO-xylene solution was obtained and used. As long as 0.5 mm is short compared to the mean free light path in the cell and the absorption in the wavelength region by the PPO is much greater than the absorption of the radiation products, then the emission spectrum obtained from a 0.5 mm sample is a good approximation to an average PPO spectrum. Computations are presented with both types of spectra. Each spectrum, whose range is roughly 320 to 500 m μ , was divided into 10-m μ intervals. The multiplier phototube used was calibrated by the Material Laboratory, New York Naval Shipyard. Solvent absorbances were read on a Beckman DK-2 Spectrophotometer, and output readings in the black cylinder were obtained from an electrometer circuit designed by R. K. Swank of Argonne National Laboratory.

The results obtained, together with estimated uncertainties, are as follows:

	<u>Ratio irradiated to unirradiated</u>		
	<u>Predicted I</u>	<u>Predicted II</u>	<u>Experimental</u>
Sample 1 irradiated 16-1/5 hrs	0.66 ± 0.04	0.71 ± 0.04	0.62 ± 0.02
Sample 2 irradiated 15-5/6 hrs	0.66 ± 0.04	0.71 ± 0.04	0.58 ± 0.02

where Predicted I is computed from the true PPO emission (neglecting absorption and re-emission) and Predicted II is computed from an estimated PPO emission. The uncertainty in the predicted results arises primarily from the fact that insufficiently precise absorbance readings were made for low absorbances; greater path lengths will be used in future. The uncertainty in the experimental result can be reduced by increasing the number of readings taken on a given sample.

The difference between predicted and experimental results should be the effect of factors (2) and (3) of paragraph 1 of Part V. From an independent experiment, the scintillation efficiency of an irradiated solution was measured with an alpha source. By irradiating xylene for 17 hours under nitrogenated conditions and then adding 2 g/l of PPO, the scintillation efficiency was reduced to a value approximately 75% of that for an unirradiated solution. Therefore, the predicted values which take into account only the absorption should be multiplied by the scintillation efficiency ratio when any comparison is made to the experimental results. Doing this, one obtains the following:

	<u>Predicted I</u>	<u>Predicted II</u>	<u>Experimental</u>
Sample 1	0.50 ± 0.04	0.54 ± 0.04	0.62 ± 0.02
Sample 2	0.50 ± 0.04	0.54 ± 0.04	0.58 ± 0.02

To obtain more accurate results one should use electrons or beta rays to measure the scintillation efficiency of an irradiated solution. This will be done shortly.

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NOTE ON THE "TWIN" SCINTILLATOR METHOD OF DETECTING NEUTRONS

I. B. Berlman

The twin scintillator method for measuring neutrons in the presence of gamma-rays has been described in previous reports and elsewhere.^(1,2) The usefulness of the twin scintillator method is dependent on having the luminescence yield of one scintillating solution to gamma-rays and protons be the same as in its twin. L. D. Marinelli has suggested a convenient method for testing the response of each scintillating solution to alpha particles and gamma-rays by means of the cell shown in Fig. 3 of an earlier report.⁽³⁾ This cell which contains a curium alpha source, used in conjunction with an external Co^{60} source, provides a means of determining the luminescence yield of each solution to alpha particles and electrons simultaneously.

Since fluorinated xylene is the less efficient solvent, one method of matching its luminescence yield to that of hydrogenated xylene is to use a much smaller concentration of PPO in the latter case. However, the α/β ratio is concentration dependent, so if the two solutions are matched for gamma-rays then they are no longer matched for alpha particles, as seen in Fig. 8. The effect of scintillator concentration on the α/β ratio, as

Fig. 8

reported by Reynolds⁽⁴⁾ suggests that a much better method of matching the two solutions to the two types of radiations is to maximize the fluorescence from the fluorinated xylene by nitrogenation and a concentration of 4 g/l of PPO. The regular xylene should contain as nearly the same PPO concentration as is feasible in order to maintain the same α/β ratio.

The excess fluorescence of the more efficient regular xylene can be reduced by the addition of a quencher. One method of quenching is prolonged oxygen bubbling of the xylene before adding the PPO; any slight inequality remaining in the fluorescence yields can be corrected electronically. See Fig. 9. It is surmised that an intermediately ionizing particle such as a proton will also produce the same response in both solutions.

Fig. 9

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INTERIM REPORT ON MEASUREMENT OF THE COSMIC-RAY NEUTRON
BACKGROUND WITH THE "TWIN" SCINTILLATION
FAST NEUTRON SPECTROMETER

R. Grismore, L. D. Marinelli and B. G. Oltman

Work has been continuing on a program of flux and spectrum measurements of the cosmic-ray-produced neutron background. Preliminary details of this program, which was initiated for the purpose of comparing the intermediate-energy natural neutron and gamma-ray dose rates being received by contemporary man, were given in an earlier report.⁽¹⁾

During the period since that report some improvements have been made in the electronic system of the "twin" scintillation fast neutron spectrometer. The anomalies in the data coming from the spectrometer which were mentioned in the earlier report⁽¹⁾ have been resolved. The pulse-height stability of the whole unit has been improved, and the gain of the coincidence system has been increased, thus decreasing the lower limit of energies which can be observed.

In addition, more background data have been taken in Room B-110, Building D-203 with the spectrometer, both with and without one-quarter-inch lead shields about the scintillators. These data tend to show that the neutron background counting rate is not quite as large compared to the gamma background counting rate as was indicated by the data presented earlier,⁽¹⁾ there now being only a small difference between the ordinary and fluorinated xylene spectra, even with the one-quarter-inch lead shields. The decrease in the difference results from a change in operating conditions made in order to match the spectra of the two scintillators better for gamma rays.

More extensive background data have not yet been obtained because it has been necessary first to better evaluate the fundamental properties of the counters themselves. One result of the attempt to do this is the above-mentioned improvement in the gamma-ray match between the two scintillating fluids. Also, pulse height versus neutron energy calibration runs of the counter cells have been made with D-D neutrons from the Cockroft-Walton linear accelerator at energies of 2.18, 2.49 and 2.99 Mev. The calibration curves drawn from these data show that pulse height is not quite linear with neutron energy in this range and indicate that the relationship must become quite non-linear before zero pulse height and energy are reached.

An even more interesting result obtained from the energy calibration runs is that the fluorinated xylene cell yields about 30-40 per cent greater pulse height for neutrons of a given energy than the ordinary xylene cell. Thus, while the two are at present matched for gamma rays, they

are definitely not matched for neutrons. Apparently the two fluids have significantly different proton-electron response ratios owing to the great difference between the concentrations of diphenyloxazole scintillator solute in the two solvents (4 g/l in the fluorinated xylene; 0.28 g/l in the ordinary xylene). Dr. I. B. Berlman of the Radiological Physics Division first called our attention to this phenomenon and has done further research on the subject. His results are contained elsewhere in this volume. Apparently the solution to the problem of matching the spectra of the two scintillators simultaneously for both neutrons and gamma rays lies in increasing the response of the fluorinated xylene cell by bubbling nitrogen gas through it and decreasing the response of the ordinary xylene cell by treatment with oxygen or other quencher until the two can be brought into match with approximately equal concentrations of solute.

Our future course of action will thus be to attempt to match the two scintillators simultaneously for gamma rays and neutrons in the above-described manner. After this has been accomplished, we plan to calibrate the spectrometer more completely from about 200 kev to 17 Mev of neutron energy, using D-T and D-D neutrons at various angles and also neutrons at two or three lower energies to be obtained with the aid of the Physics Division's Van de Graaff accelerator. Upon completion of the calibration process the spectrometer will again be applied to the problem of measuring background spectra. It is planned to assemble a bismuth cave in room B-110 in order to enhance the cosmic-ray-produced neutron background relative to the gamma-ray background for measurement purposes.

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DIFFUSION AND ABSORPTION OF GASES IN PLASTIC-WALLED IONIZATION CHAMBERS

R. J. Lari* and J. H. Marshall

Introduction

The design of tissue-equivalent⁽¹⁾ ionization chambers would be facilitated by a knowledge of the solubility and permeability of the wall materials to the enclosed gases. Direct measurement on full scale chambers is difficult because changes in gas pressure may be small except over periods of months. In order to reduce the measurement time and magnify the effects, diffusion and solubility measurements have been made on thin disks of several plastics. The results have then been applied to a typical ionization chamber design.

If at $t = 0$ a gas is admitted to one side of a plane slab of material initially free of that gas, there will be a delay before the gas reaches an equilibrium rate of diffusion through the slab (see Fig. 10). This delay, L , is directly related to the diffusion coefficient, D , and to the thickness of the slab, w :⁽²⁾

$$L = \frac{w^2}{6D} \quad (1)$$

After a time $t \cong 2L$, there will be a constant rate of flow through the slab (assuming the concentrations C_1 and C_0 are held constant) which is given by:

$$F = A D \frac{\alpha(C_1 - C_0)}{w} \quad (2)$$

where

- αD = permeability coefficient (cm^2/sec)
- D = diffusion coefficient (cm^2/sec)
- A = area of slab (cm^2)
- F = atoms penetrating slab per unit time (atoms/sec)
- C_1 = concentration on one side (atoms/ cm^3)
- C_0 = concentration on other side (atoms/ cm^3)
- w = thickness of slab (cm)
- α = solubility of gas in slab material (dimensionless)

The solubility α is defined as the following concentration ratio: At equilibrium, the number of atoms of a gas dissolved in unit volume of a material is taken to be α times the number of atoms per unit volume

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of that gas in contact with it (see Fig. 11). This definition of solubility is convenient because one can then use simple units (cm^2/sec) for both the diffusion coefficient, D , and the permeability coefficient, αD .

Fig. 10

Diffusion through a slab as a function of time illustrating the delay time, L .

Fig. 11

Steady-state concentrations in a slab showing the effect of the solubility, α .

Measurement of the time delay, L , for a slab of known thickness yields D , and measurement of the subsequent permeation rate, F , yields αD . Therefore, measurement of the penetration of gas through a slab as a function of time provides values of both the diffusion coefficient, D , and the solubility, α , from which the behavior of the gas filling in an ionization chamber of that material may be calculated.

Measurements

Figure 12 shows a diagram of the cell* used in these measurements. The upper and lower chambers of this cell were separated by a thin disk of the plastic to be tested. Each disk was 2 inches in diameter, from 0.010 to 0.020 inch thick, and was supported by a stainless steel grid. A tight seal was provided by neoprene O-rings both above and below the plastic disk. A mercury well just outside the O-rings completed the seal. The

*Modified version of design proposed by G. Failla, Radiological Research Laboratory, Columbia University, New York, N.Y.

Fig. 12

Diffusion Cell

area of the disk used in expression (2) was that included within the O-rings; replacing the supporting steel grid with a fine glass frit produced no change in permeation rate, so any reduction of the effective disk area by the grid was assumed to be negligible. With the disk sealed in place, the upper and lower chambers were evacuated by a mechanical pump for a time comparable to 3 L. The lower and upper valves were then closed and the small pressure rise due to outgassing was measured as a function of time by the thermocouple gauge. At time zero, the test gas was admitted to the upper chamber at 760 mm Hg pressure. The subsequent pressure rise in the lower chamber as a function of time was measured by the thermocouple gauge and a Brown recorder over the range 10 - 100 microns Hg (C_0 was thus always negligible compared to C_1). The time necessary for one run was between one and ten hours, depending on the gas tested. The thermocouple gauge was calibrated separately for each gas used. Measured values are given in Table 7 for several gases in a commercial plastic,

Table 7

Material	Thickness, w (cm)	Gas	°C	L (min)	Solubility, α	Diffusion coefficient, $D(\text{cm}^2/\text{sec})$	Permeability coefficient $\alpha D(\text{cm}^2/\text{sec})$
Polystyrene Equivalent	0.0271	He	27	1.6	0.000	1.3×10^{-6}	7.6×10^{-8}
Polystyrene Equivalent	0.0271	CO	27	60.0	0.12	3.4×10^{-8}	3.5×10^{-9}
Polystyrene Equivalent	0.0271	N ₂	28	60.1	0.067	3.4×10^{-8}	2.3×10^{-9}
Marlex 50	0.0254	CO ₂	28	10.1	0.18	1.8×10^{-7}	3.1×10^{-8}
Marlex 50	0.0254	He	28	4.5	0.040	4.0×10^{-7}	1.6×10^{-8}
Marlex 50	0.0254	O ₂	27	9.4	0.032	1.9×10^{-7}	6.1×10^{-9}
Marlex 50	0.0254	CH ₄	28	14.2	0.045	1.3×10^{-7}	5.6×10^{-9}
Marlex 50	0.0254	CO	27	16.2	0.023	1.1×10^{-7}	2.5×10^{-9}
Marlex 50	0.0254	CO	26	13.5	0.018	1.3×10^{-7}	2.3×10^{-9}
Marlex 50	0.0254	N ₂	28	13.1	0.014	1.4×10^{-7}	1.9×10^{-9}
Tissue Equivalent - Muscle	0.0368	CO ₂	27	142.0	0.57	2.0×10^{-8}	1.5×10^{-8}
Muscle	0.0368	He	28	3.3	0.0096	1.1×10^{-6}	1.1×10^{-8}
Muscle	0.0368	O ₂	27	88.8	0.097	4.2×10^{-8}	4.1×10^{-9}
Muscle	0.0368	CH ₄	27	86.3	0.071	4.5×10^{-8}	3.2×10^{-9}
Muscle	0.0403	CH ₄	27	102	0.063	4.4×10^{-8}	2.8×10^{-9}
Muscle	0.0368	CO	27	139	0.050	2.7×10^{-8}	1.4×10^{-9}
Muscle	0.0368	N ₂	27	141	0.040	2.7×10^{-8}	1.1×10^{-9}
Bone Equivalent	0.0266	He	28	3.8	0.0098	5.2×10^{-7}	5.1×10^{-9}
Bone Equivalent	0.0266	CO ₂	27	298	0.76	6.0×10^{-9}	5.0×10^{-9}
Bone Equivalent	0.0266	CH ₄	26	180	0.058	1.1×10^{-8}	6.4×10^{-10}

Marlex 50, and in three of the new plastics developed for tissue-equivalent ionization chambers.⁽¹⁾ These measurements should be accurate to about 25%. Within this accuracy the values for Marlex 50 are consistent with those given by the Phillips Chemical Company.

Application

Assuming that the solubility and permeability coefficients we have measured apply to thick walls of these plastics, we can calculate the equilibrium times and the pressure changes to be expected for actual ionization chambers. Notice that the time necessary to reach equilibrium permeation, $\sim 2L$, increases as the square of the wall thickness and may be days or weeks for chamber walls. This is important in evaluating data from actual chambers because at times much less than L after a chamber has been filled with gas, the rate of flow into the chamber walls - and consequently the pressure drop - may be much larger than the final permeation rate. This initial flow rate due to absorption of gas in the wall is given by

$$F = \frac{A D \alpha C_1}{2\sqrt{\pi D t}} \quad (3)$$

where the symbols have the same meanings as defined above.

Suppose that the chamber has been sitting in a vacuum for a time much longer than L so that the walls contain negligible absorbed gas. Maintaining the vacuum outside the chamber, we admit a filling gas to the inside at $t = 0$ and hold the pressure of this gas constant until time t . At time t we close off the filling connection and ask what the subsequent rate of pressure drop will be. Since the gas flow rate into the wall, F , is directly proportional to the inside concentration, C_1 , we can calculate the fractional rate of pressure drop, p , as

$$p = \frac{F}{C_1 V} \quad (4)$$

where V = gas volume of chamber (in cm^3).

For times t much less than L , we take F from expression (3) and

$$p = \left(\frac{A}{V} \right) \left(\frac{\alpha \sqrt{D}}{2\sqrt{\pi}} \right) t^{-0.5} \quad (5)$$

For times t much greater than L , we take F from equation (2) and

$$p = \left(\frac{A}{V} \right) \left(\frac{1}{w} \right) (\alpha D) \quad (5)$$

Note that if the pressure drop is large, these expressions are exact only for a short time after the chamber is closed off because C_1 has been assumed constant in the expressions for F . Also, for simplicity, we have neglected the chamber geometry and have used the plane diffusion solutions in equations (1), (2), and (3) instead of exact solutions for a cylinder or a sphere.⁽²⁾ If A is taken as an average of the inner and outer areas of the chamber wall, little error is incurred. Expressions (5) and (6) with values of α and D from Table 7 are shown in Figure 13 for several gases in the tissue equivalent chamber "W" designed by Rossi and Failla.⁽³⁾ A diagram of this chamber is shown in Fig. 14. The wall thickness is $1/4$ inch or $w = 0.64$ cm. The wall area is $A \sim 20$ cm² and the gas volume $V \sim 4$ cm³ or $A/V = 5$ cm⁻¹. Notice that both expressions (5) and (6) contain A/V so that a change in this ratio would shift both segments of the curve in Fig. 13 for each gas. A change in wall thickness, w , would shift only the final equilibrium rate of pressure drop. The intersection of the two segments occurs at $t = \frac{w^2}{4\pi D}$, which is about one half the time L defined in expression (1).

The behavior of the total pressure in a chamber with a mixed filling can be estimated by summing the partial pressure drop due to each filling gas considered separately. After a time, L , (for N_2 or O_2) the pressure increments due to the inward diffusion of atmospheric N_2 and O_2 should also be included.

Fig. 13

Calculated rate of gas pressure drop immediately after sealing a chamber with wall area-to-gas volume ratio $A/V = 5$ cm⁻¹ and wall thickness $1/4$ " ($w = 0.64$ cm) as a function of the time the chamber wall has been exposed to the filling gas. (Vacuum is assumed outside chamber.)

Fig. 14

W chamber of Rossi and Failla for which Fig. 13 has been calculated.

Conclusions

It is clear that small tissue-equivalent chambers that must have constant gas compositions for long periods of time should have large auxiliary gas reservoirs and thick walls. Furthermore, their walls should be equilibrated with the filling gases for very long periods. Simple application of heat will probably not speed equilibration because it will not only increase D but also decrease α . However, the planned measurements of α versus plastic temperature may in the near future provide a method of high pressure, high temperature equilibration. Further measurements of α and D versus temperature will also be valuable for estimating the effects of room temperature changes upon the composition of a chamber gas filling. The assumption of additivity for the flow rate of two or more gases will be checked.

The use of an O-ring seal in a small chamber designed for long-term stability is probably not advisable. The permeability of neoprene to N_2 and He is about ten times that which we have measured in tissue equivalent plastic, and the other gases probably follow suit. The O-ring's area, A , in the Rossi W chamber is a little less than 1 cm^2 , or only about $1/20$ that of the plastic wall, but its width, w , is only about $1/4$ that of the wall, so that the gas permeation through the O-ring may be twice the final rate of permeation through the plastic wall (the equilibration time for the O-ring would be only a few hours).

All of the plastics used were developed and molded into disks by Francis R. Shonka of the Physical Sciences Laboratory of St. Procopius College, Lisle, Illinois, to whom we are deeply indebted. We also wish to thank J. E. Rose for suggesting and guiding this work and Billie G. Oltman for technical assistance.

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DRIFT VELOCITY OF ELECTRONS

J. C. Bowe

Calculations of the drift velocity and the energy distribution of electrons moving under the action of a uniform electric field have been performed for helium, neon, argon and nitrogen. The results are presented here in tabulated form. These calculations take into account the thermal motion of the gas atoms.⁽¹⁾

A. Table 8: The Electron Drift Velocity

Values of the drift velocity $\bar{w}$ were computed from the formula

$$\bar{w} = \frac{16 \pi e E}{m} \int_0^{\infty} \frac{\epsilon^2 \sigma_t(\epsilon) f_0(\epsilon) d\epsilon}{6 m \epsilon \sigma_t^2(\epsilon) k T + M e^2 E^2}$$

where e and m are the electronic charge and mass respectively, E is the electric field strength, ϵ the electron energy, k the Boltzmann constant, M the mass of the gas atom or molecule, T the absolute temperature of the gas, $f_0(\epsilon)$ the distribution function and $\sigma_t(\epsilon)$ the transport collision cross section. The $\sigma_t(\epsilon)$ data used in the noble gas calculations were those computed by Barbieri⁽²⁾ from the angular scattering data of Ramsauer and Kollath.⁽³⁾ Graham and Ruhlig⁽⁴⁾ also computed $\sigma_t(\epsilon)$ over the same energy range using the phase shift data which Westin⁽⁵⁾ derived from available scattering measurements. The $\sigma_t(\epsilon)$ values which they obtained are in good agreement with Barbieri's results for neon, but are in disagreement for helium and argon. This discrepancy is difficult to understand, inasmuch as Westin used the Ramsauer-Kollath data exclusively at low energy. For computational purposes, Barbieri's $\sigma_t(\epsilon)$ values were fitted with polynomials by the method of least squares. These polynomials were also made to fit the Graham-Ruhlig $\sigma_t(\epsilon)$ for neon and helium above the excitation potentials.⁽⁶⁾ For the calculation in nitrogen, the microwave $\sigma_t(\epsilon)$ data were used.

B. Tables 9, 10, 11, 12: The Distribution of Energy

The velocity distribution function, expressed in terms of the energy variable ϵ , is given by

$$f_0(\epsilon) = A e^{-6m} \int_0^{\epsilon} \frac{\epsilon \sigma_t^2(\epsilon) d\epsilon}{6 m \epsilon \sigma_t^2(\epsilon) k T + M e^2 E^2}$$

where A is the normalizing constant and is determined by setting

$4\pi \int_0^{\infty} v^2 f_0(v) dv = 1$. This function is an approximate solution⁽⁷⁾ of the Boltzman transport equation which is formulated under the assumption that collisions between electrons and gas atoms or molecules are elastic and that electron-electron collisions do not occur. The energy distribution function is given by $4\pi(2/m^3)^{1/2} \sqrt{\epsilon} f_0(\epsilon) d\epsilon$. In these tables $[\sqrt{\epsilon} f_0(\epsilon)/A]$ is given to five decimal places as a function of energy for a range of E/P_0 values and for a gas temperature of 23°C. The polynomials for $\sigma_t(\epsilon)$ listed in Table 8 were used in the evaluation of $f_0(\epsilon)$ except for helium where $\sigma_t = 28 \text{ cm}^{-1}$ was used.

C. Table 13: The Normalizing Constant

Values of the normalizing constant A were determined from the relation

$$A^{-1} = 4\pi \sqrt{\frac{2}{m^3}} \int_0^{\infty} \sqrt{\epsilon} \left[f_0(\epsilon)/A \right] d\epsilon$$

and are given in units of (gram/electron-volt)^{3/2} $\times 10^{-42}$. All computations were performed on the Argonne IBM 704 digital computer. The integrals were evaluated within an estimated error of less than 1%.

Table 8

E/P_0 , volts/cm, mm Hg	$\sigma_t, \text{cm}^{-1} \sim \text{sec}$									
	Helium		Neon		Argon		Nitrogen			
	$\gamma_t(\epsilon)^a$		$\gamma_t - 28 \text{ cm}^{-1}$		$\gamma_t(\epsilon)^b$		$\gamma_t(\epsilon)^c$		$\gamma_t(\epsilon)^d$	
	T = 0°K	T = 296°K	T = 0°K	T = 296°K	T = 0°K	T = 296°K	T = 0°K	T = 296°K	T = 0°K	T = 296°K
0.001	-	-	-	-	-	-	-	-	0.027	0.010
0.005	-	0.082	0.055	0.024	-	-	0.099	0.093	0.046	0.031
0.01	0.086	0.054	0.077	0.047	0.116	0.109	0.121	0.116	0.057	0.044
0.02	-	-	-	0.084	0.162	0.157	-	-	0.071	0.060
0.025	0.136	0.110	0.122	0.095	-	-	-	-	-	-
0.03	-	-	-	0.108	-	-	-	-	0.081	0.072
0.04	-	-	-	0.131	0.226	0.222	-	-	0.089	0.081
0.05	0.191	0.171	0.173	0.151	-	-	-	-	0.096	0.088
0.06	-	-	-	0.169	0.273	0.270	-	-	0.102	0.092
0.08	-	-	-	0.200	0.312	0.309	-	-	-	-
0.10	0.270	0.255	0.244	0.227	0.345	0.342	0.220	0.217	-	-
0.15	-	-	-	-	0.411	-	-	-	-	-
0.2	0.381	0.370	0.344	0.333	0.466	0.464	0.258	0.256	-	-
0.3	0.466	0.458	0.422	0.412	-	-	-	-	-	-
0.4	0.538	0.531	0.487	0.479	0.618	0.616	0.303	0.301	-	-
0.5	0.601	0.594	0.544	0.537	0.675	0.673	-	-	-	-
0.6	0.658	0.651	0.597	0.590	0.726	0.725	0.334	0.332	-	-
0.7	0.709	0.704	-	-	0.770	0.769	-	-	-	-
0.8	0.758	0.752	0.689	0.683	0.813	0.812	0.358	0.356	-	-
0.9	0.803	0.799	-	-	0.850	0.849	-	-	-	-
1.0	0.848	0.844	0.770	0.765	0.889	0.888	0.378	0.376	-	-
1.1	0.893	0.889	-	-	-	-	-	-	-	-
1.2	0.936	0.932	0.816	0.839	-	-	0.397	0.395	-	-
1.3	0.980	0.977	-	-	-	-	-	-	-	-
1.4	1.024	1.021	0.912	0.906	-	-	0.412	0.411	-	-
1.5	1.069	1.066	-	0.939	1.047	1.046	-	0.417	-	-
1.6	1.111	1.113	0.974	0.970	-	-	0.427	0.425	-	-
1.7	1.161	1.159	-	-	-	-	-	-	-	-
1.8	1.208	1.121	1.037	1.030	-	-	0.438	0.436	-	-
1.9	1.258	1.257	-	-	-	-	-	-	-	-
2.0	1.306	1.304	1.089	1.086	-	-	-	0.436	-	-
2.5	1.599	1.600	-	1.215	-	-	-	-	-	-
3.0	1.996	1.998	-	1.331	-	-	-	-	-	-

a. $\gamma_t = 22.9 \text{ cm}^{-1}$, $0 \leq \epsilon \leq 1.8 \text{ ev.}$

$\gamma_t(\epsilon) = 19.915 + 2.471\epsilon - 0.433\epsilon^2$, $1.8 \leq \epsilon \leq 5.37 \text{ ev.}$

$\gamma_t(\epsilon) = 27.181 - 1.416\epsilon + 0.0333\epsilon^2 - 0.00037\epsilon^3 + 0.0000016\epsilon^4$, $5.35 \leq \epsilon \leq 85 \text{ ev.}$

b. $\gamma_t(\epsilon) = 5.428 + 0.364\epsilon - 0.00725\epsilon^2 + 0.0000363\epsilon^3$, $0 \leq \epsilon \leq 85 \text{ ev.}$

c. $\gamma_t(\epsilon) = 1.262 + 3.431\epsilon + 0.553\epsilon^2 - 0.0373\epsilon^3$, $0 \leq \epsilon \leq 12.5 \text{ ev.}$

d. $\gamma_t(\epsilon) = 6.049 + 155.61\epsilon - 165.88\epsilon^2 - 72.316\epsilon^3$, $0 \leq \epsilon \leq 0.19 \text{ ev.}$

Table 9

Energy, ev	Helium $\sqrt{e} f_{ij}(e)/A$ for $\sigma_t = 28 \text{ cm}^{-1}$ E/P _j , volts/cm/mm/Hg							
	0.005	0.01	0.05	0.1	0.2	0.5	1.0	1.5
	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
0.0005	0.02230	0.02234	-	-	-	-	-	-
0.001	0.03134	0.03153	-	-	-	-	-	-
0.002	0.04347	-	-	-	-	-	-	-
0.0025	-	0.04921	-	0.04999	-	-	-	-
0.004	0.05839	-	-	-	-	-	-	-
0.005	0.06342	0.06706	0.07049	-	-	-	-	-
0.006	0.06734	-	-	-	-	-	-	-
0.0075	-	0.07817	-	-	-	-	-	-
0.008	0.07291	-	-	-	-	-	-	-
0.01	0.07623	0.08519	0.09877	0.09968	-	-	-	-
0.012	0.07795	-	-	-	-	-	-	-
0.014	0.07849	-	-	-	-	-	-	-
0.015	0.07844	0.09148	0.11918	-	-	-	-	-
0.016	0.07814	-	-	-	-	-	-	-
0.018	0.07712	-	-	-	-	-	-	-
0.02	0.07560	0.09135	0.13487	0.13965	0.14097	-	-	-
0.025	0.07038	0.08753	0.14702	-	-	-	-	-
0.03	0.06402	0.08167	0.15629	0.16841	-	-	-	-
0.035	0.05734	0.07479	0.16308	-	-	-	-	-
0.04	0.05077	0.06754	0.16771	0.19035	0.19716	-	-	-
0.045	0.04457	0.06035	0.17043	-	-	-	-	-
0.05	0.03885	0.05346	0.17146	0.20715	0.21921	0.22289	-	-
0.055	0.03368	0.04704	0.17100	-	-	-	-	-
0.06	0.02906	0.04115	0.16925	0.21966	0.23805	-	-	-
0.065	0.02498	0.03582	0.16638	-	-	-	-	-
0.07	0.02140	0.03105	0.16256	0.22845	-	-	-	-
0.075	0.01828	0.02683	0.15794	-	-	-	-	-
0.08	0.01558	0.02310	0.15267	0.23393	0.26892	-	-	-
0.085	0.01324	0.01984	0.14688	-	-	-	-	-
0.09	0.01124	0.01699	0.14070	0.23647	-	-	-	-
0.095	0.00952	0.01453	0.13422	-	-	-	-	-
0.10	0.00805	0.01239	0.12755	0.23640	0.29236	0.31219	-	-
0.11	0.00575	0.00897	0.11398	-	-	-	-	-
0.12	0.00408	0.00646	0.10055	0.22966	0.30958	-	-	-
0.13	0.00288	0.00463	0.08769	-	-	-	-	-
0.14	0.00203	0.00330	0.07567	0.21606	0.32135	-	-	-
0.15	0.00143	0.00234	0.06468	0.20738	0.32537	-	-	-
0.16	0.00100	0.00166	0.05481	0.19779	0.32824	-	-	-
0.17	0.00070	0.00117	0.04606	-	-	-	-	-
0.18	0.00050	0.00083	0.03843	0.17676	0.33076	-	-	-
0.19	0.00034	0.00058	0.03184	-	-	-	-	-
0.20	0.00024	0.00041	0.02620	0.15459	0.32939	0.42492	0.44150	-
0.22	-	-	0.01744	0.13258	0.32460	-	-	-
0.24	-	-	0.01136	0.11166	0.31682	-	-	-
0.25	0.00004	0.00007	0.00910	0.10182	0.31196	-	-	-
0.26	-	-	0.00726	0.09248	0.30652	-	-	-
0.28	-	-	0.00456	0.07541	0.29412	-	-	-
0.3	0.00000	0.00001	0.00282	0.06060	0.28004	0.48843	-	-

Table 10A

Neon					
$\sqrt{1 + (v/c)^2}$					
L.P. volts (cm/sec) H _r					
Energy, eV	0.01	0.02	0.04	0.06	0.1
0	0.0000	0.0000	0.0000	0.0000	0.0000
0.02	0.14009	0.1410	0.14134	0.14136	0.14140
0.04	0.19074	0.1916	0.1919	0.19194	0.19197
0.06	0.22447	0.2252	0.2254	0.2254	0.2254
0.08	0.25073	0.2514	0.2515	0.2515	0.2515
0.10	0.27073	0.2714	0.2715	0.2715	0.2715
0.12	0.28612	0.2868	0.2869	0.2869	0.2869
0.14	0.29897	0.2996	0.2997	0.2997	0.2997
0.16	0.30964	0.3103	0.3104	0.3104	0.3104
0.18	0.31851	0.3192	0.3193	0.3193	0.3193
0.20	0.32594	0.3266	0.3267	0.3267	0.3267
0.22	0.33223	0.3329	0.3330	0.3330	0.3330
0.24	0.33754	0.3382	0.3383	0.3383	0.3383
0.26	0.34197	0.3426	0.3427	0.3427	0.3427
0.28	0.34564	0.3463	0.3464	0.3464	0.3464
0.30	0.34854	0.3492	0.3493	0.3493	0.3493
0.32	0.35073	0.3514	0.3515	0.3515	0.3515
0.34	0.35217	0.3528	0.3529	0.3529	0.3529
0.36	0.35384	0.3545	0.3546	0.3546	0.3546
0.38	0.35573	0.3564	0.3565	0.3565	0.3565
0.40	0.35784	0.3585	0.3586	0.3586	0.3586
0.42	0.36017	0.3608	0.3609	0.3609	0.3609
0.44	0.36273	0.3634	0.3635	0.3635	0.3635
0.46	0.36554	0.3662	0.3663	0.3663	0.3663
0.48	0.36864	0.3693	0.3694	0.3694	0.3694
0.50	0.37207	0.3727	0.3728	0.3728	0.3728
0.52	0.37584	0.3765	0.3766	0.3766	0.3766
0.54	0.37997	0.3806	0.3807	0.3807	0.3807
0.56	0.38447	0.3851	0.3852	0.3852	0.3852
0.58	0.38934	0.3900	0.3901	0.3901	0.3901
0.60	0.39457	0.3952	0.3953	0.3953	0.3953
0.62	0.40017	0.4008	0.4009	0.4009	0.4009
0.64	0.40614	0.4068	0.4069	0.4069	0.4069
0.66	0.41247	0.4131	0.4132	0.4132	0.4132
0.68	0.41917	0.4198	0.4199	0.4199	0.4199
0.70	0.42624	0.4269	0.4270	0.4270	0.4270
0.72	0.43367	0.4343	0.4344	0.4344	0.4344
0.74	0.44147	0.4421	0.4422	0.4422	0.4422
0.76	0.44964	0.4503	0.4504	0.4504	0.4504
0.78	0.45817	0.4588	0.4589	0.4589	0.4589
0.80	0.46707	0.4677	0.4678	0.4678	0.4678
0.82	0.47634	0.4770	0.4771	0.4771	0.4771
0.84	0.48607	0.4867	0.4868	0.4868	0.4868
0.86	0.49624	0.4969	0.4970	0.4970	0.4970
0.88	0.50687	0.5075	0.5076	0.5076	0.5076
0.90	0.51794	0.5186	0.5187	0.5187	0.5187
0.92	0.52947	0.5301	0.5302	0.5302	0.5302
0.94	0.54147	0.5421	0.5422	0.5422	0.5422
0.96	0.55394	0.5546	0.5547	0.5547	0.5547
0.98	0.56687	0.5675	0.5676	0.5676	0.5676
1.00	0.58024	0.5809	0.5810	0.5810	0.5810
1.02	0.59407	0.5947	0.5948	0.5948	0.5948
1.04	0.60834	0.6090	0.6091	0.6091	0.6091
1.06	0.62307	0.6237	0.6238	0.6238	0.6238
1.08	0.63824	0.6389	0.6390	0.6390	0.6390
1.10	0.65387	0.6545	0.6546	0.6546	0.6546
1.12	0.66994	0.6706	0.6707	0.6707	0.6707
1.14	0.68647	0.6871	0.6872	0.6872	0.6872
1.16	0.70347	0.7041	0.7042	0.7042	0.7042
1.18	0.72094	0.7216	0.7217	0.7217	0.7217
1.20	0.73887	0.7395	0.7396	0.7396	0.7396
1.22	0.75724	0.7579	0.7580	0.7580	0.7580
1.24	0.77607	0.7767	0.7768	0.7768	0.7768
1.26	0.79534	0.7960	0.7961	0.7961	0.7961
1.28	0.81507	0.8157	0.8158	0.8158	0.8158
1.30	0.83524	0.8359	0.8360	0.8360	0.8360
1.32	0.85587	0.8565	0.8566	0.8566	0.8566
1.34	0.87694	0.8776	0.8777	0.8777	0.8777
1.36	0.89847	0.8991	0.8992	0.8992	0.8992
1.38	0.92047	0.9211	0.9212	0.9212	0.9212
1.40	0.94294	0.9436	0.9437	0.9437	0.9437
1.42	0.96587	0.9665	0.9666	0.9666	0.9666
1.44	0.98924	0.9899	0.9900	0.9900	0.9900
1.46	1.01307	1.0137	1.0138	1.0138	1.0138
1.48	1.03734	1.0380	1.0381	1.0381	1.0381
1.50	1.06207	1.0627	1.0628	1.0628	1.0628
1.52	1.08724	1.0879	1.0880	1.0880	1.0880
1.54	1.11287	1.1135	1.1136	1.1136	1.1136
1.56	1.13894	1.1396	1.1397	1.1397	1.1397
1.58	1.16547	1.1661	1.1662	1.1662	1.1662
1.60	1.19247	1.1931	1.1932	1.1932	1.1932
1.62	1.21994	1.2206	1.2207	1.2207	1.2207
1.64	1.24787	1.2485	1.2486	1.2486	1.2486
1.66	1.27624	1.2769	1.2770	1.2770	1.2770
1.68	1.30507	1.3057	1.3058	1.3058	1.3058
1.70	1.33434	1.3350	1.3351	1.3351	1.3351
1.72	1.36407	1.3647	1.3648	1.3648	1.3648
1.74	1.39424	1.3949	1.3950	1.3950	1.3950
1.76	1.42487	1.4255	1.4256	1.4256	1.4256
1.78	1.45594	1.4566	1.4567	1.4567	1.4567
1.80	1.48747	1.4881	1.4882	1.4882	1.4882
1.82	1.51947	1.5201	1.5202	1.5202	1.5202
1.84	1.55194	1.5526	1.5527	1.5527	1.5527
1.86	1.58487	1.5855	1.5856	1.5856	1.5856
1.88	1.61824	1.6189	1.6190	1.6190	1.6190
1.90	1.65207	1.6527	1.6528	1.6528	1.6528
1.92	1.68634	1.6870	1.6871	1.6871	1.6871
1.94	1.72107	1.7217	1.7218	1.7218	1.7218
1.96	1.75624	1.7569	1.7570	1.7570	1.7570
1.98	1.79187	1.7925	1.7926	1.7926	1.7926
2.00	1.82794	1.8286	1.8287	1.8287	1.8287
2.02	1.86447	1.8651	1.8652	1.8652	1.8652
2.04	1.90147	1.9021	1.9022	1.9022	1.9022
2.06	1.93894	1.9396	1.9397	1.9397	1.9397
2.08	1.97687	1.9775	1.9776	1.9776	1.9776
2.10	2.01524	2.0159	2.0160	2.0160	2.0160
2.12	2.05407	2.0547	2.0548	2.0548	2.0548
2.14	2.09334	2.0940	2.0941	2.0941	2.0941
2.16	2.13307	2.1337	2.1338	2.1338	2.1338
2.18	2.17324	2.1739	2.1740	2.1740	2.1740
2.20	2.21387	2.2145	2.2146	2.2146	2.2146
2.22	2.25494	2.2556	2.2557	2.2557	2.2557
2.24	2.29647	2.2971	2.2972	2.2972	2.2972
2.26	2.33847	2.3391	2.3392	2.3392	2.3392
2.28	2.38094	2.3816	2.3817	2.3817	2.3817
2.30	2.42387	2.4245	2.4246	2.4246	2.4246
2.32	2.46724	2.4679	2.4680	2.4680	2.4680
2.34	2.51107	2.5117	2.5118	2.5118	2.5118
2.36	2.55534	2.5560	2.5561	2.5561	2.5561
2.38	2.60007	2.6007	2.6008	2.6008	2.6008
2.40	2.64524	2.6459	2.6460	2.6460	2.6460
2.42	2.69087	2.6915	2.6916	2.6916	2.6916
2.44	2.73694	2.7376	2.7377	2.7377	2.7377
2.46	2.78347	2.7841	2.7842	2.7842	2.7842
2.48	2.83047	2.8311	2.8312	2.8312	2.8312
2.50	2.87794	2.8786	2.8787	2.8787	2.8787
2.52	2.92587	2.9265	2.9266	2.9266	2.9266
2.54	2.97424	2.9749	2.9750	2.9750	2.9750
2.56	3.02307	3.0237	3.0238	3.0238	3.0238
2.58	3.07234	3.0730	3.0731	3.0731	3.0731
2.60	3.12207	3.1227	3.1228	3.1228	3.1228
2.62	3.17224	3.1729	3.1730	3.1730	3.1730
2.64	3.22287	3.2235	3.2236	3.2236	3.2236
2.66	3.27394	3.2746	3.2747	3.2747	3.2747
2.68	3.32547	3.3261	3.3262	3.3262	3.3262
2.70	3.37747	3.3781	3.3782	3.3782	3.3782
2.72	3.42994	3.4306	3.4307	3.4307	3.4307
2.74	3.48287	3.4835	3.4836	3.4836	3.4836
2.76	3.53624	3.5369	3.5370	3.5370	3.5370
2.78	3.59007	3.5907	3.5908	3.5908	3.5908
2.80	3.64434	3.6450	3.6451	3.6451	3.6451
2.82	3.69907	3.6997	3.6998	3.6998	3.6998
2.84	3.75424	3.7549	3.7550	3.7550	3.7550
2.86	3.80987	3.8105	3.8106	3.8106	3.8106
2.88	3.86594	3.8666	3.8667	3.8667	3.8667
2.90	3.92247	3.9231	3.9232	3.9232	3.9232
2.92	3.97947	3.9801	3.9802	3.9802	3.9802
2.94	4.03694	4.0376	4.0377	4.0377	4.0377
2.96	4.09487	4.0955	4.0956	4.0956	4.0956
2.98	4.15324	4.1539	4.1540	4.1540	4.1540
3.00	4.21207	4.2127	4.2128	4.2128	4.2128
3.02	4.27134	4.2720	4.2721	4.2721	4.2721
3.04	4.33107	4.3317	4.3318	4.3318	4.3318
3.06	4.39124	4.3919	4.3920	4.3920	4.3920
3.08	4.45187	4.4525	4.4526	4.4526	4.4526
3.10	4.51294	4.5136	4.5137	4.5137	4.5137
3.12	4.57447	4.5751	4.5752	4.5752	4.5752
3.14	4.63647	4.6371	4.6372	4.6372	4.6372
3.16	4.69894	4.6996	4.6997	4.6997	4.6997
3.18	4.76187	4.7625	4.7626	4.7626	4.7626
3.20	4.82524	4.8259	4.8260	4.8260	4.8260
3.22	4.88907	4.8897	4.8898	4.8898	4.8898
3.24	4.95334	4.9540	4.9541	4.9541	4.9541
3.26	5.01807	5.0187	5.0188	5.0188	5.0188
3.28	5.08324	5.0839	5.0840	5.0840	5.0840
3.30	5.14887	5.1495	5.1496	5.1496	5.1496
3.32	5.21494	5.2156	5.2157	5.2157	5.2157
3.34	5.28147	5.2821	5.2822	5.2822	5.2822
3.36	5.34847	5.3491	5.3492	5.3492	5.3492
3.38	5.41594	5.4166	5.4167	5.4167	5.4167
3.40	5.48387	5.4845	5.4846	5.4846	5.4846
3.42	5.55224	5.5529	5.5530	5	

Table 10B

Neon

Energy, ev	$\sqrt{\epsilon} f_0(\epsilon)/A$		
	E/P_0 , volts, cm/mm Hg		
	0.1	0.2	0.4
0	0.00000	0.00000	0.00000
0.2	0.44287	0.44612	-
0.4	0.60784	0.62619	0.63088
0.6	0.70743	0.75716	-
0.8	0.75935	0.85837	0.88526
1.0	0.77133	0.93674	-
1.2	0.74961	0.99554	1.0695
1.4	0.70080	1.0367	-
1.6	0.63208	1.0614	1.2105
1.8	0.55089	1.0708	-
2.0	0.46433	1.0661	1.3174
2.2	0.37863	1.0483	-
2.4	0.29871	1.0188	1.3944
2.6	0.22798	0.97889	-
2.8	0.16828	0.93024	1.4437
3.0	0.12010	0.87441	-
3.2	0.08283	0.81307	1.4669
3.4	0.05519	0.74792	-
3.6	0.03551	0.68058	1.4654
3.8	0.02205	0.61260	-
4.0	0.01321	0.54540	1.4411
4.2	0.00763	0.48022	-
4.4	0.00425	0.41813	1.3959
4.6	0.00228	0.35998	-
4.8	0.00118	0.30638	1.3322
5.0	0.00058	0.25775	-
5.2	-	0.21430	1.2531
5.4	-	0.17606	-
5.6	-	0.14291	1.1615
5.8	-	0.11459	-
6.0	-	0.09074	1.0609
6.2	-	0.07096	-
6.4	-	0.054783	0.95466
6.6	-	0.041752	-
6.8	-	0.03141	0.84620
7.0	-	0.02331	-
7.2	-	0.01707	0.73863
7.4	-	0.01234	-
7.6	-	0.00879	0.63472
7.8	-	0.00618	-
8.0	-	0.00428	0.53680
8.4	-	-	0.44665
8.8	-	-	0.36553
9.2	-	-	0.29413
9.6	-	-	0.23262
10.0	-	-	0.18077
10.4	-	-	0.13799
10.8	-	-	0.10343
11.2	-	-	0.07611
11.6	-	-	0.05496
12.0	-	-	0.03893
12.4	-	-	0.02705
12.8	-	-	0.01843
13.2	-	-	0.01231
13.6	-	-	0.00806
14.0	-	-	0.00517
14.4	-	-	0.00325
14.8	-	-	0.00200

Table 10C

Neon

Energy, ev	$\sqrt{\epsilon} f_0(\epsilon)/A$			
	E/P_0 , volts, cm/mm Hg			
	0.6	0.8	1.0	1.5
0	0.00000	0.00000	0.00000	0.00000
0.5	0.70587	0.70641	-	-
1.0	0.99275	0.99592	0.99738	0.99884
1.5	1.2040	1.2131	-	-
2.0	1.3703	1.3893	1.3982	1.4071
2.5	1.5022	1.5362	-	-
3.0	1.6045	1.6591	1.6850	1.7110
3.5	1.6794	1.7606	-	-
4.0	1.7285	1.8423	1.8976	1.9538
4.5	1.7526	1.9051	-	-
5.0	1.7528	1.9496	2.0482	2.1505
5.5	1.7304	1.9762	-	-
6.0	1.6869	1.9855	2.1413	2.3073
6.5	1.6243	1.9779	-	-
7.0	1.5450	1.9542	2.1791	2.4270
7.5	1.4515	1.9153	-	-
8.0	1.3470	1.8622	2.1642	2.5110
8.5	1.2343	1.7963	-	-
9.0	1.1168	1.7191	2.1000	2.5599
9.5	0.99757	1.6320	-	-
10.0	0.87941	1.5370	1.9919	2.5745
10.5	0.76495	1.4358	-	-
11.0	0.65640	1.3303	1.8471	2.5561
11.5	0.55552	1.2224	-	-
12.0	0.46358	1.1139	1.6742	2.5065
12.5	0.38136	1.0064	-	-
13.0	0.30920	0.90152	1.4829	2.4278
13.5	0.24703	0.80056	-	-
14.0	0.19443	0.70469	1.2830	2.3234
14.5	0.15073	0.61480	-	-
15.0	0.11508	0.53158	1.0840	2.1968
15.5	0.08651	0.45547	-	-
16.0	0.06402	0.38671	0.89393	2.0523
16.5	0.04663	0.32531	-	-
17.0	0.03343	0.27112	0.71939	1.8943
17.5	0.02359	0.22386	-	-
18.0	0.01637	0.18310	0.56477	1.7276
18.5	0.01118	0.14835	-	-
19.0	0.00751	0.11906	0.43245	1.5567
19.5	0.00497	0.09465	-	-
20.0	0.00323	0.07452	0.32292	1.3859
20.5	0.00207	0.05812	-	-
21.0	0.00130	0.04489	0.23513	1.2192
21.5	0.00081	0.03435	-	-
22.0	0.00049	0.02603	0.16695	1.0597
22.5	-	0.01953	-	-
23.0	-	0.01452	0.11560	0.91033
23.5	-	0.01069	-	-
24.0	-	0.00780	0.07807	0.77285
24.5	-	0.00564	-	-
25.0	-	0.00404	0.05144	0.64857
25.5	-	0.00286	-	-
26.0	-	0.00201	0.03307	0.53808
26.5	-	0.00097	-	-
27.0	-	0.00045	0.02076	0.44144
28.0	-	-	0.01273	0.35818
29.0	-	-	0.00763	0.28753
30.0	-	-	0.00447	0.22841
31.0	-	-	-	0.17961
32.0	-	-	0.001437	0.13986
33.0	-	-	-	0.10789
34	-	-	-	0.08247
35	-	-	-	0.06250
36	-	-	-	0.04697
37	-	-	-	0.03503
38	-	-	-	0.02593
39	-	-	-	0.01906
40	-	-	-	0.01393
41	-	-	-	0.01011

Table 11A

Argon			
$\sqrt{E} f_0(\epsilon)/A$			
Energy,	E/P ₀ , volts/cm/mm/Hg		
ev	0.005	0.01	0.1
0	0.00000	0.00000	0.00000
0.02	0.14126	-	-
0.04	0.19964	0.19976	-
0.06	0.24213	-	-
0.08	0.27671	0.28129	0.28283
0.10	0.30493	-	-
0.12	0.32768	0.34157	-
0.14	0.34537	-	-
0.16	0.35814	0.38888	0.39989
0.18	0.36696	-	-
0.20	0.36917	0.42569	-
0.22	0.36754	-	-
0.24	0.36193	0.45264	0.48951
0.26	0.35076	-	-
0.28	0.33621	0.46965	-
0.30	0.31812	-	-
0.32	0.29707	0.47644	0.56469
0.34	0.27371	-	-
0.36	0.24874	0.47274	-
0.38	0.22289	-	-
0.40	0.19689	0.45861	0.63035
0.42	0.17140	-	-
0.44	0.14702	0.43459	-
0.46	0.12422	-	-
0.48	0.10337	0.40174	0.68886
0.50	0.08471	-	-
0.52	0.06835	0.36174	-
0.54	0.05430	-	-
0.56	0.04248	0.31675	0.74145
0.58	0.03272	-	-
0.60	0.02482	0.26927	-
0.62	0.01854	-	-
0.64	0.01364	0.22186	0.78876
0.66	0.00989	-	-
0.68	0.00706	0.17688	-
0.70	0.00497	-	-
0.72	0.00345	0.13626	0.83112
0.74	0.00236	-	-
0.76	0.00159	0.10128	-
0.78	0.00106	-	-
0.80	0.00070	0.07255	0.86847
0.82	0.00045	-	-
0.84	0.00029	0.05003	-
0.86	0.00018	-	-
0.88	0.00012	0.03319	0.90067
0.90	0.00007	-	-
0.92	0.00004	0.02116	-
0.94	0.00003	-	-
0.96	0.00002	0.01297	0.92739
0.98	0.00001	-	-
1.00	0.00001	0.00764	-
1.02	0.00000	-	-
1.04	-	0.00432	0.94821
1.06	-	-	-
1.08	-	0.00235	-
1.12	-	0.00123	0.96267
1.16	-	0.00062	-
1.2	-	0.00039	0.97022
1.24	-	0.00014	-
1.28	-	0.00006	0.97938
1.32	-	0.00003	-
1.36	-	0.00001	0.98270
1.40	-	0.00000	-
1.44	-	-	0.94682
1.48	-	-	-
1.52	-	0.92255	-
1.60	-	0.88990	-
1.68	-	0.84912	-
1.76	-	0.80076	-
1.84	-	0.74563	-
1.92	-	0.68486	-
2.00	-	0.61986	-
2.08	-	0.55222	-
2.16	-	0.48368	-
2.24	-	0.41604	-
2.32	-	0.35101	-
2.40	-	0.29011	-
2.48	-	0.23461	-
2.56	-	0.18538	-
2.64	-	0.14296	-
2.72	-	0.10746	-
2.80	-	0.07863	-
2.88	-	0.05594	-
2.96	-	0.03864	-
3.04	-	0.02589	-
3.12	-	0.01689	-
3.20	-	0.01055	-
3.28	-	0.00641	-
3.36	-	0.00376	-
3.44	-	0.00212	-
3.52	-	0.00116	-
3.60	-	0.00061	-
3.68	-	0.00031	-
3.76	-	0.00015	-
3.84	-	0.00007	-
3.92	-	0.00003	-
4.00	-	0.00001	-

Table 11B

Argon			
$\sqrt{E} f_0(\epsilon)/A$			
Energy,	E/P ₀ , volts/cm/mm/Hg		
ev	0.2	0.4	0.5
0.0	0.00000	0.00000	0.00000
0.1	0.31622	-	-
0.2	0.44716	0.44720	0.44721
0.3	0.54752	-	-
0.4	0.63193	0.63232	0.63240
0.5	0.70596	-	-
0.6	0.77237	0.77404	0.77435
0.7	0.83271	-	-
0.8	0.88786	0.89278	0.89369
0.9	0.93829	-	-
1.0	0.98424	0.99603	0.99823
1.1	1.0257	-	-
1.2	1.0626	1.0871	1.0917
1.3	1.0946	-	-
1.4	1.1214	1.1674	1.1762
1.5	1.1425	-	-
1.6	1.1575	1.2371	1.2525
1.7	1.1659	-	-
1.8	1.1672	1.2956	1.3210
1.9	1.1609	-	-
2.0	1.1465	1.3416	1.3815
2.1	1.1239	-	-
2.2	1.0939	1.3736	1.4334
2.3	1.0535	-	-
2.4	1.0061	1.3695	1.4760
2.5	0.95169	-	-
2.6	0.88937	1.3875	1.5031
2.7	0.82197	-	-
2.8	0.75017	1.3657	1.5286
2.9	0.67548	-	-
3.0	0.59951	1.3230	1.5360
3.1	0.52396	-	-
3.2	0.45049	1.2580	1.5293
3.3	0.38064	-	-
3.4	0.31573	1.1739	1.5073
3.5	0.25684	-	-
3.6	0.20467	1.0704	1.4692
3.7	0.15960	-	-
3.8	0.12166	0.95185	1.4147
3.9	0.09055	-	-
4.0	0.06574	0.82325	1.3441
4.1	0.04650	-	-
4.2	0.03201	0.69048	1.2583
4.3	0.02143	-	-
4.4	0.01393	0.55990	1.1593
4.5	0.00879	-	-
4.6	0.00537	0.43755	1.0494
4.7	0.00318	-	-
4.8	0.00182	0.32848	0.93200
4.9	0.00101	-	-
5.0	0.00054	0.23611	0.81075
5.2	-	0.16197	0.68969
5.4	-	0.10568	0.57277
5.6	-	0.06537	0.46359
5.8	-	0.03822	0.36507
6.0	-	0.02105	0.27923
6.2	-	0.01089	0.20709
6.4	-	0.00528	0.14668
6.6	-	0.00239	0.10316
6.8	-	0.00161	0.06971
7.0	-	0.00040	0.04457
7.2	-	0.00014	0.02767
7.4	-	0.00005	0.01651
7.6	-	0.00002	0.00945
7.8	-	0.00000	0.00519
8.0	-	-	0.00273
8.2	-	-	0.00137
8.4	-	-	0.00066
8.6	-	-	0.00030
8.8	-	-	0.00013
9.0	-	-	0.00006

Table 11C

Argon

Energy, ev	$\sqrt{\epsilon} f_0(\epsilon)/A$						
	E/P ₀ , volts/cm/mm Hg						
	0.8	1.0	1.2	1.4	1.5	1.6	1.8
0	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
0.4	0.63242	0.63243	0.63244	0.63244	0.63245	0.63245	0.63245
0.8	0.89402	0.89416	0.89424	0.89429	0.89431	0.89432	0.89435
1.2	1.0934	1.0941	1.0945	1.0948	1.0949	1.0949	1.0950
1.6	1.2579	1.2604	1.2618	1.2626	1.2629	1.2632	1.2635
2.0	1.3957	1.4023	1.4059	1.4081	1.4089	1.4096	1.4105
2.4	1.5076	1.5224	1.5305	1.5355	1.5372	1.5387	1.5409
2.8	1.5902	1.6196	1.6358	1.6457	1.6492	1.6521	1.6566
3.2	1.6377	1.6905	1.7200	1.7380	1.7444	1.7498	1.7579
3.6	1.6427	1.7300	1.7795	1.8100	1.8210	1.8301	1.8440
4.0	1.5984	1.7324	1.8100	1.8585	1.8762	1.8907	1.9131
4.4	1.5009	1.6925	1.8069	1.8798	1.9065	1.9287	1.9629
4.8	1.3515	1.6071	1.7663	1.8700	1.9085	1.9406	1.9906
5.2	1.1590	1.4769	1.6857	1.8260	1.8790	1.9234	1.9933
5.6	0.93921	1.3071	1.5658	1.7464	1.8160	1.8750	1.9687
6.0	0.71356	1.1084	1.4104	1.6319	1.7192	1.7943	1.9151
6.4	0.50413	0.89590	1.2276	1.4858	1.5908	1.6824	1.8323
6.8	0.32855	0.68649	1.0287	1.3146	1.4353	1.5424	1.7214
7.2	0.19600	0.49614	0.82678	1.1273	1.2597	1.3798	1.5855
7.6	0.10628	0.33656	0.63519	0.93453	1.0732	1.2022	1.4297
8.0	0.05205	0.21335	0.46500	0.74716	0.88569	1.0183	1.2602
8.4	0.02290	0.12591	0.32348	0.57493	0.70674	0.83719	1.0845
8.8	0.00902	0.06898	0.21336	0.42508	0.54449	0.66720	0.91028
9.2	0.00317	0.03501	0.13323	0.30161	0.40458	0.51494	0.74465
9.6	0.00100	0.01645	0.07870	0.20525	0.28977	0.38469	0.59343
10.0	0.0003	0.00715	0.04397	0.13395	0.20004	0.27815	0.46070
10.4		0.00288	0.02327	0.08390	0.13319	0.19476	0.34854
10.8		0.00108	0.01168	0.05049	0.08562	0.13220	0.25719
11.2		0.00038	0.00558	0.02927	0.05325	0.08714	0.18535
11.6		0.00013	0.00255	0.01639	0.03213	0.05590	0.13071
12.0		0.00004	0.00112	0.00890	0.01887	0.03501	0.09041
12.4			0.00047	0.00471	0.01083	0.02149	0.06152

Table 12

V ₁ = 100 V									
V ₂ = 100 V									
V ₃ = 100 V									
V ₄ = 100 V									
V ₅ = 100 V									
V ₆ = 100 V									
V ₇ = 100 V									
V ₈ = 100 V									
V ₉ = 100 V									
V ₁₀ = 100 V									
V ₁₁ = 100 V									
V ₁₂ = 100 V									
V ₁₃ = 100 V									
V ₁₄ = 100 V									
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V ₂₀ = 100 V									
V ₂₁ = 100 V									
V ₂₂ = 100 V									
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V ₂₄ = 100 V									
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V ₃₀ = 100 V									
V ₃₁ = 100 V									
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V ₃₄ = 100 V									
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V ₃₇ = 100 V									
V ₃₈ = 100 V									
V ₃₉ = 100 V									
V ₄₀ = 100 V									
V ₄₁ = 100 V									
V ₄₂ = 100 V									
V ₄₃ = 100 V									
V ₄₄ = 100 V									
V ₄₅ = 100 V									
V ₄₆ = 100 V									
V ₄₇ = 100 V									
V ₄₈ = 100 V									
V ₄₉ = 100 V									
V ₅₀ = 100 V									

Table 13

Normalizing constant

$\frac{E}{P_0}$ volts/cm/mm Hg	A^* (gram/ev) $\times 10^{42}$			
	Helium**	Neon	Argon	Nitrogen
0.001	-	-	-	309.22
0.005	360.43	-	11.321	124.60
0.01	278.63	23.967	5.8455	77.077
0.02	175.02	9.2687	-	42.399
0.025	143.72	-	-	-
0.03	120.44	-	-	30.088
0.04	88.800	3.5046	-	23.404
0.05	68.779	-	-	19.139
0.06	55.232	1.9854	-	16.213
0.08	38.442	1.3312	-	
0.10	28.693	0.98096	0.86154	
0.15	-	-	-	
0.2	11.063	0.38672	0.51518	
0.3	6.1989	-	-	
0.4	4.0854	0.15874	0.31280	
0.5	2.9499	0.12019	-	
0.6	2.2571	0.096274	0.23434	
0.7	-	0.079662	-	
0.8	1.4767	0.067874	0.19102	
0.9	-	0.058821	-	
1.0	1.0618	0.051971	0.16273	
1.1	-	-	-	
1.2	0.81002	-	0.14276	
1.3	-	-	-	
1.4	0.64410	-	0.12727	
1.5	-	0.031797		
1.6	0.52799			
1.7	-			
1.8	0.44349			
1.9	-			
2.0	0.37896			
2.5	0.27168			
3.0	0.20686			

*T = 296°C

** $\sigma_t = 28 \text{ cm}^{-1}$

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POSITIVE ION EMISSION

Introduction

H. A. Schultz

The positive ion emission project had its origin in the need for a sensitive instrument to study atmospheric diffusion by use of a gaseous tracer. A modified General Electric Freon detector provided adequate sensitivity⁽¹⁾ but was sometimes unstable. The reasons for instability were sought through a study of the mechanism of operation. It has been established that a multiple-step process is involved, and it has been shown that some of the steps may be isolated for independent study.^(2,3)

I. Release of Alkali by Chlorine

H. A. Schultz and R. G. Hussey*

There appear to be several different routes by which halogens may enter the reactions taking place in the halogen-sensitive positive ion emitters which have been under study. It has been suggested that halogens (a) may increase the rate of release of ionizable material, (b) may react with this material in the gaseous phase to make ionization more probable, or (c) may react with the ionizing surface to increase the probability of ionization.⁽²⁾

The experiment described below was designed to examine the first possibility, by investigating the effect of chlorine upon the rate of release of potassium from a "boat" of the type that has been used in ionization experiments.⁽⁴⁾ Qualitative tests of this possibility, using alkali salts in a combustion furnace, gave a positive result,⁽⁵⁾ but later experimentation yielded a device ("the injector-type detector") which was halogen-sensitive even though the alkali was picked up by a separate air stream surrounded by the stream carrying the halogen.⁽²⁾ In isolating this single step, it is assumed that the gaseous tracer, such as Freon-12, has already been decomposed thermally to release Cl_2 .⁽⁵⁾

*Resident Student Associate, Summer 1958. Student Aide, Summer 1957.

Apparatus

The vaporization experiments were carried out in interchangeable one-liter Florence flasks, mounted neck down and sealed to the rest of the system by a ground glass tapered joint. The other portion of the joint carried the boat, its supports and leads, and the tubing for exhausting and filling the flask. When assembled, the boat was approximately at the center of the flask.

The platinum foil boat was of the C-shaped type used currently in other types of experiments, with the entire C heated, but with only the central third filled with potassium carbonate and controlled in temperature.⁽⁴⁾ A Pt vs Pt-Rh thermocouple was attached at one end of the active portion. A new type of current lead was used to provide improved mechanical support.

The power supply for heating the boat was thyatron-controlled to maintain constant resistance across the active portion of the boat, and therefore, constant average temperature.^(3,6)

The vaporization took place in argon (or argon plus a trace of Cl_2) at atmospheric pressure. Tank argon was processed in the same train as used for other recent experiments.⁽⁷⁾ When chlorine was to be used, it was injected directly into the center of the gas stream flowing into the flask, by means of a hypodermic syringe and a long needle inserted through a rubber stopper and through the plug of a stopcock in a short side arm. Before an injection, a short needle was inserted into the stopper, the stopcock was opened, and the side arm was flushed with argon, at about 250 ml/min for 10 minutes or more. As soon as an injection was completed, the long needle was pulled back through the stopcock and the side arm was shut off from the rest of the system until it was needed again.

The flask was exhausted through two liquid nitrogen traps by a mechanical pump. A thermocouple gauge was used to follow the progress of the pumping and to warn of serious leaks. A Helium Leak Detector* was used to get the system tight originally.

Procedure

Flasks were changed as quickly as possible to minimize the sorption of water from the room. The flask was then exhausted and filled with argon, at least once and sometimes twice, before it was filled with the gas

*Veeco Mass Spectrometer Leak Detector, Vacuum Electronics Engineering Company, 86 Denton Avenue, New Hyde Park, L. I., New York.

to be used during the experiment. The temperature of the boat was raised gradually to operating temperature (in 12 minutes) to avoid spitting due to any water vapor picked up by the potassium carbonate in the boat.⁽⁸⁾

The alkali was carried by convection currents in the flask and deposited on a circular area of the wall directly over the boat. In some cases the deposit was observed directly as a white spot which developed if the open flask was allowed to stand in room air on a day with high humidity.

The deposit was taken up in 5 ml of 0.01N nitric acid, pipetted into and out of the flask, and the potassium concentration was determined with a Beckman Model D.U. Flame Spectrometer* with calibration made and maintained by use of synthetic mixtures stored in polyethylene bottles. The flame photometry was carried out in accordance with the Beckman Handbook Number 334.⁽⁹⁾ Quantitative techniques were performed according to the methods of Pierce and Haenisch.⁽¹⁰⁾

Results

The effect of chlorine was positive each time it was used. The yield depended upon boat temperature, chlorine concentration and time. The results are summarized in Table 14 and Fig. 15. Blanks were, of course, run and additional tests to be sure that there were no appreciable gains of potassium when the flask was washed or losses when the deposit was taken up.

After each reaction involving chlorine, a succession of tests was run, using only argon, in order to remove from the boat any residual KCl which had been deposited during the reaction. The data in the table are presented in terms of percent of the ideal value calculated on the basis of one K atom per Cl atom added. Losses as high as 10% to 17% are to be expected, 10% due to thermal expansion when the boat is turned on and up to 3% to 7% due to the division of the Cl₂ between the reaction flask and the connecting glass tubing. The 3% applies to Series No. 2, 4, 6, 8, the 7% to Series 1, 3, 5, 7, depending upon whether the vacuum gauge was shut off from the tubing or not.

The greater yield, in terms of percent of the ideal, was obtained at the higher temperature, with the higher Cl₂ concentration. Note, however, that only 30% gross, 26% net came off during the reaction; the balance was vaporized afterward in six additional hours of operation, as shown in

*Beckman Scientific Instruments Division, 2500 Fullerton Road, Fullerton, California.

Table 14

Potassium released from a boat by chlorine as a function of Cl_2 concentration, temperature, and time

Chronological order	Reaction conditions			Ideal ^(b) deposit, μg	Deposits, in % of ideal				
	Cl_2 , ^(a) ml	Temp., $^{\circ}\text{C}$	Time, hr		During reaction Gross %	Net, ^(c) %	After reaction Time, hr	Net, ^(c) %	Total net, %
7	0.1	27	0.25			3	1	1	4
	0 (d)	726	1	309					
6	0.1	720	1	309	34	31	5	21	52
5	0.1	720	2	308	55	49	4	2	51
3	0.1	791	0.5	309	34	26	1.5	3	29
8	0.1	808	0.5	312	34	26	1	4	30
1	0.1	788	1	312	37	21	1	0	21
2	0.1	785	1	312	34	18	3	6	24
4	0.2	794	0.5	618	30	26	6	48	74

(a) At atmospheric pressure and temperature.

(b) Ideal deposit calculated at the rate of one K atom per Cl atom, with no allowances.

(c) Net is gross less average background of $9.3 \mu\text{g/hr}$ at 720°C (3 tests, 1, 2, and 2 hrs) or $48 \mu\text{g/hr}$ at 790°C (4 tests, $1/2$, $1/2$, 1 and 1 hrs). The energy activation of the background is 2.1 electron volts.

(d) Argon and Cl_2 were replaced with argon before heating.

Fig. 15. Potassium release after removal of Cl_2 from the flask as a function of time since the start of the Cl_2 reaction.

Figure 15 Both gross and net values are given for the reaction since the background may be suppressed during this interval through a mechanism to be discussed later.

Much lower percent yields were obtained at the higher temperature when the chlorine concentration was half as great (Series 3, 8, 1, and 2). In these cases there was very little residual yield after the chlorine was removed.

Tests at the lower temperature, with the lower chlorine concentration, gave intermediate yields. There was considerable residual material after a one-hour run (Series 6), but much less after a two-hour run (Series 5). Both gave about the same total net yield, as shown in the table and in the graph

Chlorine had very little effect when it was allowed to react only at room temperature (Series 7).

Discussion

The data are too few to give a complete description of the relation between reaction rate, temperature, and chlorine concentration. They do show that the yield, in per cent of the ideal, is greater at lower boat temperature and at higher initial chlorine concentration. Also, the highest yields were not deposited at once, residual yields were obtained after the Cl_2 had been removed. From the conditions of the experiment, and from the results obtained, it appears that the reaction depends greatly upon diffusion processes

A tentative explanation of the results will now be given. Consider first background without chlorine. Natural convection currents rise along the sides of the boat but leave a stagnant zone directly over the potassium carbonate which is down inside the boat. The carbonate and its vapor are in equilibrium at the surface, but the partial pressure of the vapor decreases with increasing distance from this surface. There is a concentration gradient, and the vapor diffuses out of this zone continually to be picked up by the convection current and carried to the top of the flask, where it is deposited.

If the flask contains only a very slight trace of Cl_2 , it will react with the alkali vapor above the boat, converting it to KCl , but it will increase the rate of deposit of potassium only by penetrating into the stagnant zone to change the concentration gradient of the carbonate vapor and therefore, its rate of diffusion. At very low Cl concentrations in the flask most of the Cl_2 enters into reaction near the boundaries of this zone, but the Cl_2 does penetrate the zone to some extent.

At higher Cl_2 concentrations and lower boat temperatures, an appreciable amount of Cl_2 will reach the surface of the carbonate in the boat. Potassium chloride will be deposited here only if it is being formed at a rate greater than that required to maintain the vapor pressure of the KCl at the surface. All told, there are three diffusion processes, involving Cl_2 , K_2CO_3 , and KCl .

Consider next the problem of subtracting off the background. From a practical standpoint, the entire background should, of course, be subtracted since one is interested in the net change in ion current due to a halogen compound, such as Freon-12. However, from the somewhat more fundamental standpoint assumed at present, one is interested in the yield of KCl ; then one should subtract off only the K which is deposited in some form other than KCl . When chlorine is present throughout the flask, as in at least the early part of the reaction phase of the above experiments, practically all the alkali deposited may be in the form of KCl . The true yield for the reaction phase will, therefore, lie between the gross and the net yields.

When there is no free Cl_2 in the flask, as in the case of the runs "after reaction" the situation is different. If any residual KCl remains in the boat, it probably exists as one component of a mixture, and each component vaporizes independently of the other; therefore, the entire background should be subtracted. In extreme cases, the original carbonate might be completely covered with KCl for a time, but this does not seem very likely in view of interdiffusion. (The melting point of KCl is 776°C , between the two temperatures used.)

These questions regarding background might be answered more satisfactorily by analyzing the deposit for Cl as well as for K .

A quantitative description of the proposed mechanism requires the solution of second order nonlinear differential equations. It appears that the simplified, one-dimensional version may be solved readily by a process of iteration,⁽¹¹⁾ but the integrations have not been carried out.

The results of this experiment suggest the following criteria to be used in practical design. The ionizable material should be in a form which has a low vapor pressure relative to that of the chloride at the same temperature. This material should combine readily with Cl_2 to produce the chloride. Forced convection should be used rather than natural convection, and the source of ionizable material should be so arranged that there is good contact between it and the gas stream containing the chlorine. In the Freon detector, the Cl_2 results from the thermal decomposition of Freon.⁽⁵⁾

Mr. William Prepejchal improved the boat design, constructed the boat and the boat power supply, and also assisted in the experimental work.

II. Effect of Various Gases upon Positive Ion Emission: Hydrogen, Oxygen, Propane, and Methane

H. A. Schultz

The previous semiannual report⁽⁷⁾ pointed out that it was important to study the effects of various gases which might be found in the atmosphere if the principle under study is to be used in atmospheric diffusion studies. Also, it gave some results of adding oxygen to purified argon and pointed out that hydrogen was one of the other important gases to study, due to its stabilizing action in the modified General Electric Freon Detector.⁽¹⁾

The study of additive gases has been continued, and attention has been focused primarily upon hydrogen, oxygen, and two hydrocarbons, propane and methane, added to argon. The effect of propane depends upon the history of the equipment, particularly upon the recent use of oxygen or hydrogen. Methane has much less effect than propane.

A trace of propane sometimes produces two very obvious effects, an increase in the power required to maintain the platinum ionizing filament at a constant resistance value, and a change in the appearance of the filament, as observed through the optical pyrometer, or directly when the filament is cold. These effects are inhibited by the prior use of hydrogen mixed with the argon. Also, these effects may be at least partially reversed by the use of oxygen or of hydrogen in the argon following the use of the propane.

These results are most likely related to those obtained earlier with butane.⁽¹²⁾ Possibly any hydrocarbon heavier than methane has a similar effect. The results may also be related to the stabilizing action of hydrogen in the modified commercial Freon detector and to small and rare, but unexplained, changes observed in power requirements. Thus, there are a number of reasons for examining the propane-hydrogen-oxygen system carefully. The work is still in progress.

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RADON RETENTION OF EMBEDDED AND SECTIONED BONE

R. E. Rowland and J. H. Marshall

Through the cooperation of Dr. Charles W. Mays and Dr. Webster S. S. Jee of the Radiobiology Laboratory at the University of Utah, bones from beagle dogs which have carried pure Ra^{226} for periods ranging from 58 to 1606 days have been made available to us. The radon retention of some of these dogs had been measured in vivo,⁽¹⁾ and, in some cases, an in vitro measurement of the retention was also available.

The radon retention of these dogs is known to increase with the time the burden has been carried.⁽¹⁾ The question to be resolved in this study was: Does the radon retention of bone embedded in methyl methacrylate and sectioned for autoradiography differ from the in vivo retention?

The measurement of the radon retention of the samples is based upon the detection of the gamma-rays emitted by the daughter products of radon when the sample is sealed in glass. The fractional radon retention within each sample is calculated directly from the observed increase in the number of these gamma-rays from the time of sealing until radioactive equilibrium is reached, about a month later. Care must be taken to allow the bone samples to come into radioactive equilibrium before they are sealed and measurements made. For example, if embedding and sectioning were to increase the radon retention, this increase would not be measurable by the gamma-ray technique until the new radon retention level in the sample was achieved, requiring a time long compared with the half-life of radon. On the other hand, if sectioning the embedded samples were suddenly to lower the radon retention of the sample, the new radioactive equilibrium of the daughter products of radon would be achieved within four hours. All of the measurements discussed below were made on samples that had been first allowed to age in the laboratory a sufficient time to reach their new radioactive equilibrium condition.

From the bone of one dog, T12R5, three measurements were made. First, a length of cortical bone, cleaned in ethyl alcohol, then air dried, was sealed in a test tube without any embedding to determine the in vitro retention of this one-inch length of intact femur. Second, a similar length was completely embedded in methyl methacrylate, then sealed in a test tube without any cuts having been made through the embedding medium. Third, a similar length of cortical bone, embedded in methyl methacrylate, was cut into 58 sections, each 100 μ thick, and the radon retention of all of these sections was measured at one time.

This dog, which had carried its Ra^{226} for 225 days, had a measured in vivo retention of 17.4%. Measurements by C. Mays⁽²⁾ show a retention of 18.4% for these bones. Our measurements gave (1) a retention of 20.1% for the cleaned and dried length of cortical bone, (2) a retention of 96% for the embedded length of cortical bone, and (3) a retention of 39% for the 58 sections.

A second study was made on smaller quantities of bone from 10 dogs which had carried Ra^{226} for various periods. These bone samples were embedded in methyl methacrylate and sectioned to a thickness of 150 microns. Since in some cases an insufficient number of sections were available for accurate analysis, the sections from two dogs were combined and counted together. The basis for combination was the fact that the period over which the radium was carried was nearly equal for the two dogs. This combination is indicated by the use of a bracket in Table 15.

Table 15

Radon retention of 150- μ thick bone sections from Utah dogs

Dog	Injection: $\mu\text{c Ra}^{226}/\text{kg}$	Weight at injection, kg	Age at injection, months	Days to sacrifice	<u>In vivo</u> radon retention, %	Measured retention of embedded bone sections, %
T8R5	1.92	5.52	10	58	13.7	34.0
T9R5	1.94	10.4	75	58	13.8	
T13R5	9.76	7.59	13	188	16.2	39.2
T12R5	9.72	12.3	13	225	17.4	30.3
M5R5	10.1	10.9	15	1220	}	31.9
M7R5	11.9	8.85	15	1288		
T2R5	4.39	8.40	30	1368	25.0	40.6
M2R5	10.8	8.85	15	1380	22.6	
M4R4	3.47	8.83	14	1518	}	36.0
M1R4	3.51	9.08	16	1606		

The measured values of the radon retention, shown in Table 15, indicate that the retention varied from a low of 30.3% to a high of 40.6%, but that the retention in embedded sections is obviously independent of the in vivo retention. These values of retention are in agreement with the values we have previously published⁽³⁾ for retention in human bone sections treated in similar manner.

The influence of section thickness upon radon retention was not explored in this study. However, in the range of 100 to 150 microns the thickness is shown to be of secondary importance. The initial study described above, on dog T12R5, resulted in a retention of 39% for 58 sections of 100 microns thickness. A sample of this dog bone was included in the second study, and from Table 15 we see a retention of 30.3% for a group of sections 150 microns thick. Since increasing section thickness should be expected to increase the retention of radon rather than decrease it, the conclusion may be drawn that another variable is of greater importance than section thickness in the retention of radon in these samples.

It is apparent that embedded and sectioned bone retains a greater fraction of the radon produced within it than was retained in vivo or even retained within cleaned and dried in vitro samples. Part of this retention must be due to the filling of the microscopic cavities of the bone with embedding medium (for a bone sample completely surrounded with the embedding medium was shown to hold almost all of the radon produced). The heating of the bone during the embedding procedure may well be responsible for some of the retention, for although the oven temperature never exceeds 38°C, the exothermic reaction during polymerization may raise the bone temperature considerably higher than this. Such temperature increases have been shown previously⁽³⁾ to result in increased radon retention.

The significant conclusion of this work is that under the conditions of embedding and sectioning of bone samples in this laboratory, the radon retention of bone containing Ra^{226} is $35 \pm 5\%$. This value is independent of in vivo radon retention of the animal studied, and is not strongly dependent upon the thickness of the section prepared for autoradiography.

The bone sections for this study were prepared by Charles Williams. The gamma-ray measurements were made by John B. Corcoran, employing the Radiological Physics Division's Steel Room, 8 inch by 4 inch NaI(Tl) crystal, and 256-channel analyzer.

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MICRORADIOGRAPHY AND MICRODOSIMETRY OF HUMAN BONE BURDENED WITH Ra^{226} *

R. E. Rowland, J. H. Marshall, and J. Jowsey

Ra^{226} , incorporated in the bones of the dial painters and others who acquired radium therapeutically, eventually produces changes in the mineral structure of bone. These changes, microscopic in size, can be shown by means of microradiography. The most prevalent abnormality is the plugging of the haversian canals; so many canals are plugged that severe restriction of blood flow to many volumes of bone must result.

The activity of Ra^{226} deposits in bone has been measured, and the terminal dose distributions calculated for 12 patients studied in detail. The total lifetime doses to microscopic volumes in bone have been estimated from the terminal dose distribution and the total skeletal elimination curve.

Measured terminal radium concentrations in the bones of individuals who received constant weekly injections of Ra^{226} for periods ranging from 18 to 45 weeks (30 years ago) have given considerable insight to the mechanisms which remove radium from bone. Since the magnitude of the original radium concentrations can be predicted in these cases, the difference between the expected original distribution and that actually observed terminally gives an indication of the magnitude and mechanisms of removal. In cortical bone, for example, it appears that long-term exchange may remove considerably more activity than direct resorption over a thirty-year period.

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EXCRETION AND RETENTION OF Ra^{226} IN HUMANS:
A 2312-DAY FOLLOW-UP OF A SINGLE
INHALATION CASE

L. D. Marinelli, H. F. Lucas, P. F. Gustafson* and C. E. Miller

This summary brings to a close a 6.5-year period of observations on technician G who accidentally inhaled some RaSO_4 on June 13, 1952. Earlier data on body contents and excreta have been reported in the open literature⁽¹⁾ and later in some of the reports of our Division,^(2,3) as well as elsewhere in this report. They are shown in Tables 16 and 17 in their

Table 16

Retention of Ra^{226} following
single inhalation of RaSO_4

Days	Curies $\times 10^8$		
	Gamma	Radon	Total
3	24.8	9.0	33.8
13	22.2	3.0	25.2
35	23.2	5.0	28.2
102	6.7	3.0	9.7
235	2.8	2.2	5.0
622	0.7	1.6	2.3
1012	0.58	1.12	1.7
2312	0.22	0.67	0.89

Table 17

Excretion of Ra^{226} following
single inhalation of RaSO_4

Days	Curies	Days	Curies
3	2.06×10^{-8}	235	2.18×10^{-10}
7	5.6×10^{-9}	246	2.11×10^{-10}
13	4.4×10^{-9}	297	1.68×10^{-10}
34	3.2×10^{-9}	334	9.1×10^{-11}
57	1.7×10^{-9}	484	5.6×10^{-11}
61	1.6×10^{-9}	597	2.2×10^{-11a}
92	8.25×10^{-10}	622	1.8×10^{-11a}
102	7.8×10^{-10}	1012	1.1×10^{-11a}
122	5.1×10^{-10}	2300	5×10^{-12a}
185	3.56×10^{-10}		

^aCorrected for 2×10^{-12} normal intake.

(These curves are plotted on log log paper in Fig. 16.)

*Biological and Medical Research Division

entirety The body contents, when plotted on log log paper, yield a good straight line between 100-2312 days, which is described analytically by

$$q \text{ (curies retained)} = 3.6 \times 10^{-6} t^{-.78} \quad (1)$$

where t is in days.

The differential of this curve, namely the excretion curve

$$dq/dt \text{ (curies excreted)} = 2.8 \times 10^{-6} t^{-1.78} \quad (2)$$

fits as well as any the measurements obtained from excreta in the same interval (Fig. 16). For either curve no fitting was attempted at less than 10^2 days for it was exhaustively shown by scanning⁽¹⁾ *in vivo* that, at early times, most of the radioelement was in the lung and was being released, therefore, with a half-life of the order of 30 days. This fact seems to have been overlooked in at least one paper dealing with the behavior of radium excretion from humans.⁽⁴⁾ The consistency of both the gamma-ray and breath analysis on the one hand, and the excreta measurements on the other, do confirm the validity of our body measurements in absolute terms.

Fig. 16

The above-mentioned equations are also consistent with the findings of Norris et al.⁽⁵⁾ in patients observed more than 20 years after I.V. injection of radium; thus equations (1) and (2) predict daily coefficients of elimination (computed as $\frac{dq/dt}{q} = bt^{-1}$) of 9.6×10^{-5} and 9.05×10^{-5} at 8030 and 8620 days, respectively, in good agreement with Norris' values of 7.45×10^{-5} and 8.05×10^{-5} for equivalent periods.⁽⁵⁾ It would seem from these data that the power law correctly applies to radium metabolism in adult humans for long periods of time.

As for the description of excretion in terms of a sum of exponential terms, the data of Table 17 can be described adequately by the equation

$$\begin{aligned} dq/dt = & 1.53 \times 10^{-8} e^{-3.01 \times 10^{-2}t} + 2.87 \times 10^{-11} e^{-5.13 \times 10^{-3}t} \\ & + 1.43 \times 10^{-12} e^{-5.06 \times 10^{-4}t} \end{aligned} \quad (3)$$

but, in addition to its clumsiness, this expression suffers also of prophetic blindness, inasmuch as it predicts at late times a coefficient of elimination of 5×10^{-4} .^{*} This is close to what is experimentally found at 2312 days (Tables 16 and 17) but at variance with Norris' data at later times. The latter, it will be remembered, must be substantially correct for they were obtained from a total of 10 cases, and they were supported by independent remeasurements of the Elgin cases 760 days afterwards. In the second measurement the average contents of a large number of patients decreased by only 6.5%, consistent with a coefficient of elimination of 7.5×10^{-5} but not with its value of 5×10^{-4} . It will be very difficult to follow the excretion of this subject any further since corrections for normal radium intake in food and water have become necessary, and this would require hospitalization with metabolic ward facilities. Therefore, to settle any doubts^(6,7,8) beyond dispute it is proposed to remeasure the whole-body burdens of the Elgin patients very soon in order to obtain an average coefficient of elimination utilizing observations covering a span of almost a decade. This seems to be much the most economic, direct, accurate and less punitive way of obtaining meaningful values of retention in long-lived mammals.

^{*}By plotting equation (2) on semilog paper it is possible to match the straight line with the following exponential expression:

$$\begin{aligned} dq/dt = & 1.75 \times 10^{-10} e^{-3.3 \times 10^{-2}t} + 1.42 \times 10^{-11} e^{-7.7 \times 10^{-4}t} \\ & + 6.12 \times 10^{-13} e^{-8.75 \times 10^{-5}t} \end{aligned}$$

This equation will predict the correct excretion coefficient at later time ($\lambda = 8.75 \times 10^{-5}$); the trouble is that its form cannot be predicted from observations obtained at $t = 2 \times 10^3$ days if these are described as a series of exponential terms as in equation (3).

On the other hand, observations on excreta, beside their relatively higher cost and their inherent higher susceptibility to error, suffer also from considerable scatter due to their sensitivity to transient metabolic variables such as calcium content in diet, constipation, thyroid activity etc.⁽⁹⁾ The experience of Van Dilla et al.⁽¹⁰⁾ with measurements in a large number of beagles amply documents these facts; their long-term observations as well as M. P. Finkel's⁽¹¹⁾ with Sr^{90} in mice confirm the validity of the power law for alkaline earths in other mammals.

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MICROSCOPIC METABOLISM OF Ra^{226} IN CANINE BONE

R. E. Rowland and J. H. Marshall

The specific activity of Ra^{226} hot spots in the bone of Elgin State Hospital patients 26 - 28 years after a series of weekly intravenous injections has been found to be a factor of ten lower than the average specific activity of the blood during the injection period.^(1,2) In view of the narrow distribution of hot spot activities (the absence of more intense and the scarcity of less intense hot spots), we concluded that the observed hot spots were due to activity deposited in haversian bone which was forming during the injection period, but that subsequently there had been a tenfold loss of activity in unresorbed bone due to long-term exchange.⁽³⁾ This conclusion was based upon the assumption that the specific activity of newly-formed bone ($\text{Ra}^{226}/\text{Ca}$) would be equal to the specific activity of the blood during its formation. The present experiment was performed to verify this assumption.

A 15-kg mongrel dog between one and two years of age was injected, I.V., once a week for a total of eight weeks with a RaCl_2 solution containing $12.6 \mu\text{c Ra}^{226}$. This solution was in 0.1 N HCl with BaCl_2 added as a carrier (1.5 mg Ba per injection) and was neutralized immediately before each injection with NaOH.

The injection level of $12.6 \mu\text{c Ra}^{226}$ was comparable in magnitude to that given to the Elgin patients ($10 \mu\text{c Ra}^{226}$ per injection). This level was chosen so that the $\text{Ra}^{226}/\text{Ca}$ ratio in the blood of the dog would be greater than that which existed in the human patients, so that any effects due to dose, dose rates, or increased radium concentrations would be enhanced in this experiment.

Blood samples were obtained at least twice each week. Somewhat more than 3 ml of whole blood were removed for each sample; from this quantity 3 ml were obtained, by means of a volumetric pipette, and transferred to a small glass tube, the end of which was then flamed to form a seal. Thus, within this tube, the radon formed from the radium in the blood was confined, and the gamma-ray emitting daughters of radon were accumulated. After each sample of blood had aged at least a month, to allow the daughter products to come into radioactive equilibrium with the radium, the samples were counted in a well-type NaI(Tl) crystal.

Implicit in these measurements are thus the following assumptions, which define the accuracy of this experiment. (1) The calcium content of the plasma is constant and has the value of 10 mg Ca/100 ml plasma, (2) the plasma content of the whole blood is constant and is one-half by volume.

It was realized that considerable difficulty would be involved in attempting to maintain the weekly radium injections equal in magnitude, but since it is actually the plasma radium levels which are employed in the calculations, no great effort was expended in this direction.

The observed Ra/Ca ratios in the blood plasma are plotted in Fig. 17 and the average over the 8-week injection period is indicated by the dashed line. Also indicated in Fig. 17 are the values calculated from the empirical equation which describes the plasma levels for the radium-injected beagles at the University of Utah AEC project,⁽⁴⁾

$$P = 2.64 \times 10^{-3} t^{-1.24} \quad \% \text{ dose/g plasma}$$

where t is time in days, for 1 day to 700 days. These calculated values were normalized to fit at the end of the 8th week; the agreement between the observed and calculated values is quite satisfactory.

Fig. 17 Observed values of the plasma specific activity, expressed as the ratio of Ra/Ca, based on whole blood measurements. Calculated ratios, based on the loss of Ra^{226} from the Utah beagles, are normalized to fit at the end of the 8th week. The average plasma specific activity is indicated by the dashed line.

Four weeks after the last injection the right humerus and radius-ulna were removed by amputation. Quantitative alpha-track autoradiographs, no-screen autoradiographs, and microradiographs were made of embedded and sectioned bone, and the diffuse and hot spot radium concentrations measured by techniques which have been described previously.⁽¹⁾

Hot Spot Concentrations

Both nonhaversian and haversian bone were analyzed quantitatively to determine the ratio of Ra to Ca incorporated in new mineral. Both types of bone were found to contain the same ratio of Ra/Ca; the magnitude of this ratio is essentially identical to the average value calculated from the blood specific activity measurements. These values are shown in Table 18.

Table 18

Ra/Ca in newly formed mineral	
Average blood plasma	1.38 $\mu\text{c Ra/g Ca}$
Nonhaversian bone	1.41 $\mu\text{c Ra/g Ca}$
Haversian bone	1.28 $\mu\text{c Ra/g Ca}$

Thus, our previous assumption that newly-formed mineral incorporates radium in the same ratio to calcium as the two elements exist in the blood is shown to be correct. It thus follows that the postulated loss of radium from the hot spots of the Elgin State Hospital patients is verified in this respect.

Trabecular Concentrations

An attempt was made to measure the specific activity of newly-formed trabecular bone. Difficulty was experienced due to the fact that the bands of new mineral formed on the surfaces of trabeculae were very narrow. The alpha-track counts are normally made within an area defined by a square with sides 80 microns in length, whereas the trabecular bands of activity were seldom this wide.

Some relatively wide distributions of alpha tracks were found, which yielded on analysis low specific-activity values. When the microradiographs were examined, no trabeculae were visible in the location where the alpha track counts had been made. Turning to the bone sections themselves, trabeculae were found in these locations; however, the thickness of the trabeculae was not the same as the full thickness of bone section.

A microradiograph of a 100-micron thick bone section, exposed to record satisfactorily the maximum mineral density present, will not record any region in which the mineral density is less than 70% of the maximum value. Thus, if a trabecula is not the full thickness of the bone section, it will appear on the x-ray as if it consisted of low density mineral. If the trabecular thickness is less than 70% of the section thickness, it will not produce any visible image on the microradiograph.

Thus, in the regions described above, when we observed fewer alpha-track counts than normal, the reduction may have been due to the fact that the bone from which the alpha particles originated was not in contact with the emulsion, but rather covered with a layer of embedding material. Since the range of the alpha particles in bone is about 30 microns and is roughly the same in methyl methacrylate, the tracks in this case come from a 30-micron layer of methyl methacrylate and bone. To analyze this situation properly, then, the thickness of embedding material lying between each trabecula and the NTA emulsion must be known. For this reason, then, no values were obtained for the ratio of Ra/Ca incorporated into newly-formed trabeculae. It should be pointed out that without the microradiographs to delineate this difficulty, we might have reported erroneous values.

Diffuse Concentration

The average diffuse specific activity was found to be $0.051 \mu\text{c Ra}^{226}/\text{g Ca}$. Expressed as the ratio of diffuse to the uniform label, the ratio is 0.75. By uniform label⁽¹⁾ we mean the body burden divided by the bone mass, i.e., the concentration that would exist if the radium were uniformly distributed throughout the bones. A body-burden measurement was not made at amputation, so the burden instead is based upon the retention given by

$$R = 0.79 t^{-0.2}$$

where t is time in days. This again is from the Utah beagles,⁽⁴⁾ and is plotted in Fig. 18.

Fig. 18

Calculated Ra^{226} body burden as a function of time for this dog which received 8 weekly injections

Augmentation Rate

From the measured diffuse level and the integral of the blood specific activity, the augmentation rate, f , can be calculated.⁽³⁾

$$f = \frac{a}{\int_0^{T_s} B dt}$$

where f = the augmentation rate (Ca/g Ca day)
 a = diffuse specific activity at time T_s ($\mu\text{c/g Ca}$)
 T_s = time of sacrifice (days after injection of isotope)
 B = blood specific activity ($\mu\text{c/g Ca}$)

and

$\int_0^{T_s} B dt$ is the integral of the blood specific activity.

The measured value of f , 7×10^{-4} g Ca/g Ca day, agrees well with previous values of the order of 5×10^{-4} g Ca/g Ca day⁽³⁾ measured with Ca⁴⁵. It is however, about a factor of ten higher than the only value of an augmentation rate we have for man, 5×10^{-5} g Ca/g Ca day.⁽⁵⁾

For three Elgin patents who received 18 to 45 weekly injections, hotspot to diffuse ratios of 107, 150, and 218 were found. Here, with this dog, we find the hotspot to diffuse ratio to be as low as 28 to 1, with only 8 injections. Since the hotspots in each case are formed at the specific activity of the blood, the difference between man and dog is found in the diffuse level. The lower augmentation rate in man, even though the blood is not cleared of radium as fast in man as in dog, still results in a lower diffuse uptake of radium. Thus, the lower hotspot to diffuse ratio found in the dog is illustrative of a significant difference between the species, the augmentation rate.

Augmentation Rate in Newly-Formed Bone

The augmentation rate, evaluated above, was based on a diffuse activity measurement over a region of bone characterized by secondary haversian systems. It would be of interest if such a rate could be correlated with the age of a given volume of bone mineral. Does newly-formed bone show a significantly higher uptake of activity, i.e., a higher augmentation rate?

An answer to this question is provided by the autoradiographs. Figure 19 shows a fully-labeled haversian system from this dog and a micro-radiograph of the same region. This type of system was analyzed for the Ra/Ca ratio in haversian system hotspots. Figure 20, on the other hand,

Fig. 19. (a) A microradiograph and (b) an autoradiograph of a newly-formed haversian system completely labelled with Ra^{226} . The autoradiograph covers the outlined area of the microradiograph.

Fig. 20. (a) A microradiograph and (b) an autoradiograph of a haversian system, part of which is labelled with Ra^{226} . The autoradiograph covers the outlined area of the microradiograph.

shows a partially-labeled system. This particular haversian canal had started to fill before the first injection; the postinjection mineral is clearly differentiated from the preinjection mineral by the radium deposition. The number of alpha tracks over the preinjection mineral in this haversian system which is known to be very young, and shown by the microradiograph to be of very low mineral density, is considerably less than the number of α tracks over the postinjection mineral. Since the hotspot to diffuse ratio is only 28 to 1, and since the number of tracks over the preinjection mineral is very similar to that over the adjacent, older haversian systems, it is apparent that the augmentation rate of newly-formed mineral cannot be greater than two or three times the rate in older mineral.

Growth Rate Measurements

The stripping film autoradiographs of the radium-labeled regions of bone, (a typical low-power photomicrograph is shown in Fig. 21), provide

Fig. 21. A low-power microphotograph of a stripping film autoradiograph showing broad bands of bone which are labelled with Ra^{226} . From such bands both growth rate measurements and Ra/Ca ratios were determined.

a direct measurement of the rate at which mineral is laid down appositionally. Since the blood specific activity may be considered to have been held at a constant specific activity for 56 days in this animal, we can obtain a

Table 19

Linear Growth Rates

Subperiosteal bone	0.6 μ /day
Endosteal bone	0.9 μ /day
	1.3 μ /day
	1.5 μ /day
	1.8 μ /day
Haversian systems	1.3 μ /day
	1.4 μ /day
	1.6 μ /day
	1.9 μ /day

linear growth rate from the measurement of the width of the bands laid down in this period. Some values obtained for the largest haversian systems, endosteal bands, and subperiosteal bands are listed in Table 19. These values could be low if the growth in the regions measured took place during only a part of the eight-week interval.

We are deeply indebted to Dr. Robert Flynn for performing the amputation on this dog, and to Durwood Banister, who did all of the injecting and blood sampling on this animal for us.

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MICROSCOPIC METABOLISM OF CALCIUM IN BONE*

J. H. Marshall and R. E. Rowland

Abstract

Recent experiments with radioactive tracers in dogs have revealed an important component of calcium metabolism in bone. It has been found that calcium, as well as the calcium-like isotopes Ca^{45} and Ra^{226} , diffuse into and out of bone mineral at an appreciable rate even in regions of mature bone which have been thought to be metabolically inert. Called, more accurately, long-term exchange, this component may account for as much calcium transfer between blood and bone in an adult animal as the well-known components, accretion and resorption. However, because practically all bone crystallites are involved in this exchange, the time constant is long, so that a radioactive isotope deposited in bone is not returned to the blood by exchange to an appreciable extent until several years after injection. The mechanism responsible for this component is probably exchange at crystallite surfaces, possibly coupled with intracrystalline diffusion. The existence of long-term exchange will modify our interpretation of accretion rate measurements based on radioactive uptake and will increase our estimates of the availability of skeletal calcium to the circulating fluids. These studies were carried out using new techniques of quantitative autoradiography, microradiography, and computer analysis of blood and bone specific activities.

Introduction

The various processes involved in the mineral metabolism of the skeleton have been recently reviewed by Neuman and Neuman.⁽¹⁾ They have shown that new crystal formation, recrystallization, crystal surface exchange, intracrystalline exchange, crystal growth, resorption, and the aging of each element of bone structure probably all contribute to the complexity of interrelated events occurring in the skeleton. The problem before us is to isolate some of these processes for quantitative study in living animals.

*Some of this work, including experimental details, has been reported in Radiation Research, February 1959. This is the text of a paper delivered by J. H. Marshall to the conference on Bone as a Tissue, Lankenau Hospital, Philadelphia, October 30, 1958, the proceedings of which will be published by Blakiston and Company.

Elimination of Resorption by Choice of Location

The first problem of isolation is one of location. We should like to concentrate our attention on individual regions of bone that are as homogeneous as possible with respect to their structure, their stage of development, and to the metabolic processes taking place within them. By focusing our attention on microscopic volumes of bone we can hope to obtain results more definitive than those obtained by measurement of the average properties of large volumes of bone. Our method is to inject an animal intravenously with Ca^{45} , sacrifice it after an appropriate interval, and then analyze the bone by quantitative autoradiography and quantitative x-ray microradiography. These techniques permit us to study the calcium metabolism of volumes of bone 10 to 100 microns on a side. We are, therefore, able to avoid resorption cavities or haversian systems of unknown age and so can eliminate from our autoradiographic analysis for Ca^{45} the effects of resorption.

Terminology

From such microscopic measurements we can only indirectly identify processes at the molecular level, so that it is important to use concepts and terminology in describing our data that do not go beyond the resolution of our measurements. First, we shall define a "microscopic volume" of bone as a cube 10 to 100 microns on a side in which no resorption is taking place. We shall use the term "accretion" to describe any process involving the net transfer of calcium to a single "microscopic volume" of bone. We shall use the term "exchange" to describe any process involving equal and opposite rates of transfer of calcium atoms to and from a single "microscopic volume" of bone. These definitions of accretion and exchange can be employed unambiguously to whole bones or to the skeleton, because one then considers merely the sum of many microscopic volumes. Exchange under this definition includes the molecular processes of ion exchange, intracrystalline diffusion, and recrystallization. Note that this definition of exchange does not specify how long it may take for Ca^{45} atoms to reach equilibrium following their injection on one side of the exchange process. In our present terminology we refer to short-term or long-term exchange, depending on whether the time necessary for the attainment of radioactive equilibrium is much shorter or much longer than a week.

Use of Blood Specific Activity Measurements

Now we need a way to isolate the processes of exchange and of accretion in a given microscopic volume of bone from all the other processes taking place in different parts of the animal; we need to eliminate from our calculation the effect of deposition, resorption, or exchange of Ca^{45} elsewhere in the skeleton and the effect of Ca^{45} excretion from the

body as a whole. This can be done by using in our calculations measured values of the specific activity of the blood. If we know the ratio of the number of Ca^{45} atoms to the number of Ca^{40} atoms in the blood throughout a given experiment, then no matter how the Ca^{45} may have been introduced into the system (whether by multiple or single injections or by continuous feeding) and no matter what processes of uptake or elimination of Ca^{45} may be going on in other locations, we can calculate from the quantity of Ca^{45} which we find in our microscopic volume of bone the rate of calcium transfer which has existed between the blood and this volume of bone during the experiment.

Elimination of Short-term Exchange

Bauer, Carlsson, and Lindquist⁽²⁾ have developed a fruitful technique of analysis which derives its power from this use of observed blood specific activities. Figure 22 shows the physical model underlying this analysis. Bone mineral is divided into two fractions: stable bone and exchangeable bone. Bauer's exchangeable fraction refers to short-term exchange and consequently reaches radioactive equilibrium with blood within minutes or a few hours after injection. The Bauer, Carlsson, Lindquist analysis provides a way to separate the processes of short-term exchange from any other processes of calcium transfer. When used together with quantitative autoradiography, the

Fig. 22. Bauer, Carlsson, Lindquist model for calcium transfer between blood and bone.

analysis is particularly simplified since, by choice of location, we can avoid the influence of resorption upon a measured Ca^{45} activity. Thus we can wait weeks after a Ca^{45} injection before sacrificing an animal for analysis, and by this time the specific activity of the blood has dropped to such low values that there is a negligible amount of Ca^{45} in the short-term exchangeable fraction of this bone. The rate of Ca transfer from blood to stable bone is then obtained by dividing the observed Ca^{45} content of the bone by the time integral of the specific activity of the blood during the experiment. Bauer has shown that in growing bone this rate of calcium transfer corresponds closely to the accretion rate. As Neuman has pointed out, and as we shall show, this correspondence does not always hold, but first we shall describe a microscopic experiment on growing bone.

Experiment on Growing Bone

Figure 23 shows a Ca^{45} autoradiograph of a cross section through the midshaft of the femur of a young puppy. This puppy was injected with Ca^{45} intravenously at the age of 2 months, again at the age of 3 months, and sacrificed at the age of 4 months. The upper autoradiograph has been exposed to show the rings of Ca^{45} activity taken up during periosteal growth at 2 months and 3 months of age. The lower autoradiograph has been over-exposed to show the diffuse uptake of Ca^{45} in bone formed before the first injection. Since the animal was sacrificed a month after the second injection, we can use the quantity of Ca^{45} in either of these two rings to calculate the accretion rate according to the Bauer, Carlsson, Lindquist method. The intensity of the first or inner ring yields an accretion rate equivalent to $16 \mu/\text{day}$, the second or outer ring a rate of $10 \mu/\text{day}$. The average rate during the month between injections was thus $13 \mu/\text{day}$, and this rate is confirmed by measurement of the distance between the two rings, at least within the 20% accuracy of the measurements. Thus at this growing periosteal surface of bone, the accretion of calcium predominates over the processes of long-term exchange discussed by Neuman. However, this is not the case in the bone that had already been formed before the first injection.

Experiments on Non-growing Bone

The diffuse uptake of Ca^{45} in bone formed before injection is better demonstrated in an adult dog. Figure 24 shows at the bottom an autoradiograph of a cross section through the midshaft of the femur of a dog a few years of age which was given a single intravenous injection of Ca^{45} and sacrificed two weeks after injection. The upper part of this figure shows the corresponding x-ray microradiograph. With an adult animal such as this, we may choose a microscopic volume of bone in which no new bone formation is taking place, and yet there is still a Ca^{45} uptake by stable bone. (Remember that the short-term exchangeable fraction of this bone will contain negligible Ca^{45} two weeks after injection.) According to the Bauer, Carlsson, Lindquist analysis the accretion rate in this bone is $0.2 \text{ g Ca/g Ca/year}$. In other words, the Ca^{45} uptake indicates that the mineral density of this bone should be increasing some 20% per year. Now you will notice on the microradiograph that some of the regions of this bone contain secondary haversian systems, and from the beautiful work of Amprino we know that over long periods of time haversian systems do increase in mineral density. However, Fig. 24 shows broad areas of cortical bone that were laid down periosteally and have not been replaced by secondary haversian systems. Such bone is often referred to as fully mineralized bone. But we can see that its diffuse uptake of Ca^{45} is only slightly less than that in the regions of secondary haversian bone. Could it be that this periosteal bone is also slowly increasing in calcium density at a rate comparable to the measured rate of calcium transfer?

Fig. 23. Autoradiograph of a cross section through the mid-shaft of the femur of a puppy injected with Ca^{45} intravenously at age 2 months, again at age 3 months, and sacrificed at age 4 months. Upper: short exposure. Lower: longer exposure.

Fig. 24. Corresponding autoradiograph (lower) and microradiograph (upper) of a cross section through the midshaft of the femur of an adult dog injected with Ca^{45} and sacrificed 2 weeks after injection. Similar autoradiographs have been obtained up to a year after injection.

Fig. 25. Rate of long-term calcium transfer from blood to bone for microscopic volumes of canine periosteal bone measured by means of Ca^{45} diffuse uptake. A single measurement for a man dying with radium poisoning is tentatively included.

To answer this question we have made microscopic measurements on corresponding volumes of canine periosteal bone of various ages. Figure 25 shows the results of the autoradiographic measurements. The ordinate is the rate of long term Ca transfer from blood to bone calculated from the observed Ca^{45} content weeks after injection. According to the Bauer, Carlsson, Lindquist model, this rate of transfer should be identical to the accretion rate. The abscissa is the age of this bone, a few months less than the age of the dog. It is interesting that the rate of Ca transfer from blood to the cortical bone of 3-month-old rats measured by Tomlin, Henry and Kon⁽³⁾ using continual Ca^{45} feeding is consistent with the present data. A single measure-

ment from the Ca^{45} diffuse uptake in cortical bone of a 50-year-old man is tentatively included on this same graph. Now the most significant thing about Fig. 25 is that the rate of Ca transfer is much too large to represent purely accretion. For example, microscopic volumes of periosteal bone 6 years of age would have to contain 50% more calcium than periosteal bone 2 years of age. We have directly measured the calcium content of this periosteal bone as a function of bone age by means of quantitative microradiography. The results are shown in Fig. 26. It is clear that at more than a few months after its formation, this bone shows an almost constant microscopic calcium density. This remarkable paradox has been pointed out by Arnold, Jee, and Johnson,⁽⁴⁾ but here we have it in quantitative form.

Fig. 26. Microradiographic measurements of the calcium content of microscopic volumes of canine periosteal bone. Preliminary measurements for rat and man indicate a shift in the curve to the levels indicated.

Long-term Exchange

From the diffuse uptake of Ca^{45} we have measured a rate of calcium transfer from blood to microscopic volumes of bone in which no resorption is occurring, and yet there is no corresponding increase in their calcium content. We are forced to conclude that there exists a comparable rate of calcium transfer back to blood and that most of the observed Ca^{45} is therefore engaged in an exchange process. But if this is so, how could this Ca^{45} be retained in bone for weeks or months after Ca^{45} injection? This must be an exchange in which each exchanging calcium atom spends on the average more than a year in bone before returning to the blood. In other words, for long periods of time after injection, radioactive equilibrium is not attained. We are therefore observing a process of long-term exchange.

Under what conditions is it possible to explain this long retention of Ca^{45} in an exchange process? The currently accepted explanation is essentially that the rate of exchange decreases so rapidly with bone age that Ca^{45} is trapped on the bone side of the exchange. The rate of exchange certainly does decrease, but this is not a sufficient explanation; for bone a few years of age the decrease in rate during a single year is not large (Fig. 25) and so cannot be primarily responsible for the long Ca^{45} retention. We are left with the conclusion that the exchange itself has a long time constant, or in other words, that the exchangeable fraction - the calcium available for this exchange - is a large fraction of all the calcium in any microscopic volume of mature bone.

Explanation for Rapid Uptake

It is not obvious how such a process of exchange could take up Ca^{45} within a day after injection and then hold it for a year or more. To understand this we must consider the actual magnitudes of the blood and bone

Fig. 27. Calculated values for the Ca^{45} or Ra^{226} diffuse specific activity of canine periosteal bone 1 year old as described in text and appendix.

specific activities. Because some of the following experiments have employed Ra^{226} instead of Ca^{45} as the tracer, we shall refer to either Ca^{45} or radium as calcium-like radioisotopes. Their blood specific activity curves⁽⁵⁾ and their diffuse uptake in dogs we find to be quite similar. Figure 27 shows a typical curve for blood specific activity in a one-year-old dog following a single injection of a calcium-like radioisotope. The ordinate is the fraction of the total activity injected per

gram of calcium in the blood. * The diffuse specific activity in the cortical bone of a one-year-old dog is a little less than 10^{-3} of the dose per gram calcium, so that during the first day after injection, the specific activity of the blood exceeds that of a microscopic volume of nongrowing bone by a factor of 1000. Therefore the transfer of Ca^{45} from blood to bone can proceed rapidly despite the long-term nature of the exchange.

Calculation of Retention and Eventual Loss

Now how long could Ca^{45} be expected to be retained? We know the rate of exchange as a function of bone age (Fig. 25)** and we know the specific activity of the blood as a function of time after injection. Therefore, we can calculate the time dependence of the specific activity of a microscopic volume of bone. For simplicity we assume that bone can be represented by a single compartment model. *** (See Appendix) Clearly the longest retention would be produced if all of the calcium in the microscopic volume were taking part in the exchange; and this case is shown by curve 2 (Fig. 27) for a dog injected at one year of age. If only 20% of the calcium were involved, then the specific activity of the microscopic volume would decrease more quickly, as in curve 3 (Fig. 27). For comparison, curve 1 shows the behavior to be expected if the uptake of Ca^{45} were due primarily to accretion and not to long-term exchange, though this possibility has been ruled out by the x-ray measurements.

Now notice three things about this calculation. First with experiments of much less than a year's duration it is impossible to tell from the behavior of the radioactive tracer whether it has been deposited in bone by a process of accretion or is only temporarily deposited in a process of long-term exchange. Second, since we know that diffuse activity appears fixed in bone for many months after injection, we are forced to conclude that the exchangeable fraction for this long-term exchange must be a large fraction of the total cortical bone calcium. And third, no matter how large an exchangeable fraction we assume, this process of exchange requires that over periods of years an appreciable loss of calcium-like radioactivity should occur even from mature bone without the influence of resorption.

*A 9-kgm dog has a blood content of about 0.05 gm_{Ca} and a skeletal content of about 140 gm_{Ca}.

**To be precise, we know that for canine periosteal bone at least the middle part of the curve in Fig. 25 (1-10 years) represents primarily long-term exchange.

***More complex models are treated in Argonne National Laboratory, Radiological Physics Division Semiannual Report No. ANL-5919 (September 1958) p. 35, but they show that the essential point is the large-compartment behavior of the exchange.

This prediction has now been confirmed in 3 experiments. First, a one-year-old beagle was injected with Ca^{45} and his left radius-ulna amputated at one month as a control. At one year after injection the radius of the opposite leg showed a 10-20% loss of diffuse activity in regions not subject to resorption. Second, a group of 11 beagles was injected with radium* and autoradiographically analyzed for diffuse activity at different times after injection. The animals sacrificed four years after injection show approximately one-half the diffuse specific activity of the animals sacrificed a few months after injection. Third, bone from radium-injected human beings has been examined 25-30 years after injection and shows that haversian systems laid down at the time of injection have lost by exchange about 90% of their original radium content. (6)

Mechanism

As to the mechanism for this long-term exchange, it probably involves an exchange at crystallite surfaces, possibly coupled with intracrystalline diffusion, and perhaps accompanied by recrystallization. Detailed analysis of long-term tracer experiments may be able to distinguish between these possibilities. The physical explanation for the existence of a large calcium compartment in exchange with blood is provided by the extremely small size of bone crystallites in which a large fraction of the calcium is in or near the crystallite surfaces.

Conclusions

To sum up, we have shown that the uptake and retention of calcium-like radioisotopes by mature cortical bone in dogs cannot be explained as a process of accretion and therefore must be predominantly a process of long-term exchange. The measured rates of this exchange as a function of bone age require that over a period of years the radioactivity should begin exchanging back again to the blood, and this prediction has now been confirmed by 3 experiments. Although we have discussed this long-term exchange primarily with regard to the diffuse component in mature bone, it also produces a long-term loss of radioactivity from hot spots or bone formed during the injection period, though the time dependence may not be exactly the same.

In conclusion, the existence of long-term exchange has three effects upon our concepts of skeletal metabolism. First, as shown in Fig. 28, the Bauer, Carlsson, Lindquist analysis includes in its measurement of accretion rate the transfer rate of long-term exchange. This refinement in the meaning of their parameter A should not detract from its diagnostic value

*This experiment is being performed in cooperation with W.S.S.Jee of the Radiobiology Laboratory, College of Medicine, University of Utah, Salt Lake City. Complete results will appear later.

Fig. 28. Suggested model for calcium transfer between blood and bone.

because both mechanisms, accretion-resorption and long-term exchange, result in a turnover of skeletal calcium which is not detectable by over-all calcium balance measurements. Second, mature bone is not completely diffusion-locked and inert but is able to maintain a slow exchange of Ca with the blood. In adult skeletons, since there is much more mature bone than young bone the contribution of long-term exchange to the blood-bone interchange of Ca is

comparable to that of accretion and resorption. And finally, calcium-like radioactivity which has been deposited in the skeleton and not resorbed will be lost from even mature bone over periods of years due to long-term exchange.

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Addendum Concerning the Power Function Behavior
of Radioisotope Retention

The age dependence of the rate of long-term exchange of calcium between bone and blood shown in Fig. 25 may provide an important link in our understanding of the empirical power function description for whole-body retention. If the rate of long-term exchange were inversely proportional to bone age (which, is almost true for Fig. 25),

$$f = b t_{\text{age}}^{-1} \quad (1)$$

where

f = exchange rate in $\text{gm}_{\text{Ca}}/\text{gm}_{\text{Ca}} \text{ year}$

b = dimensionless constant

t_{age} = bone age in years

then the amount of Ca^{45} retained (assuming a single compartment) as a function of time after injection is given by

$$R = a t_{\text{age}}^{-b} \quad (2)$$

or

$$R = a (t + t_0)^{-b} \quad (3)$$

where

R = Ca^{45} retention

t = time after injection

t_0 = bone age at injection

These relations follow from the differential equation for a single compartment with time dependent rate constants:

$$\frac{dR}{dt} = -f R \quad (4)$$

Substituting (2) in (4) and performing the differentiation:

$$-b a t_{\text{age}}^{-b-1} = -f (a t_{\text{age}}^{-b})$$

and $f = b t_{\text{age}}^{-1}$ which agrees with (1). Therefore, the power function (2) is the solution of (4) (instead of the usual exponential) when the exchange rate varies inversely with time.

The value of b for the present data would be the numerical value of f in units of year^{-1} at the time of one year, or about 0.3. Of course for whole-body retention one must also take into account activity that has been resorbed and make a correction for redeposition from the blood, but it is encouraging that the large-compartment behavior of long-term exchange together with its decreasing rate yield an approximate power function behavior for the activity in individual regions of bone.

APPENDIX - Calculation for Fig. 27.

The following differential equation representing one bone compartment coupled to blood by a decreasing rate of calcium transfer was solved for the initial condition $v = 0$ at $t = 0$:

$$\frac{dv}{dt} = f \left(B - \frac{v}{\alpha} \right)$$

where

v = average specific activity of a microscopic volume of bone as a function of time

B = observed specific activity of the blood as a function of time

f = observed rate of long-term calcium transfer as a function of time (taken from Fig. 25)

α = fraction of the calcium content of the microscopic volume taking part in the exchange

t = time measured from time of injection.

Bone age at time of injection taken as one year.

Curve 1 (accretion) calculated setting $\alpha = \infty$ (or $\frac{v}{\alpha} = 0$)

Curve 2 $\alpha = 1$

Curve 3 $\alpha = 0.2$

A MATHEMATICAL THEORY OF THE RETENTION OF BONE-DEPOSITING RADIOACTIVE ELEMENTS

J. C. Reynolds

Introduction

A wide variety of experimental evidence, covering a number of elements in both humans and experimental animals, has established the following empirical formula for the in vivo retention of bone-depositing radioactive materials:

$$R(t) = (1-b)t^{-b} ; \quad t \geq 1 \quad , \quad (1)$$

where t is the time in days after receiving a dose, and $R(t)$ is the proportion of the dose retained in the body at time t . The constant b is between 0 and 1, usually from 0.3 to 0.5.

From a theoretical standpoint, such a formula is unusual. One would expect at first that for large t the amounts of the radioactive material in various parts of the body would come into equilibrium, and that $R(t)$ would then show the behavior of an exponential rather than a power function.

Attempts have been made to treat expression (1) as the sum of the amounts of material contained in a number of compartments, each of which releases material at a rate proportional to its contents and absorbs a certain proportion of the material released by other compartments. Such a model leads to a retention function which is a sum of exponential terms. Its weakness is that a large number of such terms are necessary to approximate (1) over the range for which it has been experimentally verified. Furthermore, the fact that these terms appear in just the proportions necessary to give a simple function such as (1) is not explained and becomes an extraordinary coincidence.

It is the purpose of this paper to attempt to explain the power function behavior of expression (1) on an entirely different basis, in terms of the diffusion of the radioactive atoms within individual bone crystals.

If we limit ourselves to calcium-like elements, and to initial doses by injection, there is apparently no organ in the body other than the bone which will retain a substantial proportion of radioactive material for a length of time much greater than a month. Thus it appears that if (1) holds for large t , the power function behavior must be the result of the retention of the material within the bone itself, and not just of the interaction of the bone and other compartments.

If we examine the deposition of such elements in bone we find two distinct forms of deposit. Some of the radioactive material is concentrated in small and intense regions called "hot spots"; the remainder is distributed diffusely throughout the bone. Microscopically, the hot spots may be associated with the capture of radioactive atoms by crystals of bone which are just being formed, while the diffuse activity may be associated with the surface adsorption of atoms by older crystals. In both cases, the atoms are released by diffusing out of the crystal. (We are ignoring the release of material by resorption.) In the hot spots, however, the initial distribution of the atoms within a crystal is probably uniform, while in the diffuse activity the initial distribution is taken to be concentrated near the surface.

The Diffusion Process

Consider a radioactive atom which has been bound at some point inside the bone crystal. When it is released, it will move through the crystal, undergoing frequent collisions, which we assume send the atom off in a random direction. If it does not leave the crystal, it will eventually be captured and bound again. We will describe this process by two parameters: the inverse mean free path between scatterings λ_s , and the inverse mean free path between captures λ_c . Once the atom has been bound, we assume that the probability of its release is a simple exponential, with an inverse mean lifetime of γ .

Now an individual atom may be described by its position and time coordinates. In addition it is useful to introduce a quantity s , called the path length, which is the total distance the atom has travelled in the crystal since its initial release. This artifice allows us to separate the diffusion and capture processes. We define $f(r, s)$ as the probability per unit volume that an atom will be found at r after it has travelled a distance s since its initial release. The probability per unit length of scattering is the constant λ_s , which is independent of whether the particle has been captured once or more while travelling the distance s . (Strictly speaking, this is not true, since a capture process changes the direction of motion of an atom just as a scattering process does. However, if scattering is much more frequent than capture the resulting correlation may be ignored.) Thus $f(r, s)$ will depend only upon s , λ_s , and the geometry of the problem.

On the other hand we may define $p(s, t)$ as the probability per unit length that an atom which has been bound (once or more times) for a total time t since its initial release will have travelled a path length s . The probability per unit length of capture is the constant λ_c , which is independent of the scattering history of the particle. Thus $p(s, t)$ is independent of the geometry.

Finally we may combine $f(r, s)$ and $p(s, t)$ to obtain the probability per unit length that a particle of age t and any path length will be found at r :

$$\int_0^{\infty} ds f(r, s) p(s, t) \quad (2)$$

and the probability that such a particle will be found anywhere within the crystal, i. e., will not have been released:

$$\int (dr) \int_0^{\infty} ds f(r, s) p(s, t) \quad (3)$$

where the integral (dr) is over the volume of the crystal.

Now we wish to find the function $p(s, t)$. Consider $p(s, t + \Delta t)$. It consists of two terms: the first is due to particles which remain bound during the time interval Δt and thus do not change their path length s . The second term is due to particles which had a path length s' at time t but which are released during the interval Δt and move a distance $s - s'$ before being recaptured. Note that we assume the time spent in moving through the crystal is negligible compared to the time spent while bound. We obtain the equation:

$$p(s, t + \Delta t) = (1 - \gamma \Delta t) p(s, t) + \int_0^s ds' p(s', t) \gamma \Delta t \lambda_c e^{-\lambda_c(s-s')} \quad (4)$$

where $(1 - \gamma \Delta t)$ is the probability that a bound particle will not be released in the time interval Δt , $\gamma \Delta t$ is the probability that it will be released, and $\lambda_c e^{-\lambda_c(s-s')}$ is the probability that it will travel a distance $s - s'$ between release and recapture. If we let Δt go to zero, we obtain the integro-differential equation:

$$\frac{\partial p}{\partial t} = -\gamma p + \gamma \lambda_c \int_0^s ds' e^{-\lambda_c(s-s')} p(t, s') \quad (5)$$

We must also determine the initial condition. $p(s, 0)$ is simply the probability per unit length that a particle will travel a distance s from its initial position before being recaptured:

$$p(s, 0) = \lambda_c e^{-\lambda_c s} \quad (6)$$

We introduce the Laplace transform:

$$\bar{p}(k, t) = \int_0^{\infty} ds e^{-ks} p(s, t) \quad (7)$$

Taking the Laplace transform of (5) gives:

$$\frac{\partial \bar{p}}{\partial t} = -\gamma \bar{p} + \frac{\gamma \lambda_c}{k + \lambda_c} \bar{p} = -\frac{\gamma k}{k + \lambda_c} \bar{p} \quad (8)$$

The initial condition becomes:

$$\bar{p}(k, 0) = \frac{\lambda_c}{k + \lambda_c} \quad (9)$$

This gives

$$\bar{p} = \frac{\lambda_c}{k + \lambda_c} e^{-\frac{\gamma k}{k + \lambda_c} t} \quad (10)$$

Under certain circumstances this expression for the Laplace transform of $p(s, t)$ is of direct significance. Suppose that we have obtained $f(r, s)$ as a sum (or integral) over terms containing the exponential e^{-ks} . Then the integral in (2) simply gives the Laplace transform (7), and is equivalent to the substitution in $f(r, s)$ of:

$$e^{-ks} \quad \text{by} \quad \frac{\lambda_c}{k + \lambda_c} e^{-\frac{\gamma k}{k + \lambda_c} t} \quad (11)$$

It is still useful to find $p(s, t)$ itself. The inverse Laplace transform of (10) is:

$$p(s, t) = \lambda_c e^{-\lambda_c s - \gamma t} I_0(2\sqrt{\lambda_c s \gamma t}) \quad (12)$$

where $I_0(x)$ is the imaginary Bessel function:

$$I_0(x) = J_0(ix) = \sum_{n=0}^{\infty} \frac{1}{(n!)^2} \left(\frac{x}{2}\right)^{2n} \quad (13)$$

For large values of x ,

$$I_0 \simeq \frac{e^x}{\sqrt{2\pi x}} \quad (14)$$

Substituting this in (12) gives:

$$p(s, t) \simeq \lambda_c \frac{1}{\sqrt{4\pi \sqrt{\lambda_c s \gamma t}}} e^{-(\sqrt{\lambda_c s} - \sqrt{\gamma t})^2} \quad (15)$$

Thus for large values of s and t , $p(s, t)$ has a sharp peak at $\lambda_c s = \gamma t$, i. e., a particle of age t almost certainly has an age near $s = \frac{\gamma}{\lambda_c} t$. For this reason, the dependence of (2) and (3) on t for large t will be similar to that of $f(r, s)$ (and its integral over the crystal volume) on s for large s .

The Case of Uniform Initial Distribution

Next we determine the function $f(r, s)$ for the case in which the radioactive material is distributed uniformly throughout the volume of the bone crystal. $f(r, s)$ may be defined in terms of another function $g(r, n, s)$, which gives the probability per unit volume per unit solid angle that a particle will be found at a point r moving in a direction n (n is a unit vector) after it has travelled a distance s . Then $f(r, s)$ is given by the integral of $g(r, n, s)$ over all n :

$$f(r, s) = \int d\Omega g(r, n, s) \quad (16)$$

The function $g(r, n, s)$ is determined by the transfer equation:

$$\frac{\partial g}{\partial s} = -n \cdot \nabla g - \lambda_s g + \lambda_s \int \frac{d\Omega}{4\pi} g \quad (17)$$

The first term gives the propagation of g that would occur if no scattering were present. The second gives the decrease in g due to particles being scattered away from the direction n , and the last term gives the increase in g due to particles being scattered into the direction n .

The methods of solving (17) are extremely complex. In the case of a uniform initial distribution, we may make the approximation of using the much simpler "diffusion" equation instead of (17). We obtain the diffusion equation from (17) by making the approximation (see reference 1, p. 183):

$$g = \frac{1}{4\pi} f + n \cdot J \quad (18)$$

where f and J are independent of n , and by assuming we may ignore $\frac{\partial J}{\partial s}$. These approximations lead to the diffusion equation:

$$\frac{\partial f}{\partial s} = \frac{1}{3\lambda_s} \nabla^2 f \quad (19)$$

The actual "best boundary condition" on (19) is (see Reference 1, p. 186):

$$f = -0.7104 \frac{1}{\lambda_s} \frac{\partial f}{\partial r} \quad (20)$$

on the surface of the bone crystal, where $-\frac{\partial f}{\partial r}$ is the inward normal gradient. However, this boundary condition is equivalent to $f = 0$ on a surface slightly outside the bone crystal, and since we hardly know the size of the bone crystal to within $1/\lambda_s$, we will use the simpler boundary condition $f = 0$ as holding on the surface of the crystal.

We assume that the bone crystal is an infinitely long cylinder of radius ℓ . For an initial condition we set $f(r, 0) = \frac{1}{\pi \ell^2}$ which gives an initial distribution of one unit per unit length of the cylinder. Under these conditions and $f = 0$ on the surface, the solution of (19) is

$$f(r, s) = \frac{2}{\pi \ell^2} \sum_{n=1}^{\infty} \frac{J_0(\eta_n \frac{r}{\ell})}{J_1(\eta_n) \eta_n} e^{-\frac{\eta_n^2}{\ell^2} \frac{1}{3\lambda_s} s} \quad (21)$$

(see Reference 2, p. 271) where J_0 and J_1 are the conventionally defined Bessel functions of the first kind, and η_n is the n th zero of J_0 . The integral of this function over the volume (actually the cylindrical cross-section) of the bone crystal is:

$$\int (dr) f(r, s) = \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} e^{-\frac{\eta_n^2}{\ell^2} \frac{1}{3\lambda_s} s} \quad (22)$$

The expression (3) may be evaluated by using the substitution (11). This gives:

$$\int (dr) \int_0^{\infty} ds f(r, s) p(s, t) = \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} \frac{3\ell^2 \lambda_s \lambda_c}{3\ell^2 \lambda_s \lambda_c + \eta_n^2} e^{-\frac{\eta_n^2}{3\ell^2 \lambda_s \lambda_c + \eta_n^2} \gamma t} \quad (23)$$

One caution must be observed in interpreting (23). It is the proportion of the initial deposit which remains at a time t after the material was initially released from the bound state. What we wish to find is the proportion of the initial deposit which remains at a time t after the material was initially bound into the crystal. Now if a unit amount is deposited throughout the bone crystal at time 0, during an interval $\Delta\tau$ at time τ an amount $\gamma e^{-\gamma\tau}$ of this material will be released for the first time. At a later time

t the proportion of this which will remain in the crystal will be:

$$\int (dr) \int_0^{\infty} ds f(r, s) p(s, t - \tau) \quad (24)$$

Therefore the total amount remaining in the crystal at time t will be:

$$\begin{aligned} G_0(t) &= \int_0^t d\tau \gamma e^{-\gamma\tau} \int (dr) \int_0^{\infty} ds f(r, s) p(s, t - \tau) + e^{-\gamma t} \\ &= \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} \left[e^{-\frac{\eta_n^2}{3\ell^2\lambda_s\lambda_c + \eta_n^2} \gamma t} - e^{-\gamma t} \right] + e^{-\gamma t} \end{aligned} \quad (25)$$

Now:

$$\sum_{n=1}^{\infty} \frac{4}{\eta_n^2} = 1 \quad (26)$$

Therefore:

$$G_0(t) = \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} e^{-\frac{\eta_n^2}{3\ell^2\lambda_s\lambda_c + \eta_n^2} \gamma t} \quad (27)$$

We may simplify this expression by defining some new parameters.

Let

$$\begin{aligned} \tau = \gamma t, \quad \mu &= \frac{\lambda_c}{\lambda_s}, \quad \text{and } \sigma = \ell\lambda_s. \quad \text{Then} \\ G_0(\tau) &= \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} e^{-\frac{\eta_n^2}{3\sigma^2\mu + \eta_n^2} \tau} \end{aligned} \quad (28)$$

It may be seen that (28) exhibits an exponential behavior for large τ . This difficulty cannot be circumvented by altering the bone crystal shape or the initial distribution; it is an inherent property of diffusion processes in finite media that they exhibit exponential behavior for very large times. However, it is quite possible that during an actual lifetime τ never reaches a value large enough for the exponential behavior to occur. This is the only possibility that would lead to a highly nonexponential form such as (1). Unfortunately, G_0 has the property that for such small values of τ it does not decrease far from its initial value of unity, i. e., by the time it has dropped to one-half of its initial value, it is almost a pure exponential. We shall see that this difficulty does not occur for the case of a surface initial distribution.

The Case of Surface Initial Distribution

We now wish to find $f(r, s)$ corresponding to an initial distribution where a unit amount of radioactive material crosses the surface of the bone crystal in an inward direction at time 0. In this case we cannot use the simple diffusion equation (19), since our initial condition contradicts the approximation (18). Thus we must consider original transfer equation (17), and find $f(r, s)$ as the integral (16) of $g(r, n, s)$.

First we introduce the Laplace transform

$$\bar{f}(r, k) = \int_0^{\infty} ds e^{-ks} f(r, s) \quad (29)$$

We may impart a simple physical interpretation to $\bar{f}(r, k)$. Consider the steady-state situation in which a unit amount of material per unit s (i. e., a unit amount per unit time with a unit velocity) crosses the crystal surface of a crystal which is similar to our original one, but which absorbs material at a rate of k per unit length. Then the amount of material absorbed per unit time (actually per unit s) per unit volume at r is $f(r, s)$ times the probability of absorption ke^{-ks} integrated over s :

$$\int_0^{\infty} ds ke^{-ks} f(r, s) = k \bar{f}(r, k) \quad (30)$$

Thus we may find $k \bar{f}(r, k)$ by solving the steady-state absorbtive transfer equation:

$$-n \cdot \nabla g - (\lambda_s + k) g + \lambda_s \int \frac{d\Omega}{4\pi} g = 0 \quad (31)$$

Now we have noted that the range of τ in which we are interested is limited, since for large τ the retention function will exhibit exponential behavior. This exponential behavior will occur when a substantial number of particles have path lengths of the order of the crystal dimensions. If we limit ourselves to much smaller times, this will be equivalent to limiting s to values which are small compared to the crystal dimensions. In this case, the actual shape of the crystal is immaterial, and we may consider the simpler situation of a semi-infinite bone crystal with a plane surface. This has the advantage of reducing (31) to the one-dimensional form:

$$-\cos \theta \frac{\partial g(x, \theta)}{\partial x} - (\lambda_s + k) g(x, \theta) + \lambda_s \int \frac{d\Omega}{4\pi} g(x, \theta') = 0 \quad , \quad (32)$$

(where the crystal surface is at $x = 0$). In addition, we have removed all dependence on the crystal dimensions.

Now consider an atom which enters the crystal at an angle θ_0 to the normal to the surface. Eventually this atom will either be absorbed or re-emitted from the surface. We define $1/\mu S(\mu_1 \mu_0)$ where $\mu = \cos \theta$,

$\mu_0 = \cos \theta_0$, as the probability that this atom will be re-emitted within $d\Omega$ of an angle θ (outward). From this function we may form the total probability K that an atom which entered the crystal in a random inward direction would be emitted in any direction:

$$K = 2\pi \int_0^1 d\mu \int_0^1 \frac{d\mu'}{\mu'} S(\mu, \mu') \quad (33)$$

Using (32) and considerations of reversability, it is possible to derive the restriction on S :

$$S(\mu, \mu_0) = S(\mu_0, \mu) \quad (34)$$

and the integral equation for S :

$$\left(\frac{1}{\mu} + \frac{1}{\mu_0} \right) S(\mu, \mu_0) = \frac{\lambda_s}{4\pi(\lambda_s + k)} \left[1 + 2\pi \int_0^1 S(\mu'', \mu_0) \frac{d\mu''}{\mu''} \right] \left[1 + 2\pi \int_0^1 S(\mu, \mu') \frac{d\mu'}{\mu'} \right] \quad (35)$$

(see Reference 1, pages 16 to 19). If we integrate (35) over μ_0 and μ , and substitute (33) and (34), we may obtain the simple algebraic equation:

$$4K = \frac{\lambda_s}{\lambda_s + k} (1 + K)^2 \quad (36)$$

which has the solution (the second solution is not physically meaningful):

$$K = \frac{\lambda_s + 2k}{\lambda_s} - \sqrt{\left(\frac{\lambda_s + 2k}{\lambda_s} \right)^2 - 1} \quad (37)$$

If K is the amount which is re-emitted, then the amount which is absorbed is just:

$$1 - K = k \int (dr) \bar{f}(r, k) \quad (38)$$

Therefore:

$$\int (dr) \bar{f}(r, k) = 2 \left\{ \sqrt{\frac{1}{k\lambda_s} + \frac{1}{\lambda_s^2}} - \frac{1}{\lambda_s} \right\} \quad (39)$$

We now return to our original time-dependent, non-absorbtive problem, by taking the inverse Laplace transform. Using the Fourier-Mellin inversion integral:

$$\int (dr) f(r, s) = \frac{1}{2\pi i} \int_{r-i\infty}^{r+i\infty} dk e^{ks} 2 \left\{ \sqrt{k \frac{1}{\lambda_s} + \frac{1}{\lambda_s^2}} - \frac{1}{\lambda_s} \right\} \quad (40)$$

The actual value of this inverse transform is

$$\int (dr) f(r, s) = e^{-\frac{\lambda_s}{2} s} \left\{ I_1 \left(\frac{\lambda_s}{2} s \right) + I_0 \left(\frac{\lambda_s}{2} s \right) \right\} \quad (41)$$

where I_0 and I_1 are imaginary Bessel functions. However, for the purpose of computing (3), it is simpler to consider the contour integral (40), since in this form the integration (3) may be performed by means of the substitution (11). We obtain a function which we label $F(t)$:

$$F(t) = \int (dr) \int_0^\infty ds f(r, s) p(s, t) = \frac{1}{2\pi i} \int_{r-i\infty}^{r+i\infty} dk \frac{\lambda_c}{\lambda_c - k} e^{\frac{\gamma k}{\lambda_c - k} t} 2 \left\{ \sqrt{\frac{1}{k\lambda_s} + \frac{1}{\lambda_s^2}} - \frac{1}{\lambda_s} \right\} \quad (42)$$

The path of integration must be considered carefully. In the complex k -plane the integrand of (40) has branch points at $k = 0$ and $k = -\lambda_s$, and the path of integration is a counterclockwise contour enclosing the branch points. However, the integrand of (42) has an additional essential singularity at $k = \lambda_c$. It is important to notice that the contour of integration must exclude this point. This follows from the fact that the substitution (11) is valid only if $\int_0^\infty e^{ks} p(s, t) ds$ converges, and this only occurs for $k < \lambda_c$.

We now make the change of variable $z = \frac{\lambda_s}{k} - \frac{\lambda_s}{\lambda_c}$, and define $\mu = \frac{\lambda_c}{\lambda_s}$. This gives

$$F(t) = -\frac{1}{2\pi i} \int dz \frac{1}{z} e^{\frac{\gamma t}{\mu z}} \left\{ \frac{2\mu}{z\mu+1} \left(\sqrt{z+1+\frac{1}{\mu}} - 1 \right) \right\} \quad (43)$$

In the z -plane, the branch points are at $z = -1 - \frac{\lambda_s}{\lambda_c}$ and at infinity, and the essential singularity is now at the origin. Note that there is no singularity at $z = -\frac{\lambda_s}{\lambda_c}$. The path of integration has been mapped into a clockwise contour around the origin:

In (43) we now expand the exponential in a power series and the expression in parentheses in a Taylor series:

$$F(t) = -\frac{1}{2\pi i} \int dz \frac{1}{z} \sum_{m, n=0}^{\infty} \frac{1}{m!} \left(\frac{\gamma t}{\mu z} \right)^m z^n \frac{1}{n!} \frac{d^n}{dz^n} \left\{ \frac{2\mu}{z^{\mu+1}} \left(\sqrt{z+1+1/\mu} - 1 \right) \right\}_{z=0} \quad (44)$$

This is actually a Laurent series. Using Cauchy's integral theorem, we pick out the coefficient of $1/z$, obtaining:

$$\begin{aligned} F(t) &= \sum_{n=0}^{\infty} \frac{1}{(n!)^2} \left(\frac{\gamma t}{\mu} \right)^n \frac{1}{z^n} \left\{ \frac{2\mu}{z^{\mu+1}} \left(\sqrt{z+1+1/\mu} - 1 \right) \right\}_{z=0} \\ &= \sum_{n=0}^{\infty} \frac{1}{(n!)^2} \left(\frac{\gamma t}{\mu} \right)^n \sum_{k=0}^n \frac{n!}{k! (n-k)!} \frac{z^{n-k}}{z^{\mu+1}} \left(\frac{2\mu}{z^{\mu+1}} \right) \frac{d^k}{dz^k} \left(\sqrt{z+1+1/\mu} - 1 \right)_{z=0} \end{aligned} \quad (45)$$

$$= \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{\gamma t}{\mu} \right)^n \sum_{k=0}^n \frac{1}{k! (n-k)!} 2 (-1)^{n-k} (n-k)! \mu^{n-k+1} \begin{cases} \sqrt{1+\frac{1}{\mu}} - 1 & (k=0) \\ \frac{1}{2} \left(-\frac{1}{4} \right)^{k-1} \frac{(2k-2)!}{(k-1)!} \left(1 + \frac{1}{\mu} \right)^{\frac{1}{2}-k} & (k > 0) \end{cases}$$

Simplifying terms we obtain a power series expression for $F(t)$;

$$F(t) = 2\mu \left(\sqrt{1+\frac{1}{\mu}} - 1 \right) e^{-\gamma t} - \frac{1}{\sqrt{1+\frac{1}{\mu}}} \sum_{n=1}^{\infty} \frac{(-\gamma t)^n}{n!} \sum_{k=1}^n \frac{(2k-2)!}{(k-1)! k!} \frac{1}{(4+4\mu)^{k-1}} \quad (46)$$

We note that for $t = 0$, F is not unity. Instead

$$F(0) = 2\mu \left(\sqrt{1+\frac{1}{\mu}} - 1 \right) \quad (47)$$

This corresponds to the physical situation that in a crystal where both scattering and binding occur, some of the atoms which enter the crystal will not be bound at all, but will scatter out of the crystal at once. From the physical standpoint, however, only those particles which are bound at least once are of interest; thus we should renormalize $F(t)$. If we do this, and also set $\tau = \gamma t$, we may define a function $G_1(\tau)$, analogous to G_0 , which gives the proportion of the material which was bound by surface

adsorption at time 0 which remains at time τ :

$$G_1(\tau) = e^{-\tau} \frac{1}{2\mu(\sqrt{1+\frac{1}{\mu}}-1)\sqrt{1+\frac{1}{\mu}}} \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \sum_{k=1}^n \frac{(2k-2)!}{(k-1)k!} \frac{1}{(4+4\mu)^{k-1}} \quad (48)$$

Such a power series gives little information about the behavior of G_1 for large τ . To determine this behavior, we return to expression (42). Since the integral over small circles around the branch points goes to zero, we may replace the contour integral by twice the integral from zero to $-\lambda_s$ along a line just above the branch line. (Note that -1 disappears since it is continuous across the branch line.) This gives

$$F(t) = \frac{1}{\pi i} \int_{0+i\epsilon}^{-\lambda_s+i\epsilon} dk \frac{\lambda_c}{\lambda_c - k} e^{\frac{\gamma k}{\lambda_c - k} t} \frac{2}{\lambda_s} \sqrt{\frac{\lambda_s}{k} + 1} \quad (49)$$

We may obtain a real integral by rotating the k -plane 180° :

$$F(t) = \frac{1}{\pi} \int_0^{\lambda_s} dk' \frac{\lambda_c}{\lambda_c + k'} e^{-\frac{\gamma k'}{\lambda_c + k'} t} \frac{2}{\lambda_s} \sqrt{\frac{\lambda_s}{k} - 1} \quad (50)$$

Now we substitute $x = \frac{k'}{\lambda_c + k'}$. This gives:

$$F(t) = \frac{1}{\pi} \int_0^{\frac{1}{1+\mu}} dx \frac{2\sqrt{\mu}}{\sqrt{x}} \frac{1}{\sqrt{x}} e^{-x\gamma t} \left\{ \frac{1}{1-x} \sqrt{(1-x)-\mu x} \right\} \quad (51)$$

For large γt , the exponential becomes negligibly small except in a region near $x = 0$. Since the expression in parentheses is analytic throughout the range of integration, and is non-zero at $x = 0$, we may obtain an approximation for large τ by replacing this expression by its value (unity) at $x = 0$. Thus

$$\begin{aligned} F(t) &\xrightarrow{t \rightarrow \infty} \frac{2\sqrt{\mu}}{\pi} \int_0^{\frac{1}{1+\mu}} \frac{1}{\sqrt{x}} e^{-x\gamma t} = \frac{4}{\pi} \frac{\sqrt{\mu}}{\sqrt{\gamma t}} \int_0^{\sqrt{\frac{\gamma t}{1+\mu}}} dy e^{-y^2} \\ &= \sqrt{\frac{4\mu}{\pi \tau}} \operatorname{erf}\left(\sqrt{\frac{\tau}{1+\mu}}\right) \quad (52) \end{aligned}$$

Since the error function $\text{erf}(x)$ quickly approaches unity for large arguments, we obtain:

$$G_1(\tau) = \frac{1}{2\mu \left(\sqrt{1 + \frac{1}{\mu}} - 1 \right)} F \approx \frac{1}{\sqrt{\pi\mu} \left(\sqrt{1 + \frac{1}{\mu}} - 1 \right)} \frac{1}{\sqrt{\tau}} \quad (53)$$

What we have done is to replace the expression in parenthesis in (51) by the first term of its Taylor series expansion about $x = 0$. By using several terms of the Taylor series more accurate asymptotic expressions may be obtained.

Of course, it must be remembered that this $\tau^{-\frac{1}{2}}$ behavior will only hold as long as our semi-infinite crystal assumption is good. For very large τ the finite size of the crystal must be felt, and G_1 will go over into an exponential. Still, it is quite possible that G_1 may exhibit $\frac{1}{\sqrt{\tau}}$ behavior over a sizeable range of time. This strongly suggests that surface absorption on the bone crystals, i. e., diffuse activity in bone macroscopically, is the major factor in determining the body retention function.

Computation of the Bone Retention Functions

A table of the functions G_0 and G_1 was computed using an IBM 650 computer, and is given in the following pages. G_0 was computed from formula (28) with certain correction terms to compensate for the finite truncation of the series. For values of τ smaller than five, G_1 was computed using formula (48). For larger values the expression (51) (renormalized to give G_1) was integrated numerically. The numerical integration was quite time consuming, running about five minutes for each value computed.

Subroutines for evaluating G_0 and G_1 by these methods, written in SOAP II, are available. They require indexing registers and floating-point arithmetic.

THE FUNCTION G_0

$\sigma^2 \mu$	τ	G_0	$\sigma^2 \mu$	τ	G_0
2000	0.1	0.99750	200	0.1	0.99220
2000	0.2	0.99512	200	0.2	0.98479
2000	0.4	0.99068	200	0.4	0.97098
2000	0.7	0.98473	200	0.7	0.95251
2000	1.0	0.97948	200	1.0	0.93626
2000	2.0	0.96556	200	2.0	0.89339
2000	4.0	0.94659	200	4.0	0.83575
2000	7.0	0.92705	200	7.0	0.77752
2000	10.0	0.91197	200	10.0	0.73342
2000	20.0	0.87475	200	20.0	0.62795
2000	40.0	0.82369	200	40.0	0.49177
2000	70.0	0.76906	200	70.0	0.35903
2000	100.0	0.72651	200	100.0	0.26731
1000	0.1	0.99647	100	0.1	0.98907
1000	0.2	0.99311	100	0.2	0.97869
1000	0.4	0.98686	100	0.4	0.95937
1000	0.7	0.97847	100	0.7	0.93354
1000	1.0	0.97108	100	1.0	0.91086
1000	2.0	0.95149	100	2.0	0.85125
1000	4.0	0.92486	100	4.0	0.77181
1000	7.0	0.89752	100	7.0	0.69263
1000	10.0	0.87649	100	10.0	0.63352
1000	20.0	0.82488	100	20.0	0.49564
1000	40.0	0.75473	100	40.0	0.32791
1000	70.0	0.68062	100	70.0	0.18427
1000	100.0	0.62368	100	100.0	0.10438
400	0.1	0.99445	40	0.1	0.98304
400	0.2	0.98917	40	0.2	0.96694
400	0.4	0.97934	40	0.4	0.93703
400	0.7	0.96618	40	0.7	0.89717
400	1.0	0.95458	40	1.0	0.86228
400	2.0	0.92391	40	2.0	0.77132
400	4.0	0.88243	40	4.0	0.65247
400	7.0	0.84017	40	7.0	0.53783
400	10.0	0.80788	40	10.0	0.45540
400	20.0	0.72951	40	20.0	0.27809
400	40.0	0.62527	40	40.0	0.10999
400	70.0	0.51851	40	70.0	0.02768
400	100.0	0.43935	40	100.0	0.00697

THE FUNCTION G_1

μ	τ	G_1	μ	τ	G_1
1.00	0.1	0.98555	0.10	0.1	0.96606
1.00	0.2	0.97149	0.10	0.2	0.93401
1.00	0.4	0.94447	0.10	0.4	0.87508
1.00	0.7	0.90652	0.10	0.7	0.79806
1.00	1.0	0.87143	0.10	1.0	0.73254
1.00	2.0	0.77191	0.10	2.0	0.57421
1.00	4.0	0.63110	0.10	4.0	0.40885
1.00	7.0	0.50374	0.10	7.0	0.30202
1.00	10.0	0.42720	0.10	10.0	0.24969
1.00	20.0	0.30418	0.10	20.0	0.17425
1.00	40.0	0.21530	0.10	40.0	0.12247
1.00	70.0	0.16279	0.10	70.0	0.09235
1.00	100.0	0.13620	0.10	100.0	0.07719
0.50	0.1	0.97926	0.05	0.1	0.96211
0.50	0.2	0.95929	0.05	0.2	0.92651
0.50	0.4	0.92149	0.05	0.4	0.86156
0.50	0.7	0.86975	0.05	0.7	0.77773
0.50	1.0	0.82328	0.05	1.0	0.70744
0.50	2.0	0.69880	0.05	2.0	0.54213
0.50	4.0	0.54130	0.05	4.0	0.37799
0.50	7.0	0.41663	0.05	7.0	0.27709
0.50	10.0	0.34842	0.05	10.0	0.22872
0.50	20.0	0.24520	0.05	20.0	0.15946
0.50	40.0	0.17287	0.05	40.0	0.11204
0.50	70.0	0.13050	0.05	70.0	0.08447
0.50	100.0	0.10913	0.05	100.0	0.07060
0.20	0.1	0.97114	0.02	0.1	0.95842
0.20	0.2	0.94369	0.02	0.2	0.91952
0.20	0.4	0.89270	0.02	0.4	0.84904
0.20	0.7	0.82492	0.02	0.7	0.75906
0.20	1.0	0.76610	0.02	1.0	0.68458
0.20	2.0	0.61856	0.02	2.0	0.51366
0.20	4.0	0.45350	0.02	4.0	0.35148
0.20	7.0	0.33915	0.02	7.0	0.25609
0.20	10.0	0.28121	0.02	10.0	0.21118
0.20	20.0	0.19665	0.02	20.0	0.14714
0.20	40.0	0.13832	0.02	40.0	0.10336
0.20	70.0	0.10433	0.02	70.0	0.07792
0.20	100.0	0.08721	0.02	100.0	0.06512

The Whole Body Retention Function

We must still establish the relationship between G_0 and G_1 and the whole body retention function $R(t)$. Since we are only interested in times larger than the holding times of non-bone compartments, we may ignore the effects of these compartments. The only factors which we must consider are the probability k that an atom released by the bone will be recaptured by the bone, and the probability β that if recaptured it will be captured by a hot-spot formation.

We first define a general bone retention function G which is the appropriate linear combination of G_0 and G_1 :

$$G(t) = \beta G_0(t) + (1 - \beta) G_1(t) \quad (54)$$

This gives the probability that an atom deposited in the bone at time zero is still in the bone at time t , without having ever left the bone.

But many of the atoms which leave the bone are recaptured. Let $R(t)$ be the probability that an atom deposited in the bone at time zero is still in the bone at time t , including the probability of recapture. $R(t)$ consists of $G(t)$ plus a term giving the retention of recaptured atoms. Specifically,

$$R(t) = G(t) + \int_0^t dt' E(t') G(t-t') \quad (55)$$

where $E(t)$ is the rate at which atoms are recaptured by bone. Now consider the atoms which leave the bone: k of them are recaptured and $1-k$ are eliminated. Thus $E(t)$ is $k/1-k$ of the rate of elimination from the body (excluding those atoms which are eliminated without ever entering the bones). But the rate of elimination is just the negative of the derivative of the whole-body retention function $R(t)$. Therefore:

$$E(t) = - \frac{k}{1-k} \frac{d}{dt} R(t) \quad (56)$$

This gives the integro-differential equation for $R(t)$:

$$R(t) = G(t) - \frac{k}{1-k} \int_0^t dt' G(t-t') \frac{d}{dt'} R(t') \quad (57)$$

We introduce the Laplace transform:

$$\bar{R}(\omega) = \int_0^\infty dt e^{-\omega t} R(t) \quad (58)$$

Taking the transform of (57), we obtain:

$$\bar{R}(\omega) = \bar{G}(\omega) - \frac{k}{1-k} \bar{G}(\omega) [\omega \bar{R}(\omega) - R(0)] \quad (59)$$

Now $R(0) = 1$, thus $\bar{R}(\omega)$ is

$$\bar{R}(\omega) = \frac{\bar{G}(\omega)}{1-k[1-\omega \bar{G}(\omega)]} \quad \text{where } \bar{G}(\omega) = \beta \bar{G}_0(\omega) + (1-\beta) \bar{G}_1(\omega) \quad (60)$$

The Laplace transform of G_0 is trivial. From (28)

$$\bar{G}_0(\omega) = \int_0^\infty dt e^{-\omega t} G_0(t) = \sum_{n=1}^{\infty} \frac{4}{\eta_n^2} \int_0^\infty dt e^{-\left(\omega + \frac{\eta_n^2 \gamma}{3\sigma^2 \mu + \eta_n^2}\right)t} = \sum_{n=1}^{\infty} \frac{4}{\eta_n^2 \left(\omega + \frac{\eta_n^2 \gamma}{3\sigma^2 \mu + \eta_n^2}\right)} \quad (61)$$

This function has an infinite sequence of simple poles at

$$\omega = -\frac{\eta_n^2 \gamma}{3\sigma^2 \mu + \eta_n^2} \quad \text{where } n = 1, 2, 3, \dots \quad (62)$$

which lie between $-\gamma$ and $-\frac{\eta_0^2 \gamma}{3\sigma^2 \mu + \eta_0^2}$ on the real axis.

The Laplace transform of G_1 is more complicated. Using (42) we obtain:

$$\bar{G}_1(\omega) = \frac{1}{2\pi i} \frac{1}{2\mu \left(\sqrt{1 + \frac{1}{\mu}} - 1\right)} \int_{\gamma+i\infty}^{\gamma-i\infty} dk \frac{\lambda_c}{\lambda_c - k} 2 \left\{ \sqrt{\frac{1}{k\lambda_s} + \frac{1}{\lambda_s^2}} - \frac{1}{\lambda_s} \right\} \int_0^\infty dt e^{-\omega t} e^{\frac{\gamma k}{\lambda_c - k} t} \quad (63)$$

Carrying out the t -integration gives:

$$\bar{G}_1(\omega) = \frac{1}{2\pi i} \frac{1}{2\mu \left(\sqrt{1 + \frac{1}{\mu}} - 1\right)} \int_{\gamma-i\infty}^{\gamma+i\infty} dk \frac{\lambda_c}{\lambda_c - k(\omega + \gamma)} 2 \left\{ \sqrt{\frac{1}{k\lambda_s} + \frac{1}{\lambda_s^2}} - \frac{1}{\lambda_s} \right\} \quad (64)$$

The integrand has branch points at zero and at $-\lambda_s$, and a simple pole at $\frac{\lambda_c \omega}{\omega + \gamma}$. The path of integration is to the left of the simple pole, since

otherwise the t -integral in (63) would not converge. If we make the change of variable $z = \frac{\lambda_s}{k}$ and introduce $\mu = \frac{\lambda_c}{\lambda_s}$, we obtain:

$$\bar{G}_1(\omega) = \frac{-1}{2\pi i} \frac{1}{2\mu\left(\sqrt{1+\frac{1}{\mu}}-1\right)} \int dz \frac{2}{z} \frac{\mu}{\mu\omega z - (\omega + \gamma)} (\sqrt{z+1}-1) \quad (65)$$

In the z -plane, the branch points are at -1 and infinity, and the simple pole is at $\frac{\omega + \gamma}{\mu\omega}$. Note that there is no singularity at zero. The path of integration has been mapped into a clockwise contour around the simple pole:

Thus we can use Cauchy's integral theorem to perform the integration, giving:

$$\bar{G}_1(\omega) = \frac{1}{(\omega + \gamma) \left(\sqrt{1 + \frac{1}{\mu}} - 1 \right)} \left[\sqrt{\frac{\omega + \gamma}{\mu \omega} + 1} - 1 \right] \quad (66)$$

This function has branch points at $\omega = 0$, and $-\frac{\gamma}{1 + \mu}$. There is no singularity at $-\gamma$. It should be noted that the $R(t)$ we have defined here is not equivalent to (1). It must be multiplied by the probability that an atom entering the body will deposit in the bone in the first place. This probability is certainly smaller or equal to k .

Although we have obtained the Laplace transform of $R(t)$, there is apparently no analytic method of obtaining the inverse transform or even of greatly simplifying the transformed function. This stems from the major difference in the forms of $G_0(\omega)$ and $G_1(\omega)$. To find $R(t)$ it will probably be necessary to use numerical techniques, either to invert the transform or to directly integrate (57). Such computations have not yet been performed.

References

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DISCUSSION OF J. C. REYNOLDS' MATHEMATICAL THEORY OF THE RETENTION OF BONE-DEPOSITING RADIOACTIVE ELEMENTS*

John H. Marshall

Abstract of Reynolds' Paper

A mathematical theory for the diffusion of radioactive atoms in bone crystallites is developed in an attempt to find a physical explanation for the empirically-determined power function retention formula. Two initial distributions of radioactive atoms within a typical crystallite are considered: A uniform distribution representing deposits in forming bone (hotspots), and a surface distribution representing deposition in the diffuse component. The loss of activity by diffusion from hotspots is obtained for cylindrical crystallites in terms of the ratio σ of crystal radius to scattering mean free path, the ratio μ of scattering to capture mean free paths, and the time τ in units of the mean lifetime (γ^{-1}) of a diffusing atom in any individual site in the crystal lattice. The loss of activity by diffusion from the diffuse component is obtained as a function of μ and τ assuming semi-infinite crystals. The results are shown in Fig. 29 as solid curves.

Fig. 29

*Previous paper in this report.

Several conclusions follow from Reynolds' results:

- (1) The diffusion loss of activity from hotspots (initially a uniform distribution within crystallites) does not yield a power function.
- (2) The diffusion loss of activity from the diffuse component (initial distribution on crystallite surfaces) yields the power function, $t^{-0.5}$, when semi-infinite crystals are assumed.
- (3) For finite crystals, both types of deposit yield a retention function which closely approximates an exponential after a time sufficiently long for diffusing atoms to attain mean displacements comparable to the crystallite radius (approximately $\tau > \sigma^2 \mu$).

Comparison with Compartment Theory

This mathematical theory may be compared with the compartment theory for the same diffusion problem.⁽¹⁾ The basic assumptions differ somewhat.

The mathematical theory describes the path of a diffusing atom as a series of straight segments punctuated by either scattering or capture events. The directions and free paths between these events are randomly distributed. For simplicity, it is assumed that only scattering events change the direction of motion and that capture events do not.

The compartment theory divides the crystallite into cylindrical shells and considers the population of each shell as a function of time. Therefore, the path of a diffusing atom is described as a series of radial (or circumferential) segments of given length, punctuated only by capture events.

Both theories make the assumption that re-emission after capture at an individual site is a random process and hence yields an exponential distribution of capture times. Long cylindrical crystallites of small diameter are considered by both theories for the hotspot type of deposit, but only by the compartment theory for the diffuse type of deposit.

In the mathematical theory, the radius of the crystallite in scattering mean free paths, σ , corresponds roughly to the number of radial shells, n , in the compartment theory. Since the compartment theory deals only with capture, it corresponds to the mathematical condition $\mu = 1$. The inverse capture lifetime γ corresponds to the probability per unit time of a jump from one radial shell to the next times a factor to account for the possibility of circumferential jumps which would not affect the population of radial shells. Roughly, only one of the six possible rectangular directions

for a jump is involved in a change of radial shell, so one might expect this factor to be 6. The results of the two theories, however, match best if this factor is 2.5 or

$$\gamma \approx 2.5 \left[\frac{f n^2}{2n-1} \right]$$

where the bracket contains the reciprocal of the time constant of a single compartment for flow into one neighboring compartment according to the compartment theory.

Note that the compartment theory does not use an arbitrary time scale but ties the compartment time constants to the observable rate of diffuse uptake f . Although this additional restraint could be built into the mathematical theory, the latter does not now describe diffuse uptake.

In Fig. 29 we compare the retention results of the two theories for $f = 10^{-3} \text{ day}^{-1}$, the long-term exchange rate characteristic of 1 year old bone. For $n = 6$, $\gamma^{-1} = 120$ days and $\sigma^2 \mu = 36$, for $n = 30$, $\gamma^{-1} = 26$ days and $\sigma^2 \mu = 900$. Note that changing the value of σ or n changes the time scale because $\tau = t / \gamma$ in the mathematical theory. The compartment theory values are plotted in Fig. 29 as circled points for $n = 30$ and as circled crosses for $n = 6$.

The agreement between the two theories is good regarding hotspot deposits in large or small crystals and regarding diffuse deposits in large crystals. The compartment theory result for the diffuse component in crystals of physiological size ($n \sim 6$), however, has no counterpart in the mathematical theory, which assumes only semi-infinite crystals for this case; the circled crosses for the diffuse component in Fig. 29 fall below the mathematical $t^{-.5}$ curve for $\mu = 1$ after about 1 year ($\tau \sim 3$) and more closely approximate an exponential than a power function.

Conclusions

Where they overlap, the compartment theory and the mathematical theory agree as to the shape of the retention curve for an isolated radioactive deposit subject to intracrystalline diffusion. The compartment theory, however, provides a solution for diffuse retention based on small crystals of physiological size and, in contrast to the mathematical theory, finds that not even the diffuse retention curve can be described by a power function.

The observed power function retention curves therefore cannot be derived simply from diffusion theory. We are forced to consider the effect of different rates of mineral turnover in different regions and of rates varying with time in individual regions of bone.

Reference

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PRELIMINARY STUDY ON THE RETENTION OF Th^{232} DAUGHTERS FOLLOWING THOROTRAST INJECTIONS

C. E. Miller and L. D. Marinelli

A survey of the literature on this subject disclosed that information on the distribution of radioactivity in humans following Thorotrast injection is incomplete.⁽¹⁾ The data seem to be consistent in establishing translocation of Th^{232} daughters from the deposits in liver and spleen many years after injection. From what is known about the metabolism of Ra and the fact that the Ra^{228} and Ra^{224} are excreted in the urine, it is possible to predict that these isotopes will be eventually found in bone, and predictions as to the amounts can be made with a modicum of confidence. Much less is known, however, about the fate of Th^{228} leaving liver and spleen. A review of findings in bone is to be found in Marinelli and Stehney's report immediately following. Of definite importance to the dosimetry of these cases is the detailed picture of the initial uptake of Th^{232} daughters present in the solution injected since the behavior of these elements cannot be strictly foretold from their behavior in ionic form.

Through the kind cooperation of Dr. S. F. Thomas of Palo Alto, we have been able to study a section of liver and rib specimen from a patient who died 9 days after the injection of 48cc of Thorotrast (Batch No. 07860). Bottles and syringes used for injection were mailed to us in order to ascertain the exact nature of the material plated out in the glassware. All analyses were done by gamma-ray spectrometry during a period of about a year in order to establish the original amount of Th^{232} in the specimen from the growth of the gamma-ray lines of Ac^{228} and the behavior of Th^{228} through the growth of the gamma rays of ThB and TBC . The solution seems to have been from 4 to 5 years old at injection, on the basis of measurement of the $\text{Ra}^{228}/\text{Th}^{232}$ and $\text{Th}^{228}/\text{Th}^{232}$ ratios and the assumption that soon after preparation time Ra^{228} was absent and $\text{Th}^{228}/\text{Th}^{232} = 1$. Fractions remaining in the bottles and syringe were roughly 50% Ra^{228} and 17% Th^{228} , giving an "injected" radioactive equilibrium $\text{Th}^{232}/\text{Ra}^{228}/\text{Th}^{228} = 100:19:35$ to be compared to 100:10:40 reported by Hursh.⁽²⁾

Findings in the liver disclosed that in the liver specimen these ratios were roughly 100:7:20, whereas in the rib samples they were 100:9:18. We may compute that, if Thorotrast were injected at equilibrium, the initial activity ratio in liver would be of the order 100:40:63 and in the ribs 100:50:54.

It is to be noted that Ra^{228} in the injected solution is far from being completely washed out from the liver soon after injection and that the Th^{228} therein is present in "metabolic" ratios comparable to those found at much later times when the Th^{228} present is grown from its parents. Therefore,

the deciding dosimetric factor in the liver seems to be the radioactive equilibrium of the material injected, and this may vary from bottle to bottle as well as with the age of the Thorotrast. As to the material deposited in the ribs, the high fractions of Th^{232} and Th^{228} suggest that the marrow in the specimen retained most of these elements. Autoradiographic studies have been initiated to ascertain their exact location.

The fate of Ra^{224} remains to be explored in fresh specimens obtained soon after death. The role of in vivo spectrometric techniques in bone is dubious at the present time, for it faces the difficulty of designing an experimental technique which allows the spectrometry of compact bone wholly devoid of marrow.

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RADIOACTIVITY IN BONES OF PATIENTS INJECTED WITH THOROTRAST

L. D. Marinelli and A. F. Stehney

Specimens of bone from patients injected with Thorotrast have proved to be difficult to obtain. During the last year it has been possible to gather from three different sources scattered bone measurements from 5 patients. The essential data are given in Table 20; they have been normalized to an injection of 20 cc of Thorotrast; namely, 4 g(0.44 μ c) of Th^{232} ; skeletal contents were computed on the basis of a total fresh weight of 7×10^3 gram and ash weight of 2.8×10^3 grams for the entire skeleton.

Table 20
Analysis of bone from Thorotrast patients

Patient	Duration of burden	Specimen	Time between biopsy and analysis	Skeletal activity (10^{-8}C) per injection of 4g ($44 \times 10^{-8}\text{C}$) Th^{232}			
				Th^{232}	Ra^{228}	Th^{228}	Source
R. C.	18 yr.	Molar tooth 0.9g ashed wt.	350 days	.22	-	.74	A. F. Stehney, personal communication 1/10/59; ashed weights
No. 1	15-20 yrs.	Body of vertebra (12g)	2 wks.	-	8.3	9.3	J. B. Hursh, personal communication 11/19/57; fresh weights
		Head of femur (2.7g)		-	3.5	2.9	
No. 2	20 yr.	Sternum bone (1.41 and 1.35g)	≤ 180 days	38.0(?)	6.8	6.5	J. B. Hursh et al., Acta Rad. 47:481, 1957; ashed weights
				-	6.2	7.3	
A. D.	19 yr.	Cancellous bone (25 mg ashed)	-	11.6(?)	-	-	J. B. Hursh et al., Acta Rad. 47:481, 1957; ashed weights
		Compact bone (83 mg ashed)		.24	-	-	
		Trabecular bone (4.2 μ g)		-	-	.76	W. S. Jee and C. Mays, personal communication 1958, based on autoradiography of single bone section
M. H.	17 days	Cancellous bone (97 mg ashed)	-	2.64	-	-	J. B. Hursh, Acta. Rad. 1957; ashed weights
		Compact bone (20 mg ashed)		.24	-	-	
Theoretical	Maximum			-	.18	5.5	Note: Average background radiation dose to skeleton roughly equivalent to $0.04 \times 10^{-8}\text{C}$ Th^{228}
	Minimum			-	.18	.18	

No definite pattern of bone burden is apparent. In cases No. 2 and A.D. the Th^{232} contents are unusually high (86% and 26% of the burden), indicating possible contamination of the specimen by marrow.* Striking is the

*Personal communication from Dr. J. B. Hursh confirms this hypothesis by calling attention to the high Th^{232} concentration in marrow and to the fact that tested specimens were not freed of marrow.

similarity of Th^{232} contents in compact bone (patients A.D. and M.H.) and tooth; they could be taken as representative ($\approx 0.5\%$ of body burden) of initial Th^{232} migration to the skeleton remaining invariant thereafter for decades. Comparison between Ra^{228} and Th^{228} (patients No. 1 and No. 2) would indicate that little of the Th^{228} gets to bone directly from the reticulo-endothelial (R.E.) system but that it grows there from Ra^{228} .

However, it is difficult to visualize why the deposition of Ra^{228} is so much higher than predicted, unless the samples were contaminated by marrow,* or unusual "washout" occurred in the R. E. system. The marrow-contamination hypothesis is qualitatively consistent with the higher values observed for vertebra and sternum as compared with the head of the femur; moreover, these same results could have been obtained, even without contamination, if the thorium isotopes did concentrate preferentially on the periosteal and endosteal surfaces of bone and the samples were representative of these sites.

It is evident that, despite variations by factors as high as ten in the sampling, the Th^{228} content falls within the levels predicted by Reynolds et al.⁽¹⁾ However, the crucial point of the dosimetry of bone in Thorotrast cases is the in vivo Ra^{224} content which, as yet, has not been reported. For this task analysis of the specimen must be carried out within a short time of sample collection. For the moment, we note that the lowest figures referred to here (patient R.C.'s tooth) indicate that the minimum dose to bone due to Th^{232} and Th^{228} (the latter in virtual equilibrium with its daughters) is likely to be of the order of 25 times the average background radiation dose delivered to the normal skeleton; the highest (discounting the anomalies in Th^{232} findings) is at least ten times higher, even if we neglect the role of Ra^{224} released by the R.E. system. Thanks are due to Dr. R. J. Hasterlik, Argonne Cancer Hospital, for supplying the tooth specimen.

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1. Reynolds, J. C., P. F. Gustafson and L. D. Marinelli. Retention and Elimination of Radium Isotopes Produced by the Decay of Thorium Parents within the Body - Calculations and Comparison with Experimental Findings. ANL-5689. (November 1957).

*See previous page.

Th^{228} , Ra^{228} , AND Ra^{226} IN BONE

D. E. Wallace, A. F. Stehney and F. H. Ilcewicz

I. Contribution of Ra^{228} in Cases of Bone Tumors

A previous report⁽¹⁾ described procedures for measurement of Ra^{228} in bone and indicated the accuracy to be expected and variations encountered in samples from individual subjects. Detailed results on six radium patients were given at that time. Data on these patients are repeated in condensed form in Table 21, together with results on five additional subjects and comparisons with values reported by MIT on different specimens from the same patients. The table is meant to present the data bearing on the question of the relative importance of Ra^{226} and Ra^{228} in inducing bone tumors.

Table 21

Ra^{228} and Ra^{226} content of radium patients

Patient	Route ^a	Approx. year, t_1	Body burden, $\mu\text{g Ra}^{226}$	$\sigma_{\text{Ra}^{228}}$ at t_1 Relative to Ra^{226}		Bone tumor
				ANL	MIT	
I	M-I	1931	0.8	b		No
(L-1) ^c	D-O	1918	1.0	6.5 ± 0.8	e	Yes
(M-1) ^c	M-I	1928-30	1.2	2.7 ± 0.5		Yes
112	M-O	1928	1.3	1.2 ± 0.2	$\sim 2.3^d$	Yes
(J-1) ^c	D-O	1924	1.3	2.4 ± 0.4	~ 1.8	Yes
313	D-O	1924	1.3	b		Yes
(B-1) ^c	D-O	1923	2.7	14 ± 2		Yes
R	M-I	1931	3.6	b	< 0.1	Yes
302	D-O	1921	3.8	12 ± 1	~ 16	Yes
103	M-I	1925	6.8^f	b		Yes
118	M-O	1918	$10(?)$	b		No

^aM-I: medical-intravenous; M-O: radium water taken by mouth;
D-O: radium dial painter.

^bConcentrations of Th^{228} were within the range found in normal bones and an extrapolation to t_1 was not considered justified.

^cTemporary code designations.

^dComputed at t_1 for analysis done in 1952 or 1953, from data reported by R. D. Evans in personal communication reported by Looney, Hasterlik, et al., (5).

^eApproximately 10% (by personal communication to L. D. Marinelli from H. Q. Woodward).

^fThis value was incorrectly given as 5.3 in Reference (1).

The data indicate that out of eight patients who developed tumors with Ra^{226} burdens below $5 \mu\text{g}$ of Ra , two showed Ra^{228} contents no higher than those found in normal individuals. It would seem, therefore, that the lowest verified pure Ra^{226} carcinogenic burden is $1.3 \mu\text{g}$. This statement, however, is predicated on the assumption that no significant amount of Th^{228} could have lodged in the skeleton of patient 313 (a radium dial painter) through ingestion. The significance of the relatively low values of Ra^{228} in the other two patients with burdens below 1.3 cannot be wholly appraised until more is learned about the carcinogenic effect of Ra^{228} deposited in the skeleton and the role of the rate of intake. Subject 118 developed no bone tumors although the Ra^{226} burden is probably close to $10 \mu\text{g}$ as deduced from concentrations of about $5 \times 10^3 \mu\mu\text{g Ra}^{226}$ per g of ashed bone; literature values of 2 to $10 \mu\text{g Ra}^{226}$ have been given for this subject.⁽²⁾

II. Distribution of Th^{228} between Organic and Inorganic Portions of Bone

The appreciable concentration of Ra^{228} in bone specimens from subject B-1 provided an opportunity to demonstrate that very little, if any, Th^{228} is removed from bone when the organic matter is extracted by ethylene diamine. What was really wanted was to determine what fraction of Th^{228} formed by Ra^{228} in bone crystals goes to the organic portion of bone, but, as discussed below, no clear conclusion could be reached on this point.

Two samples of bone were carefully cleaned of surface tissue and marrow, then extracted in a Soxhlet apparatus for 70 hours with 2 portions of boiling ethylene diamine. This was followed by extraction with water to remove the base. Analyses for both Th^{228} and Ra^{226} were then performed on the inorganic portion and on the residue remaining in the extractants for each sample. Table 22 shows that only 0.6% of the total amount of Th^{228} was found in the organic residue and that 0.5% of the Ra^{226} was also present in the organic residue. Previous work had shown that blank values of less than $0.1 \mu\mu\text{c}$ of Th^{228} or Ra^{226} are obtained in the organic residue.⁽¹⁾ These results may be interpreted in two ways; namely, (1) that a small fraction of Th^{228} goes to the organic matter, but only to the same extent as does Ra^{226} ; or (2) that about 0.5% of the inorganic matter was accidentally carried over into the extractants. In the latter case, an upper limit of about 0.2% of the Th^{228} in the organic matter may be calculated after corrections for radioactive decay since death of the subject.

The above results do not conclusively prove that Th^{228} formed from Ra^{228} remains in the inorganic fraction of bone. Although a well-established procedure for the ethylene diamine extraction⁽³⁾ was used with painstaking care, complete removal of organic material was not established. Furthermore, reprecipitation of Th^{228} onto bone crystals may well have occurred in the basic medium of the amine. Thus, our conclusion is merely that further information on the distribution Th^{228} in bone cannot be obtained through extraction with ethylene diamine.

Table 22

Distribution of Ra^{226} and Th^{228} after extraction of organic material from bone with hot ethylene diamine

Subject	Bone specimen	Portion	Ashed weight, g	Ra^{226}		Th^{228}	
				Amount $\mu\mu\text{g}$	Ratio org/inorg	Amount $\mu\mu\text{c}$	Ratio org/inorg
B-1	Femur	Organic	-	30	0.005	0.19	0.006
		Inorganic	4.15	6150		33.2	
B-1	Femur	Organic	-	22	0.004	0.15	0.006
		Inorganic	3.73	5110		25.4	

III. Normal Bone

The feasibility of analyzing normal bone for Th^{228} and Ra^{226} was further investigated using the maximum amount of bone (10g fresh weight) suitable with the present procedure for Th^{228} . Table 23 shows results on two samples received from the Naval Medical Research Center. It is evident that the concentration of Th^{228} was so small as to be hardly measurable. Assuming a uniform distribution in the skeleton, the whole-body content may be estimated to be about $10^{-11}\text{c Th}^{228}$ and $5 \times 10^{-11}\text{g Ra}^{226}$ for a skeletal equivalent of 3000 g ashed bone (standard man). If this sample is representative of the Th^{228} content of normal bone the present procedure would have to be modified to permit use of larger samples for measurement at this level. On the other hand, the procedure seems adequate for detecting increases in Th^{228} content in humans, such as might accompany intake of water from some deep wells in Illinois.⁽⁴⁾

Table 23

Ra^{228} and Ra^{226} content of a normal bone

Sample	Bone specimen	Ashed weight g	Th^{228}			Ra^{226}		
			Total, $\mu\mu\text{c}$	Blank, $\mu\mu\text{c}$	Net, $\mu\mu\text{c/g}$	Total, $\mu\mu\text{g}$	Blank, $\mu\mu\text{g}$	Net $\mu\mu\text{g/g}$
NMRC-3	Cortical tibia or femur	6.44	0.023 ± 0.012	0.005 ± 0.005	0.003 ± 0.002	0.12 ± 0.01	0.01 ± 0.01	0.017 ± 0.002
NMRC-4	Cortical tibia or femur	5.88	0.020 ± 0.012	0.005 ± 0.005	0.003 ± 0.002	0.10 ± 0.01	0.01 ± 0.01	0.014 ± 0.002

We are grateful to Commander G. W. Hyatt, Tissue Bank, National Naval Medical Center, Bethesda, Maryland, for providing the normal bone samples.

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COUNTING INDIVIDUAL ALPHA ACTIVITIES IN MIXTURES

J. Sadauskis

A reproducible method for directly determining individual alpha activities in mixtures where there are sufficient differences in alpha energies to give separation of the spectrum peaks was developed. A gridded ionization chamber and a 10-channel pulse analyzer* were used along with suitable thin source preparation. The gas filling was a mixture of 90% argon and 10% methane. Bias settings on the pulse analyzer were determined which would shift the 10-channel band to adjacent parts of the spectrum to give the desired total number of channels.

The alpha spectra of thin sources consist of a main peak, a tail which extends from the main peak to zero energy, and smaller peaks caused by decay to excited states. The smaller peaks of the excited states usually merge with the main peak; if they do not and are of sufficient magnitude, they can be handled as separate alpha energies.

The problem of counting individual alpha activities can be considered as being twofold: (1) development of a method to produce thin sources in order to concentrate as many counts as possible in the main peak and to make the main peak as narrow as possible; (2) a procedure, where overlapping of spectra occurs, to determine with sufficient accuracy the total number of counts belonging to each spectrum.

Preparation of Sources

New surfaces of stainless steel, mica, and glass, as well as cleaned surfaces of platinum, were tried as backings with the same Am^{241} solution, and all produced better spectra than those with new stainless steel or platinum discs which had not been cleaned.

In the case of platinum, two sources of Am^{241} were prepared, using new discs and with identical preparation methods, except that one disc was heated with an induction heater to a red heat and allowed to cool prior to depositing the solution. For a 10-channel region which included the spectrum peak and covered a 170-keV energy region, the unflamed disc gave 54% of the total 2π count, while the flamed disc source gave 83%; a 20-channel region for the flamed disc gave 92%. Figure 30 shows the total 50-channel plot with the flamed disc. It can be readily seen that if the first 20-channel count could be obtained directly, a 10% error in the estimation

*The chamber of modified Los Alamos design uses a pulse analyzer developed by Harold A. May, Radiological Physics Division, with an electronic circuit similar to that described by D. W. Engelkemeir and L. B. Magnusson, Rev. Sci. Instr. 26: 295-302 (1955).

Fig. 30. Spectrum of Am^{241} activity. Runs at five settings of the 10-channel band in adjacent parts of the spectrum were required to get the 50-channel combination. Channel width is 17 kev.

of the remainder would only result in about 1% error in the total count. New, flamed platinum discs were found to be the best backing of the materials tried, and combined with ordinary procedures, presented a suitable standard method of source preparation.

Single Alpha Sources

The procedure of actually making a count while keeping the gain settings constant on the pulse analyzer consisted of determining the complete spectrum to sufficient accuracy, adding up a chosen number of channels which include the peak, and multiplying this total by a previously-determined factor to get the total 2π count. A total of 50 channels was chiefly used, and the spread of 17 kev per channel used throughout. The general method for estimating the contribution from the tail or low-energy part of the spectrum where overlapping occurred into another spectrum was based on the assumption that for thin sources all spectra are sufficiently similar to permit application of the same factors or ratios within the accuracy required here.

A comparison of the distribution of counts in various portions of the spectrum was made for individual sources of Am^{241} (5.48 Mev), Pu^{239} (5.15 Mev), and Th^{230} (4.68 Mev). The results are summarized in Table 24 in which some of the data have been normalized to the counting rate obtained in a standard 2π alpha counter. A consistent value of 0.95 was obtained for the fraction of total counting rate in the 50 channels (850 kev) extending downward from the high-energy portion of the peak. The fraction obtained for 26 channels (0.92) was also consistent enough to permit use where counting time was limited.

Table 24

Distribution of counts in spectra obtained for sources of Am^{241} , Pu^{239} , and Th^{230}

Nuclide	F_{50}	F_{26}	K_{27}	K_{50}	Final slope after 50 channels (% decrease per 10 channels)
Am^{241}	0.949	0.934	728	2184	13.6
Pu^{239}	0.949	0.910	295	899	46.0
Th^{230}	0.944	0.911	359	960	19.7
Average	0.947	0.918	461	1348	26.4

$$F_{50} = \frac{\text{cpm in 50 channels}}{\text{cpm in } 2\pi \text{ counter}}$$

$$F_{26} = \frac{\text{cpm in 26 channels}}{\text{cpm in } 2\pi \text{ counter}}$$

K_{50} = Ratio of the sum of counts in channels to the right of channel containing 0.1 of the number of counts in the peak channel (cf. Fig. 30) to the number of counts in channel 50.

K_{27} is defined for channel 27 as K_{50} was defined for channel 50.

The values of K and the final slopes in Table 24 are a measure of the fraction of total activity to be found in the tails of the spectra. As expected from the smaller difference between F_{50} and F_{26} for Am^{241} than for the other two nuclides, a considerably smaller fraction of the counts is found in channel 27 or channel 50 for Am^{241} than for Pu^{239} or Th^{230} . On the other hand the number of counts per channel beyond channel 50 decreases more slowly for Am^{241} as is to be expected from the constant values of F_{50} . Thus, if one extrapolates a spectrum beyond 26 channels on the basis of average values of K_{27} , K_{50} , and slope (Table 24), errors in the region from

channels 27 to 50 tend to be cancelled by errors in the region beyond channel 50. Figure 31 shows an extrapolated slope for the Am^{241} spectrum given in the preceding figure.

Fig. 31. Am^{241} data of Fig. 30 plotted on an expanded vertical scale. Crosses at channels 27 and 50 designate the positions of estimated channel counts that were used as guides for work with multiple spectra. Channel width is 17 kev.

Multiple Alpha-Particle Sources

The procedure adopted for analysis of multiple spectra was as follows:

- (1) the total counting rate of the nuclide with the highest energy alpha-particles was determined by applying standard factors (e.g. F_{26}) to the observed counts in the region of the peak;
- (2) the spectral tail of that nuclide was extrapolated using values of K_{27} , K_{50} , and the slope;
- (3) the contribution of the first spectrum to the next peak was subtracted to provide net values of the activity of the nuclide with the next most energetic alpha particles;
- (4) the total counting rate and extrapolated tail of the second group were determined, and the process continued until the spectra were fully resolved.

The accuracy of determining individual components in a mixture of alpha activities is affected by the spread in alpha-particle energies, the relative intensities of the components, and the thickness of the source. As a means of determining how well the average factors obtained in Table 24 might be applied to multiple sources, three sources consisting of mixtures of Th^{230} (4.68 Mev), Pu^{239} (5.15 Mev), and Am^{241} (5.48 Mev) of known activity were prepared where the ratio of the activity of the components was varied only 20 or 30 percent. These were analyzed for individual activities. Thirteen total runs were taken, or 39 individual activity determinations were made. A typical three-component plot is shown in Fig. 32. The total number of counts used for each activity determination did not vary much from the average value of 18,100 counts which gives a 0.9 error of 1.3%. The percentage errors of the determined count compared to the known count was tabulated for the 39 determinations. In 19 cases the error was greater than 1.3%, and 17 of these 19 were 2.5% or less, while the remaining two were 2.8% and 3.3%. The probable error computed from the errors of the 39 determinations was 1.1%.

Fig. 32. Typical spectra obtained from source containing Th^{230} , Pu^{239} , and Am^{241} activities which were used to check reliability of method for determining individual activities. Channel width is 17 kev.

Certain cases require individual variation of the above general method. The spectrum of Ra^{226} , for example, is obtained complete for 50 channels, but superimposed on a small background from higher energy

daughters. Estimating the counting rate for Ra^{226} in channel 50 by means of K_{50} and subtracting it from the observed value at that channel gives the count from other activities at this point. A line drawn from here to where the Ra^{226} should provide zero counts to the right of channel 1 enables one to estimate the contribution from other sources to the counting rate in the 50-channel region of the Ra^{226} spectrum. The activity of a Ra^{226} solution which had been calibrated by radon measurement was checked. Three runs were taken and results gave 1.8%, 1.7%, and 1.3% lower than the radon measurement.

Applications

The ratio of Th^{228} to Th^{232} activity was checked for two solutions being used as standards for Th^{228} . One which was prepared from thorium purified sometime between 1906 and 1917 and having a calculated ratio of 0.99 ± 0.005 was analyzed, and the ratio found to be 0.99 ± 0.05 . The purification of the thorium of the second solution dated back to 1946. Its calculated ratio was 0.63 ± 0.015 . Two runs were taken on it and both gave 0.67 ± 0.03 . For both solutions the small number of total counts taken was responsible for the large possible 0.9 errors.

The ratio of Th^{232} to Th^{228} was also determined for thorium isolated from a tooth obtained from a Thorotrast patient. The thorium was extracted into a mixture of carbon tetrachloride and dibutyl phosphate and the organic phase ashed on a platinum disc. Since the counting rates were small, measurements were limited to 26 channels for each peak. The Th^{228} activity was found to be 2.0 ± 0.4 disintegrations per minute and the Th^{232} 0.6 ± 0.2 dpm. No Th^{230} was detected. Similar results were obtained on the same sample by comparison of Th^{228} measured by the dibutyl phosphate (DBP) procedure⁽¹⁾ combined with the total alpha counting rate of the thorium in the ashed DBP as measured with a 2π counter: values of 2.6 ± 0.2 dpm and 0.8 ± 0.4 dpm were obtained for Th^{228} and the Th^{232} , respectively.

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A "PANCAKE" TYPE FLOW-GAS GEIGER COUNTER

H. F. Lucas

A flow-gas counter has been constructed with a window of large diameter and small volume and "pancake" shape. Operating as a G.M. counter with a helium-isobutane gas mixture, the counter was found to have a plateau slope of less than 1%/100 volts, a background counting rate (anticoincidence) of 0.4 cpm and an efficiency for $\text{Sr}^{90}\text{-Y}^{90}$ of 0.52 ± 0.02 cpm/dpm. This is about twice the efficiency and one-third the background counting rate of a commercially available, halogen-filled "pancake" counter with nearly the same physical dimensions. Four of these flow gas counters have been combined into a plug-in assembly $3\frac{1}{2}$ " high by $6\frac{1}{2}$ " wide with at least $\frac{3}{4}$ " of steel separating each unit from the others. The efficiencies and background counting rates of the counters have been stable over the year this assembly has been in operation.

Construction

The counter was machined from stainless steel type 304 to the dimensions shown in Fig. 33. The body of the counter was cleaned with concentrated HNO_3 , distilled water and acetone after which the window was attached and the center wire inserted. The window of gold-coated Mylar*, 2.0 mg/cm² thick, was attached to the body of the counter by placing the conducting surface in contact with the open end of the counter. The ring pressed firmly into place provided a smooth, taut window while R-313 adhesive** provided gas tight seals.

The center wire was formed from straight lengths of 2.5-mil tungsten wire into loops 20 to 22 mm long as shown in Fig. 33. More recently,

Fig. 33

*Hastings and Company, Inc., 2314 Market St., Philadelphia 1, Pennsylvania.

**R-313 Adhesive. Carl H. Briggs Company, 2255 Barry Ave., Los Angeles, California.

loops 11 mm long have been tested and found to give slightly superior plateau slopes. These loops are locked by crimping the supporting tubing which does not extend beyond the end of the insulator. The center wire is inserted into the counter so that the plane of the loop is parallel to the plane of the window. Sagging of the loop by $1/16$ " doubles the plateau slope. Each counter is mounted in the sample-changing assembly shown in Fig. 34. All permanent electrical and gas connections are made at the back of the outer housing. Electrical and gas connections to the counter are made through plug-in connectors which permit quick and easy removal of any counter for cleaning or repairs. The sample to be counted is mounted on a 1.5" diameter stainless steel planchet which is then placed in the well on the sample carriage. This carriage slides on pins in the tracks which extend to the edge of the shield. Suitable stops allow the carriage to extend beyond the ends of the track, thereby completely exposing the sample well for sample changing.

Fig. 34

Four counters mounted in the above sample-changing assembly are enclosed in an iron shield with 6" walls on all sides except one which, including the door, was 4" thick. Sixteen cosmic-ray counters formed a full ring around the G.M. counters.

The signals from the 4 flow-gas Geiger counters and the cosmic-ray counters were coupled with cathode followers through 10' cables to the various scalers. A unitized system of electronics was used to eliminate as much interdependence of electronics as possible. Except for the common cosmic ray signal and high voltage supply, each counter circuit is independent of the other units and can be removed for service or interchanged without noticeable effect on the other counters. The counting gas is 98% helium and 2% isobutane, which flows through all 4 units in series at about 0.5 cc/sec.

Performance

The counting efficiency of thin 1-cm diameter Sr^{90} - Y^{90} sources mounted on stainless steel and covered with 0.6 mg/cm^2 Mylar film was $0.52 \pm 0.02 \text{ cpm/dpm}$. This efficiency was determined with a National Bureau of Standards calibrated solution which had been rechecked by the Chemical Engineering Division 4π counter and includes a dead time correction of $0.3\%/1000 \text{ cpm}$.

The effect of source displacement and asymmetry was investigated by placing a 2-mm diameter source at various distances from the center of the counter and rotating through 45 degrees of arc. Counting rates at a fixed displacement were within $\pm 1\%$ except when the source was directly under the center wire between the center of the counter and the center wire support. The effect of displacement of the source for all positions except those directly under the center wire is shown by curve A of Fig. 35. A marked reduction in counting rate for all displacements directly under the center wire is shown by curve B. This effect does not appear to cause significant errors for normal 2-cm diameter samples.

The physical dimensions and operating characteristics of the flow-gas counter and the commercially available Anton "Pancake" counter are compared in Table 25. The Anton counter was substituted with a constant source to window distance of 0.31 cm. It is seen that the efficiency of the flow gas counter is nearly twice that of the Anton counter, while the background counting rate is only about one-third. The lower efficiency of the Anton counter may be caused by the post in the center of the counter.

The flow-gas and halogen-filled counters have unshielded background counting rates of about 20 and 10 cpm respectively, which are reduced to about 10 and 6 cpm respectively by the 6" iron shield. As shown in Table 25, the anticoincidence background counting rate for the flow gas counter was 0.4 cpm and for several Anton counters varied from 1 to 1.5 cpm.

A. SOURCE DISPLACEMENT FOR ALL POSITIONS NOT DIRECTLY UNDER CENTER WIRE

B. SOURCE DISPLACEMENT FOR ALL POSITIONS DIRECTLY UNDER CENTER WIRE

Fig. 35

Table 25

Comparison of "pancake" counters

	Flow-gas	Anton "pancake" (1)
Window diameter, cm	3.33	2.86
Window thickness, mg/cm ²	2.0	1.4 - 2.0
Operation potential, volts	1425	700
Plateau slope %/100 volts	1	15
Background count rate (cpm) (B)	0.4	1.0 - 1.5
Efficiency, Sr ⁹⁰ -Y ⁹⁰ , cpm/dpm (E)	0.52 ± .02	0.22 ± .01
Figure of merit, E ² /B	0.69	0.03

(1) Cat. No. 1007-T

Anton Electronics Laboratory, Inc.

1226 Flushing Ave., Brooklyn, N. Y.

Using as a criteria the conventional efficiency squared over background (E^2/B) as a figure of merit, one obtains for the flow-gas and Anton counters, 0.69 and 0.03, respectively, i.e., the flow-gas counter has 20 times the figure of merit of the Anton counter. The excellent plateau slope, high efficiency and low background counting rate, when combined with its small size and ready adaptation to a close packed counting assembly, make this flow-gas counter particularly desirable for very low level work.

Special thanks are due Mr. O. J. Steingraber for the design and construction of the electronic equipment. Thanks are also due Mr. Peter Kafalas of the Chemical Engineering Division for absolute determination of the Sr^{90} standard by 4π counting.

FURTHER STUDIES ON THE NATURAL RADIOACTIVITY OF MUNICIPAL WATER SUPPLIES

H. F. Lucas

Measurements of the naturally occurring concentration of Ra^{226} in municipal water supplies have been continued.^(1,2) As summarized in Tables 26 and 27, no unusually high radium concentrations were found.

Table 26

Summary: Ra^{226} content of municipal water supplies

Municipality	Water type ⁽¹⁾	Well No.	Ra^{226} content, $\mu\mu\text{g}/1 \pm E$ ⁽²⁾
Spain			
Barcelona	R + T	(3)	$0.20 \pm .01$
Madrid	R	(3)	$0.02 \pm .01$
Iowa			
Cedar Falls	(3)	(3)	0.78
Iowa City	(3)	(3)	0.19
Illinois			
De Kalb	R	6	4.92
De Kalb	R	7	3.16
Des Plaines	R	1	2.71
Des Plaines	R	2	3.14
Des Plaines	R	3	2.52
Des Plaines	T	1-3	0.59
Downers Grove	R	(3)	0.42
Downers Grove	T	(3)	0.04

(1) Raw (R) or treated (T) to remove hardness.

(2) Error $\pm 2\%$ or $\pm 0.03 \mu\mu\text{g}/1$, whichever is larger, except as noted.

(3) Not known.

Rn^{222} concentrations were reported⁽³⁾ in well waters at Barcelona ranging from 60 to 480 $\mu\mu\text{c}/1$ with Ra^{226} content not determined. Dr. Javier Buñuel at Madrid, kindly obtained a 2-gallon sample of water from the municipal pumping stations at Barcelona and at Madrid. The water sample from Barcelona consisted of 70% raw well water and 30% treated surface

Table 27

Radioactivity of West Chicago well waters

Water source	Ra ²²⁶ , $\mu\mu\text{g/l} \pm E(1)$	Gross alpha activity, $\mu\mu\text{c/l} \pm E(1)$	Gross beta activity, $\mu\mu\text{c/l} \pm E(1)$
Municipal			
No. 1	$0.78 \pm .04$	*	*
No. 2	$0.53 \pm .04$	$1.2 \pm .4$	$3.7 \pm .5$
No. 3	$0.84 \pm .04$	*	*
Industrial			
Northwest Chemical	$1.26 \pm .09$	*	*
Molded Products	$0.80 \pm .04$	$1.1 \pm .3$	$7.0 \pm .5$
Linde Chemical	$0.78 \pm .04$	$2.0 \pm .4$	$9.4 \pm .5$
Private	$0.54 \pm .04$	$2 \pm 5 (2)$	$12 \pm 2 (2)$

(1) 0.9 statistical counting error.

(2) Residue from 300 ml only was mounted and counted for alpha and beta activity.

*Not determined.

water of the Rio Llobregat, while the Madrid sample consisted of untreated water of the Rio Lozoya. The Ra²²⁶ content of a 0.4- and a 2.0-liter portion of the Barcelona sample was $0.20 \pm .01 \mu\mu\text{g/l}$. Similar portions of the sample from Madrid were found to contain $0.02 \pm .01 \mu\mu\text{g/l}$.

In order to assess the effect of water softeners on the accumulation of radium by the populations of interest, and since previous comparisons did not provide clear-cut conclusions, an additional study has been undertaken.⁽¹⁾ In this study, a home water softener at Downers Grove having a 20,000 grain capacity Dowex 50 ion exchanger was found to remove 98% of the calcium and magnesium while removing about $90 \pm 8\%$ of the radium. Similarly, at Des Plaines, 80% of the Ra²²⁶ was removed by the conventional lime floc water softening technique. Thus, it now appears that the use of softening equipment must be considered as an important factor in evaluating the actual intake of radium by humans.

The Illinois Department of Health reported concentrations as high as 4×10^{-10} c/l. of beta in water samples from wells in West Chicago, Illinois. Because of the possibility that this was a new area of high natural radioactivity, samples were obtained from seven of the known well sources.

The Ra^{226} , gross alpha and gross beta contents were measured and are summarized in Table 27. As expected, no unusual radioactivity was detected with radium contents from 0.5 to 1.3 $\mu\mu\text{g/l.}$, all of which fall within the range previously noted for wells penetrating limestone formations.⁽¹⁾

Portions of four of the water samples were prepared for gross alpha and beta counting as follows: one-liter portions were evaporated to a small volume, transferred to a weighed porcelain crucible, and then evaporated to dryness. After adding two 10-ml portions of concentrated HCl, the dry residue was flamed at red heat for 1 minute, scraped from the crucible, crushed into a fine powder, and then transferred to a weighed 5-cm diameter stainless steel planchet.

These samples were then counted in a Nuclear Measurements Corporation PC-2 windowless alpha proportional counter having a background counting rate of 0.1 cpm. Successive 900-minute counts agreed within statistical counting errors. Corrections for self-absorption were based on curves derived for this counter with Pu^{239} in a light inorganic medium.⁽⁴⁾ The total alpha activity obtained by this technique is consistent with the Ra^{226} plus daughters present 1 to 10 days after sample preparation.

The gross beta activity was determined by counting each of the samples in a Geiger counter having an anticoincidence counting rate of 3.8 cpm, and an efficiency for a thin distributed source of about 0.83 cpm/ $\mu\mu\text{c.}$ Corrections for self-absorption were made using the added absorber technique of Aten.⁽⁵⁾ Self-absorption factors were also calculated assuming potassium as the only activity and were found to agree with those obtained by the Aten method. Since the beta activity is also within the range of that expected from the 1 -16 mg/l. of potassium normally found in potable waters of Illinois⁽⁶⁾ it is believed that the beta activity observed results from the natural potassium content.

Thanks are due R. W. Bane for analyses of Ca and Mg in the water sample from Downers Grove. Thanks are also due Dr. Howard Kraemers for assistance in collecting the West Chicago water samples, and to Mr. F. H. Ilcewicz for performing the Ra^{226} determinations

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MESOTHORIUM MEASUREMENTS ON SOME RADIUM-BEARING
POTABLE WATERS

D. P. Krause

The dibutyl phosphate (DBP) method for the extraction and determination of Ac^{228} briefly described in the previous semiannual report,⁽¹⁾ has been further tested and applied to a number of water samples of interest as a means of determining the concentration of mesothorium (Ra^{228}). Comparison is made with the Ra^{226} values obtained on the same samples.

In order to establish the method on a quantitative basis, the procedure for Ac^{228} was applied to a standard solution prepared from pure thorium which had been separated from its daughters and other impurities in June 1946. The solution contained 4.47 mg thorium per milliliter. Calculation of the Ra^{228} activity which had grown in since purification indicated that the solution should have 796 disintegrations of Ra^{228} per minute per milliliter as of December 1, 1958. The Ac^{228} daughter is in equilibrium with the Ra^{228} and therefore will be present in like amount.

The procedure used in the extraction of Ac^{228} follows: The sample, 25 ml in volume and 0.1 N in HNO_3 , is contacted three times with one-ml portions of DBP in CCl_4 (25% DBP). The combined organic phase containing the actinium is scrubbed with 0.05 N HNO_3 . Then the actinium is transported back into an aqueous phase by stripping with 4.0 N HNO_3 , which leaves thorium in the organic phase. The aqueous solution so obtained is contacted with 1 ml of DBP, and then three times with CCl_4 to remove any entrained DBP. The solution is then evaporated to dryness. One ml of 0.1 N HNO_3 and 2 milligrams of Pb^{++} are added, and this solution is saturated with H_2S to precipitate PbS so as to remove Pb, Po and Bi activities. The precipitate is separated by centrifugation, and the supernatant liquid containing the actinium is evaporated to dryness on a stainless steel planchet. The precipitate is washed once with 1 ml of 0.1 N HNO_3 saturated with H_2S and the wash added to the planchet. The planchet is then flamed, covered with Mylar film and counted.

The counting equipment used was that described by Lucas.⁽²⁾ Calculations of concentrations of Ra^{228} were based on his calibration of the equipment with a Bureau of Standards Sr^{90} - Y^{90} solution.

Since it was expected that the Ra^{228} content of the water samples would be low, i.e., in the order of magnitude of the Ra^{226} content, one-liter samples were taken for analysis. Simple evaporation of a large sample to the 25-ml volume used in the extraction was impractical because of unknown and possibly high salt concentration. A procedure based on the co-precipitation of radium with barium sulfate was used to concentrate the radium and obtain the desired volume.

A one-liter sample to which 20 mg of Ba^{++} was added was reduced in volume to 500 ml, and barium sulphate was precipitated with ammonium sulfate at pH 1 - 1.5. The precipitate was digested overnight at room temperature, and the barium sulfate was separated by filtration. After careful washing of the precipitate and paper and ignition in a platinum crucible, the barium sulfate was fused with 1 gram of the Na_2CO_3 for one hour to convert to acid-soluble barium carbonate. The fused mass was dissolved in distilled water and the insoluble barium carbonate filtered. The barium carbonate was dissolved in 4.0 N HNO_3 , the solution was evaporated to dryness, and the resulting residue was taken up in 25 ml of 0.1 N HNO_3 . After a suitable time of standing to allow for growth of the actinium daughter, the Ac^{228} was extracted as described above.

The thorium standard was analyzed in each of two ways. In the first case, the DBP extraction was made directly on a 1-ml aliquot diluted to 25 ml in 0.1 N HNO_3 . In the second case, the extraction was preceded by a BaSO_4 precipitation of the radium as described above.

As was expected, in each case when the counting data on the final product was plotted, a curve with the 6.1-hour decay characteristic of Ac^{228} was obtained; but the shape of the curves differed after about 5 half-lives. When no BaSO_4 separation was included, the decay tailed off into a component of an approximately 3.6-day half-life, indicating the presence of Ra^{224} . This portion of the decay curve extrapolated back to zero time of separation yielded a count of about 10 cpm compared to 400 cpm for Ac^{228} . From a consideration of the analytical procedure and the amount of thorium originally present, one would expect to find a small amount of Ra^{224} (and daughter) activity present because of radioactive growth from Th^{228} .

However, in the case where a BaSO_4 precipitation was included one would not expect to see this 3.6-day tail on the decay curve because the thorium is separated at the precipitation stage. Actually, this is what was observed. The decay curve in this case tailed off to a constant value with no indication of the presence of a longer-lived component.

In addition, one other difference was noted between the samples analyzed by the two methods. After subtraction of the appropriate tail or background each method gave a curve with the 6.1-hour decay through ca. 8 to 10 half-lives but differed in the value of the count when extrapolated back to the time of separation of the Ac^{228} from the parent Ra^{228} as shown in Table 28.

Table 28

I Ac^{228} , disintegrations per minute (calculated)	II Ac^{228} , counts per minute (No BaSO_4)	III Ac^{228} , counts per minute (BaSO_4 pptn)	IV Counter efficiency II/I cpm/dpm	V Counter efficiency (Lucas) cpm/dpm
796	400	330	0.50	$0.52 \pm .02$

Using the Lucas value of 0.52 as the efficiency of the counter, a recovery of ca. 96% of the Ac^{228} is indicated for the case where no BaSO_4 precipitation is made, and a recovery of ca. 80% of the Ac^{228} is indicated when the BaSO_4 precipitation is included.

Table 29

Place	$\text{Ra}^{228} (\text{Ac}^{228}), *$ $\mu\text{c/liter}$ (Estimated error $\pm 10\%$)	Ra^{226} $\mu\text{g/liter}$
Toulon	1.3	37.2 ± 0.3 (3)
Biggsville	2.4	16.0 ± 0.2 (3)
Bushnell	3.4	18.7 ± 0.2 (3)
Joliet	7.0	6.9 ± 0.2 (5)
Lockport	7.9	11.5 ± 0.3 (5)
Lostant	6.5	10.1 ± 0.3 (4)
Ottawa	3.8	6.5 ± 0.2 (4)

*These values include a correction factor of 0.8 for the total radium recovery, based on analysis for Ra^{226} in the aqueous raffinate after extraction of the Ac^{228} .

This value of approximately 80% recovery has been verified by Ra^{226} analysis of the water samples listed in Table 29. Analyses for Ra^{226} were made on the intact samples and on the solution from which Ac^{228} was extracted after precipitation of BaSO_4 and fusion with Na_2CO_3 .

The complete procedure has been applied to several Illinois water samples from deep sandstone wells.

The results obtained are tabulated in Table 29. For comparison the values obtained by Lucas and Ilcewicz^(3,4,5) for the Ra^{226} content of these waters are also shown.

Although several of the samples contained Ra^{228} in amounts comparable to those of Ra^{226} , it is evident that the concentration of Ra^{226} was no indication of the amount of Ra^{228} to be expected. It will be of interest to measure the Ra^{228} content of water from other types of sources.

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THE HUMAN SPECTROMETER

Introduction

C. E. Miller

The following report, although incomplete, is nevertheless presented in its present form in order to obtain early publication of results of value to the many interested in this particular field of endeavor.

Most of the studies are related to the measurement of radium and are reported within three categories. (1) improving the sensitivity of radium measurements in humans with the ultimate goal of measuring the radium content of normal unexposed humans, (2) perfecting techniques for measuring the $\text{Ra}^{226}/\text{RaB}$ ratio present in intact humans which will then permit a more accurate dose determination, and (3) measuring of individuals whose exposure to radium dates back from 30 to 40 years ago.

It has been essential to determine the radioactive content of certain bulk samples of material which were used in the construction of equipment used within the Iron Room. These included samples of steel, glass and MgO which were used in the canning of the crystals for Iron Room II as well as the polishing compounds which were used to prepare the crystal. Various welding rods have also been checked which were used in the construction of collimating shields.

A number of old radium-bearing nostrums - loaned by Dr. Richard Chamberlain, University of Pennsylvania - have been analyzed to determine the amount of mesothorium present with the radium

I. Coincidence Summing Technique

C. E. Miller and O. J. Steingraber

A coincidence-summing technique designed to measure small quantities of radium when mixed with relatively large quantities of other gamma-ray emitters was discussed in the last semiannual report (1) This preliminary approach utilized two NaI(Tl) crystals - one $2\frac{1}{2}$ inches in diameter by $2\frac{1}{8}$ inches thick and one 4 inches in diameter by $1\frac{1}{2}$ inches thick. Recently, the loan of a canned 8-inch by 4-inch NaI(Tl) crystal, through the courtesy of E. C. Stewart of the Harshaw Chemical Company, permitted a re-evaluation of this technique using crystals of this size. At the same time a plastic scintillation counter 24 inches by 4 inches became available as a cosmic-ray anticoincidence detector.

Fig. 36

In brief, the electronic equipment, with the addition of a cosmic-ray anticoincidence detector, is shown in block form in Fig. 36. The pulses from the two NaI(Tl) crystals are fed to a summing network (Σ) and the sum pulse applied to the input of the 256-channel analyzer. In addition, the individual pulses from each crystal are amplified and analyzed by the wide-band single-channel pulse-height analyzers Nos. 1 and 2. The output pulses of these two single-channel analyzers are applied to the coincidence circuit. The coincidence circuit in turn gates on the 256-channel analyzer and permits it to sort the sum pulse whenever pulses are received simultaneously from both single-channel analyzers (No. 1 and No. 2). An output pulse from single-channel analyzer No. 3 - the plastic cosmic-ray counter - which is in coincidence (timewise) with the outputs of analyzers No. 1 and No. 2 cancels the coincidence output, and the sum pulse is not sorted.

The lower- and upper-level discriminators of the single channel analyzers, connected to the NaI(Tl) crystals, can be adjusted independently to span any desired energy band. The lower-level discriminator was set at 500 kev in order to reject all pulses due to gamma rays which might back-scatter from one crystal to the other crystal and thus produce a false coincidence. The upper-level discriminator was set at 1.25 Mev for most of the tests since the cascade gamma rays emitted by RaC' fall within this range. However, an energy band of 0.5 Mev to 3 Mev is a more feasible band as will be shown later.

The data from the various runs, using a lower limit of 0.5 Mev, are summarized in Table 30. The integral counts per minute in the energy band from 1 Mev to 1.9 Mev are given. This energy span was selected since it provided the highest signal-to-background ratio when RaC' was measured.

The gamma-ray background was measured with the crystal faces 1 cm, 10 cm, 12 cm, and 15 cm apart and found to be 8.5 c/m, 5 c/m, 4.4 c/m and 4.3 c/m, respectively. This reduction in background as the distance between the crystals was increased suggested that radioactivity

Table 30

Crystal spacing in cm	Coin. sum bkgd., cpm	Bkgd. with cosmic ray anticoin., cpm	Bkgd. with 2 inches of Bi bricks between crystals, cpm	0.2 μ c of radium between crystals, cpm	0.88 $\times 10^{-8}$ c of Ra in leg-equivalent phantom, cpm	0.2 μ c of Thorotrast between crystals, cpm	μ c of Ra to yield counting rate equal to bkgd.	μ c of Ra to yield 1 c/m	Standard error for 100 min-run, μ c of Ra
1	8.5								
10	5	3.5	1.7	4468			220	44	10
12	4.4			3400*	151	1742	260	60	12
15	4.3			2329			360	84	18

*Expected counting rate from 0.2 μ c Ra based on the counting rate from 0.88 $\times 10^{-8}$ source.

was present in the detectors themselves. This premise was supported by the fact that placing radioactively clean 2-inch thick bismuth bricks between the crystals reduced the counting rate (10 cm spacing) from 5 c/m to 1.7 c/m. The sum spectrum obtained when the crystals were 1 cm apart displayed small photopeaks due to radium and a large broad peak around 1.5 Mev suggestive of K^{40} . This element, although a monoenergetic emitter, may account for this peak because the gamma rays, emitted by the K in the photomultiplier glass envelope, can scatter forward in one crystal through an angle not greater than 75 degrees and then be absorbed in the second crystal. The one gamma ray will lose at least 500 kev of energy in each crystal and produce a coincident pulse which will be sorted and stored. The Ra^{226} present in the glass of the photomultiplier or the radon present in the room air may account for the radium component in background.

The use of the plastic anticoincidence detector reduced the background from 5 c/m to 3.5 c/m or a reduction of 1.5 c/m. Unfortunately, due to the limited time available and the large number of different tests which were made on the crystal, a run was not made with the cosmic-ray detector and the bismuth bricks both present. It is possible that the background might have been reduced considerably.

A 0.2- μ c National Bureau of Standards calibrated radium source was placed on the axis midway between the crystals. These data are given in column 5, Table 30. The amount of Ra^{226} , expressed in $\mu\mu$ c, which would yield a counting rate equivalent to background, is given in column 8 and the uncertainty in a 100-minute run, expressed as $\mu\mu$ c of Ra^{226} , is given in column 10.

Only a few runs were made utilizing all pulses above 0.5 Mev. The background counting rate and the radium counting rate in the 1 Mev to 1.9 Mev band did not increase if the single channel analyzers (connected to

Fig. 37

Uses of the Coincidence System

A possible application of this technique would be the measurement of radium and thorium present in normal bone; a 100-gram sample of bone (1/70th of the skeleton) would be a practical size sample to measure using the large crystals. Working at 1 c/m (or $\pm 10\%$ standard error) and employing a hundred-minute counting period, bone could be measured from an individual who had a body content of 3.5×10^{-9} c of radium. Dr. Andrew Stehney is studying one group of normal unexposed individuals who contain approximately this amount. The bone would not require any preparation other than perhaps ashing overnight, so the bone would retain all of the radon, and a one-month delay to permit the radon daughters to grow into equilibrium.

An evaluation measurement was made on the shin of subject G, whose burden is discussed elsewhere in this report. The subject's shin was placed between the crystals which were spaced 10 cm apart. A net increase of 1.6 c/m was obtained, which is equal to 98 $\mu\mu\text{c}$ of Ra^{226} .

the NaI(Tl) crystals) were set to accept pulses from 0.5 Mev to infinity. This setting, however, did yield a sum spectrum for Thorotrast which extends up to and peaks at 3.2 Mev. (See Fig. 37). The background counting rate from 2.6 to 3.5 Mev is 3 counts per minute. A 0.2- μc thorium source yields a counting rate of 1980 c/m in this same band. This background is equivalent to 300 $\mu\mu\text{c}$ of Th^{232} in equilibrium with its daughters and a 1 c/m increase is equal to 100 $\mu\mu\text{c}$ of thorium.

The 0.2 μc of Th^{232} yielded 2125 c/m in the 1.0 to 1.9 Mev band used for Ra^{226} . Thus this latter arrangement is the logical band to use when analyzing any sample since an analysis of the higher band will disclose the presence of Th^{232} , and a false analysis will not result.

Assuming that the 8-inch diameter crystals viewed $2/3$ of the fibula and tibia, which represent 6.6% of the total skeleton, then about 4.4% of the skeleton contains $98 \mu\mu\text{c}$ of RaC. This gives a value of $98 \mu\mu\text{c}/0.044$ or $2.2 \times 10^{-9}\text{c}$ of RaC' or very close to the value found by total-body measurements on the same patient.

These calculations are obviously very crude but do indicate that the technique shows promise; improvements beyond the massive increase of NaI crystals are under consideration.

II Radium Body Content of a Ra^{226} Inhalation Case 2300 Days after Exposure

C. E. Miller and J. B. Corcoran

This report concerns technician G who inhaled Ra^{226} in 1952 and whose radium content has been reported both in the open literature and in some of our previous reports.^(2,3) Because of the much higher activity of fallout Cs^{137} and natural K^{40} in the body, the "natural" gamma-ray activity of this individual overshadows the gamma rays of radium below an energy of 1.55 Mev. As mentioned before⁽⁴⁾ a radium determination using the 0 to 1.55 Mev band would have an uncertainty of at least 3×10^{-9} curie. An attempt was made, therefore, to measure the radium present by using only those gamma rays of energy from 1.55 Mev to 3.15 Mev.

Although no natural radionuclide which emits gamma rays in this energy band is present in the body, the gamma-ray spectrum of the body of an uncontaminated subject does yield counts in this band. It was reported previously⁽⁵⁾ that placing tanks of distilled water in the position occupied by the body increased the gamma-ray background in this band by about 5 counts per minute. This effect could be circumvented, however, by using the difference in spectra from subject G (5 measurements) and the spectra of a group of four unexposed individuals of the same musculature.

Table 31

Length of run, min	Background, c/m	Control subject, net c/m	Subject G, net c/m
100	65.1 ± 0.9		9 ± 1.25
50	62.4 ± 1.1	6.5 ± 1.6	11.4 ± 1.6
100	60.0 ± 0.8	4.5 ± 1.1	10.6 ± 1.1
100	59.7 ± 0.8	7.6 ± 1.1	11.0 ± 1.1
100	61.7 ± 0.8	6 ± 1.1	10.9 ± 1.2
	Average	6.1 ± 0.6	10.4 ± 0.7

The counting rate in the energy band of 1.55 Mev to 3.15 Mev of background, of subject G, and of the control subjects are given in Table 31.

Subject G was measured for a total of 450 minutes and the control subjects were measured for 100 minutes each, except for the first subject who was measured

for 50 minutes. The net weighted average value for the four control subjects is 6.1 ± 0.6 c/m higher than the background obtained without anything present in the chair. Subject G yielded an average value of 10.4 ± 0.7 counts per minute or 4.2 ± 0.9 c/m in excess of the control subjects. An evaluation of the data demonstrated that all points were valid and fell within the expected statistics.

Data obtained from several subjects who contained large quantities of radium demonstrated that 2,200 c/m would be expected in this band from one microcurie of RaB and RaC'. Thus this subject contains $2 \pm 0.4 \times 10^{-9}$ curie of RaB and RaC'.*

A single 100-minute measurement on a subject under investigation and on a control subject of the same body build would yield a value whose standard uncertainty would be of the order of 10^{-9} c of RaC'. This is equivalent of a body content, assuming 31% retention of radon, of about 3×10^{-9} curie of Ra²²⁶ or equal to that realized using only a 50-minute run and an analysis of the lower energy band.⁽⁴⁾ Thus higher sensitivities are not realized at present by using this higher energy band; the advantage of the technique is simply a practical one; namely, that of obviating the precise determination of K⁴⁰ and Cs¹³⁷ in this subject by the administration of K⁴² and Cs¹³⁷ for purposes of obtaining what his spectrum would be in the absence of Ra²²⁶. It remains to be seen if cosmic-ray anticoincidence techniques would improve the measurement by substantially reducing the background of about 60 c/m in this region.

III. Cs¹³⁷ Trend in Humans

C. E. Miller

The Cs¹³⁷ content of each of the 13 control subjects was measured in November 1958 and found in general to be about the same as those values found during July 1958.⁽⁶⁾ One of the members of the group, CEM, usually a heavy coffee drinker, stopped drinking coffee and tea about October 1, 1958, and has drunk about 8 glasses of milk per day since. CEM's Cs¹³⁷/g of K level has increased from $43 \mu\mu\text{c}$ on July 1958 to $58 \mu\mu\text{c}$ on November 22, 1958, and to $66 \mu\mu\text{c}$ on December 31, 1958. This increase reflects the increase in milk consumption in the diet.

*An independent check by a coincidence summing-up technique over the lower leg yields contents in agreement with this figure. Both are also consistent with the rate of Rn exhalation by the subject.

IV. Atmospheric Content of Cs¹³⁷ as a Function of Time

C. E. Miller

The shape of the curve indicating the amounts of Cs¹³⁷ in subjects of the Chicago area suggested that either a large quantity of Cs¹³⁷ had been suddenly injected in the biosphere some time in 1954 or that the average fallout rate had increased by a factor of about 3 during most of 1954 and had remained constant thereafter. Since Cs¹³⁷ has a relatively short biological half-time in the human body (~ 150 days), the amount of Cs¹³⁷ present in the body is indicative of the amount of Cs¹³⁷ ingested during the preceding few months. The Cs¹³⁷ content of the average human was about $10 \mu\mu\text{c Cs}^{137}/\text{g K}$ during March 1955, and increased at essentially a uniform rate until it reached $35 \mu\mu\text{c Cs}^{137}/\text{g K}$ in May 1956 and remained constant at this value until the spring of 1958 when a second rise was detected. In order to learn more about this type of fallout in the same intervening period, the Cs¹³⁷ atmospheric contents were measured by utilizing air filters collected at the Argonne National Laboratory for the past several years. Air pumps located at several different sites at Argonne pull approximately 15 m^3 of atmospheric air per hour through HV-70 filter papers. The filters in some air pumps are changed daily and the filters in six are changed weekly. A 20-cm^2 disk is cut out of each filter and a gross beta count made at the time of collection. The rest of the filter paper is then stored for future reference.

The weekly filter papers from March 1953 until the present were bunched into monthly groups and the gamma-ray spectrum of each monthly group obtained with the 8-inch by 4-inch NaI(Tl) crystal. The papers collected from March 1953 until June 1956 yielded, for all practical purposes, a pure Cs¹³⁷ spectrum. Thus the amount of activity present was determined by comparing the counting rate under the Cs¹³⁷ photopeak from the paper to that obtained from a known quantity of Cs¹³⁷ distributed over a blank paper. The Cs¹³⁷ gamma rays present in those filters collected more recently were obscured by the gamma rays emitted by short-lived fission products which had not decayed.

The filter papers collected during 1956 and 1957 were analyzed using the IBM 650 computer and a program which had been written by Mr. John C. Reynolds. Individual gamma-ray spectra were obtained for each fission product which was suspected to be present. The computer automatically subtracted the appropriate quantity of each reference spectrum from the unknown spectrum which would result in the smallest least-squares error spectrum. The program yielded very good results when tested by analyzing spectra which had been compiled by adding various amounts of the reference spectra. Naturally, a reference spectrum of each element present on the unknown must be fed to the computer.

However, the filter papers collected during 1958 have not been analyzed successfully by the computer technique because these filters contain several activation products, of which only three have been identified; i.e., Co^{60} , Sb^{125} and Mn^{54} . Chemical separations are in progress under the direction of Dr. Jacob Sedlet on the papers collected during one month to identify the remaining activation isotopes whose spectra must be added to the reference stack.

Fig. 38

USA, UK and USSR(7) are shown. The hydrogen tests are represented by vertical lines. The UK and USSR atomic tests are shown as dots and the USA series are shown by a horizontal bar which spans the complete test period. The dates of USSR tests are very incomplete.

Notice the correlation between the hydrogen tests during March 1954, October 1955, May 1956 and August 1956, and the corresponding excursions in airborne Cs^{137} activity the following months. Evidently unreported sizeable tests took place in July 1956 and January 1957. Although an analyses of the weekly papers would yield more accurate dates of these tests, the weekly groups have not been measured since the aim of this investigation was to determine if a correlation existed between airborne levels and human values.

The dpm in papers collected per month were averaged for each year. These values are listed in Table 32. Although the monthly values vary over wide limits during any one-year period, the yearly average airborne Cs^{137} values do show the same general trend as the Cs^{137} trend in humans, except for a time delay which would be expected to represent an average time of transit of Cs^{137} from air to soil and thence through food to man.

The amount of Cs^{137} (expressed as disintegrations per minute (dpm) per paper) as a function of time is given by Fig. 38. This quantity of Cs was filtered out of about 2000 m^3 of air. The absolute activity per cubic meter of air has not been calculated since the efficiency of the filter to airborne Cs is not available.

On the time abscissae of the figure, the dates of fusion and fission tests which have been published for the

Table 32

Year	Average monthly value (cpm/paper)	Year	Average monthly value (cpm/paper)
1953	16	1956	81
1954	29	1957	75
1955	74		

V. Radioactivity of Welding Electrodes

J. B. Corcoran and C. E. Miller

The activity in welding electrodes has been further studied. Some further measurements on welding electrodes may be of value to persons concerned with the construction of iron rooms.

Previous measurements⁽⁸⁾ reporting the gross activity in various types of electrodes were made by counting the whole, intact rod. To evaluate welding electrodes better for the construction of iron rooms, it is necessary to learn, besides their total activity, whether the latter is entirely in the flux, and how much actually stays with the weld melt after welding.

Three types of electrode were selected for study. Type Jet No. 1,* and LH-70,** were heavy construction types with very thick coats of flux. Type 6010,*** a general purpose electrode, bears the lowest gross activity of the types previously reported.⁽⁸⁾

The bare welding wire stripped of flux was counted and no activity found, thus confirming the hypothesis that the flux contains all of the activity.

The three types of flux were then counted and compared as follows:

Table 33

Type	Weight of flux per electrode, g	Approximate gross gamma activity per electrode,* $\mu\mu\text{c}$	Comparison with Type 6010		
			Gross activity per electrode	Radium and thorium per gram of flux	K ⁴⁰ per gram of flux
6010	4.4	7	1	1	1
LH-70	18.0	56	8	1.7	4.2
Jet 1	84.5	294	42	2.4	3.3

*gamma activity equivalent to radium in equilibrium

*A. W. S. - A. S. T. M. Type E 6024, Lincoln Electric Co., Jetweld No. 1.

**A. W. S. - A. S. T. M. Type E 7016, Lincoln Electric Co., LH-70.

***A. W. S. - A. S. T. M. Type E 6010, Metal and Thermite Co., Type R.

We obtained these samples by courtesy of the Nashville Bridge Company, Nashville, Tennessee.

A more detailed study was made of the Jet No. 1 flux with the following results:

Jet No. 1	1.8×10^{-10} c/electrode radium B + C
	3.8×10^{-11} c/electrode thorium
	1.9 g/electrode potassium

To investigate residual activity after welding, a whole, intact Jet No. 1 electrode was counted over an activity-free steel plate. The shop welder then welded a bead the length of the plate, chipped off the flux, and carefully wire brushed the bead. Three days later the plate was counted and 14% of the original activity of the rod was found to be still present. The procedure was repeated with the welder using the poorest technique that he could and trying to trap additional flux in the weld melt. When this second plate was counted three days later, however, the same 14% residual activity was again present.

The above clearly indicates that welding rods will invariably deposit activity when used on the inner surfaces of an iron room. A careful selection of type, however, can reduce this to a minimum.

VI. Measurement of Activity in Radium Nostrums

C. E. Miller and J. B. Corcoran

As has been reported in previous semiannual reports the long-term effects of bone-seeking radioisotopes is under investigation by studying individuals who accumulated body burdens of radium many years ago. The individuals studied to date received the material either therapeutically or by industrial exposure to radium compounds. In addition to those persons who accumulated radium from industrial or clinical sources many years ago, there exists a possible third group of people who may have accumulated radium from the home use of certain nostrums.

Several nostrums which used radium as the medicinal agent were available to the general public many years ago. The advisability of attempting to locate individuals who used these preparations depends in part on the half-life of the radioelements used and the amounts involved. In any case, the long-term clinical effects that would be observed in these individuals cannot be correlated to their present body contents until the percentage of mesothorium (Ra^{228}) present in the original radium preparations has been determined. Through the cooperation of Dr. R. J. Hasterlik of the Argonne Cancer Research Hospital samples of six radium nostrums, loaned by Dr. R. Chamberlain, University Hospital, Philadelphia, Pennsylvania, were obtained and analyzed with the gamma-ray spectrometer in order to determine the amount of radium and ThC' (Pb^{214}) present.

Caution was necessary in handling these sources to avoid contamination of the low background facilities. Even though the quantities present do not represent a health hazard, they contain 1000 times the activity present in some sources which are analyzed with this equipment. Since the containers as received were contaminated on the outside, the containers were heat-sealed in plastic bags. Consequently, the quantity present was estimated either by visual inspection of their labels or by weighing the container plus contents.

The trade name of each nostrum and all information printed on the container is given, followed by a short discussion of the approximate amount, activity, and in some cases, the probability of how much the subject might have accumulated by use of an appreciable amount.

Sample 1 - Radioactive Earth

Method of Employing Radioactive Earth: The earth must be used under supervision by the physician who will regulate dosage.

1. Direct Application: Mix the earth with boiling water to the consistency of an ointment, having carefully cleansed the affected part. Cover it with a layer of this, about a piece of jaconette overlapping the earth at least an inch all around and over this place a layer of cotton wool--the whole being retained in position by a bandage. If the skin is delicate or easily irritated, a poultice may be made by enclosing the layer of earth between two pieces of white lint. Application being made as before. The time of application varies from 3 hours to several days: If irritation is caused a cessation of the treatment for one or two days with an application of oxide of zinc is advised. In a protracted treatment the compress should be remade frequently.

Care must be taken that the compress is not too tight; neither should the earth be allowed to become desiccated or maceration and irritation of the skin may result.

Precaution: If the affect part is also ulcerated or the skin has an abrasion sterilized earth must be used and aseptic compresses made.

2. Application in Baths: From 1/2 to 2 lbs. of earth may be used in a bath of about 40 gallons in time 20 minutes to a half hour. Temperature 92-100°F. Massage and rinse on leaving bath: Arm and foot baths may be conveniently used.

3. Gynecological Injections: Injections may be made with water as hot as is bearable containing sterilized earth in suspense. The dosage must be fixed by a physician.

4. Application by Ionization: This method with continuous electric current has been up to the present time proved by far the most efficacious. The affected part is covered with a layer of earth about 1/4 inch in thickness; on this is placed a tin plate or other electrode which is connected to the positive pole of the source of electricity. The pole beam connected preferably to the spinal column. Application may be continued for 30-60 minutes. With a current of 5 to 30 milliamperes. It is essential that the earth should be sufficiently moistened, for which purpose radioactive water may with advantage be employed.

For thickened joints, rheumatic gouty, for arthritic inflammations and the treatment of certain skin diseases. Also for painful nerve and muscular conditions such as neuritis, sciatica, neuralgia, lumbago, shingles and intercaustal rheumatism.

The total weight of the box plus contents was $3\frac{1}{4}$ pounds. Contents estimated at 3 pounds. About 1.1 microcuries of Ra^{226} and 0.003 microcuries of ThC' were present. This concentration of thorium would not add to the dose appreciably. Subjects who used this material would probably not be of interest since this material was not for internal consumption.

Sample 2 - Radium Water

" MUIDAR "

Carl H. Schultz, N. Y.

MUIDAR

" RADIUM WATER "

Manufactured after

Dr. T. D. Boulanger's process

gamma 1% 9% beta minus
 alpha + 90%

Dept. of Commerce, NBS, Washington D. C.

Test No. 2205 (number obscured) (changed label by printing over it)

Made with pure distilled water

1 mineral-water glass (6 ounces)

Contains 29.9 mache units pure radium

Manufactured for Radium Water Corporation of New York by Carl H. Schultz,
430-444 1st Avenue, New York City

6.8 fluid ounces net

The bottle of "MUIDAR" contained 0.016 microcuries of radium. The thorium content is less than 0.3% of the radium on a radioactivity basis. If the assumption is made that a person drank 1000 bottles of this water and that the gastrointestinal absorption was 100% in the body (an absorption of 25% is more realistic), then a burden of about 0.1 microcurie might be expected at the present time.

Sample 3 - ARIUM

" ARIUM "

Reg. U. S. Patent Office

Radium Tablets

42 tablets of genuine ARIUM

Price \$1.00

Take 2 tablets with glass of water before or after each meal.

To derive the most benefit, ARIUM should be taken as directed.

Read enclosed circular. (None enclosed with sample).

Associate Radium Chemists, Inc., New York City

The full box of tablets (42) contained 0.5 microcuries of radium and no thorium. If 2 tablets were taken before each meal as directed by the label, a box (namely, $0.5 \mu\text{c}$ radium) would be taken weekly. It would thus

be necessary for a person to have taken 20 boxes of pills (20 weeks) (assuming 100% uptake from the gastrointestinal tract) if he were to have a burden of the order of $0.1 \mu\text{c}$ today.

Sample 4 - Radium Facial Cream

Small, glass bottle with metallic cap
Radium Facial Cream
Practically full
1 ounce of dried-out salve

The container did not carry any label other than "Radium Facial Cream"; the 1-ounce container was practically full of dried-out salve which contained $0.018 \mu\text{c}$ of radium. The use of this material could not have represented an internal hazard.

Sample 5 - Radium Water

Radium water for bathing purposes only. This bottle contains $4.20 \mu\text{g}$ of radium (element).
Radium Chemical Company, Pittsburgh, Pennsylvania
About 2 tablespoons left in sample.

The cork stopper was leaking and liquid was present on the outside of the bottle. The bottle contained about 1 fluid ounce of liquid which assayed $0.8 \mu\text{c}$ of radium (Ra + B). The cork stopper was replaced with a rubber one and a measurement 8 days later yielded a value of $2 \mu\text{c}$. This growth indicates the presence of $2.4 \mu\text{c}$ of Ra^{226} in the bottle. If the label was correct, namely, this bottle originally contained $4.2 \mu\text{c}$ of radium, a substantial portion of the liquid must have evaporated and left the radium behind.

Sample 6 - Paradox Radium Radioform

Alcohol 6%. A powerful radium antiseptic. Absolutely harmless even to the mucous membrane. Is nonirritating, nonpoisonous and practically without odor or color.

A thin film-like coating is formed where applied which holds the radium on the spot to destroy germs, kill pain and build up healthy cells by electric charges and penetrating light rays of the radium actually contained in every drop.

Bleeding is arrested and pain stopped quickly for cuts, bites, burns, and scalds. Will arrest poison ivy and poison oak and clear it quickly. Sunburn blisters, lip sores, hives, prickly heat, leg sores and ulcers yield readily to radioform.

Directions: Apply, spread or pour on affected surface with piece of gauze over parts with adhesive tape if condition is deep. For chronic cases use Paradox Radium Liquid X-rays or ointment.

Price: \$1.50
Net Contents: 8 ounces

Guaranteed to contain radioactive element in permanent equilibrium with minimum amount of 2,700 miche units emitting 43 million alpha particles and 71 million beta electrons per second, together with deeply penetrating gamma rays. ABSOLUTELY HARMLESS.

Paradox Radium Laboratories, Inc., Chicago, U.S.A.

CAUTION: Do not keep near unexposed photographic plates or film.

The full, unopened bottle contained $0.8 \mu\text{c}$ of radium and no detectable thorium. The stated alpha-to-beta ratio (41×10^6 alpha to 71×10^6 betas) is not correct for either radium, thorium, or a mixture of both. The guaranteed values of 41×10^6 alphas and 71×10^6 betas per second requires the presence of either $220 \mu\text{c}$ or $480 \mu\text{c}$ of radium (in equilibrium) respectively.

Conclusion

It is certain that none of these six nostrums contained an appreciable quantity of mesothorium. One cannot rule out the possibility that an appreciable quantity of 1.9 year RdTh might have been present.

VII. Status Report of Study of Long-Term Effects of Radium in Humans

Asher J. Finkel, M.D.* and Charles E. Miller

During the period from July 1 through December 31, 1958, an additional thirty-seven subjects were studied. These persons are from a large population of former radium dial painters, of which a total of ninety-three ** have been measured to date. This number represents a random sampling of individuals without reference to clinical or detailed occupational history (other than experience in the radium dial industry).

Cooperation of these persons has been secured by assurance that all information will be treated as strictly confidential. Accordingly, dates of employment, identity of the industrial firms, and locale of employment will be deferred until this study is essentially completed.

Clinical studies have included complete skeletal radiographic survey and complete blood count. An occupational and medical history is obtained at the time of measurement of body content of radium with the gamma spectrometer. The distribution of individuals who have been studied to date** is given in Table 34.

*Health Division

**In addition to the 6 radium dial painters studied at Argonne National Laboratory and reported previously.(9)

Table 34

	Measured	Clinical study
over 1 μc	14	13
0.1 μc to 1 μc	16	15
0.01 μc to 0.1 μc	38	30
less than 0.01 μc	25	17
Total	93	75

Reports of body content of radium and summaries of the X-ray and blood findings are routinely transmitted, upon written request of the patient, to the personal physician in each case. Permission of the personal physician is obtained before clinical studies are made. The active cooperation of physicians and dentists in the several communities has been secured

and cordial relations exist between our group and these persons. A local county medical society meeting sponsored by the Argonne National Laboratory has assisted us materially in having the members of the medical profession locate and refer patients to us. In turn, some patients are referred occasionally to other interested institutions more favorably located for the study.

A list of more than three hundred names and addresses has been assembled with the cooperation of patients already studied, as well as that of local physicians and dentists. It is anticipated that from seventy-five to one hundred additional subjects can be studied before June 30, 1959.

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FURTHER STUDIES ON BACKGROUND COUNTS IN LARGE NaI CRYSTALS

H. A. May and O. J. Steingraber

Introduction

Previous reports have discussed the contribution of various sources to the observed background counting rate in NaI crystals within massive shielding.⁽¹⁻³⁾ We may classify the principal sources of this residual count as follows: (a) activity within the crystal proper, (b) activity of the crystal container, optical reflector and window, (c) activity within the phototube or tubes, (d) radioactivity material within the shield mass, or on its inner surfaces, (e) high-energy ionizing particles associated with cosmic-ray phenomena—mainly mu mesons and electrons, and (f) nonionizing components (neutron and gamma rays), arising either from meson decay or interaction or from the cascade processes representing most of the cosmic-ray energy at the earth's surface.

In this report we are presenting a preliminary evaluation of contributions (e) and (f) made by comparison of background observations at our Laboratory and at a site approximately 250 feet below grade level. The overlay consisted of 36 feet of fill and topsoil, 38 feet of clay and 168 feet of limestone, the total estimated at 163 meters in equivalent depth water equivalent (m.w.e.). The penetrating component of ionizing cosmic rays is known to vary with depth according to the approximate relationship $\frac{I(d)}{I_0} = d^{-1.8}$, where d is distance in m.w.e. to top of earth's atmosphere.⁽⁴⁾ The number of events at our depth relative to the earth's surface is calculated to be 0.0083 by this method. By way of comparison, V. C. Wilson⁽⁵⁾ gives a reduction factor of 0.014 at the same depth.* Thus, this depth is sufficient to reduce the cosmic-ray intensity by approximately two orders of magnitude.

Method

Equipment was located in a tunnel being constructed as part of the City of Chicago water system. Figures 39 and 40 show a general view and closeup of the equipment on location. A smaller version of the Argonne Iron Room⁽⁶⁾ was constructed to provide identical shielding from local activity at both locations. Like its prototype, it consists of 1/4-inch steel plates built up to an 8-inch thickness and an inner lining of 1/4-inch of lead. Inside dimensions are 15 x 15½ by 27 inches, total weight is about 11,000 pounds. With an unshielded crystal, the count rate in the tunnel was found to be roughly 3/4 that observed at the basement laboratory, reflecting mostly the lower radioactivity of surrounding limestone in the tunnel as

*By coincidence Wilson's data at station 10 were taken at exactly the same depth.

Fig. 39. General view of experimental site.

Fig. 40. Plastic anticoincidence detector in position over Fe cave, electronic equipment in background.

compared to the concrete used at Argonne. Analysis of crushed limestone removed from the tunnel showed radioactive content as follows*

Radium	1×10^{-13} curies/gm
Thorium	4×10^{-7} curies/gm
Potassium (by K^{40})	0.1%

Hence, the results obtained within the Fe shield are believed to be independent of radioactivity of the surroundings; very special precautions, however, were necessary to eliminate the effects of radon and thoron daughters collecting on dielectric surfaces.

To determine the influence of highly penetrating ionizing particles, a plastic scintillator coincidence detector was employed for some of the data. It consisted of a 24-inch diameter by 4-inch thick disc of styrene base scintillating plastic, prepared in the manner described by Wouters.^{(7)**} Six circular flat surfaces were milled and polished at equal intervals around the frustum of the cylinder. Type 6363 phototubes were optically coupled to these areas. The remainder of the plastic surface was covered with TiO_2 in an acrylic binder as a reflector.*** The disc was enclosed in a close-fitting stainless steel can. Individual isolating amplifiers at each tube socket fed the output signals to an adding stage followed by a conventional slow linear amplifier (rise time $0.4 \mu\text{sec}$, gain of 2000) and a discriminator supplying a coincidence gate to the multichannel analyzer.

Results

Data in the tunnel were taken with three crystals, sizes $4 \times 1\frac{1}{2}$ inches, 5×4 inches, and $7 \times 3\frac{1}{2}$ inches. The smallest has been in use since 1954; hence, there is a great backlog of data on its response under varied conditions in both its original aluminum and Pyrex container and its present one consisting of stainless steel and quartz. It is known to have a relatively high potassium content by present-day standards. The 5-inch x 4-inch crystal was included in the program at the request of Dr. Philip Gustafson, who has reported some preliminary results elsewhere in this report. It is in electrolytic copper and quartz - the standard Harshaw type B assembly. The largest crystal, also grown by Harshaw, was canned at Argonne in mid-1958; the double-walled stainless steel container is of special design to permit

*We are indebted to Dr. P. F. Gustafson, Division of Biological and Medical Research, for this analysis.

**We acknowledge the gift of a large plastic disc from Dr. Wouters.

***Krylon Spray Enamel, Type 1501, Krylon, Inc., Norristown, Pa. No tests were performed to demonstrate optimum performance; however, the experience of others indicates that this is a satisfactory reflector.^(8,9)

an integral one-half inch layer of mercury shielding. Its potassium content is believed to be of the order of 1 ppm - the "cleanest" of the three by a wide margin.

We have found it convenient in human counting to adjust the phototube and amplifier gain so as to obtain an energy span with the 256-channel analyzer of 6.25 kev per channel, or 100 kev per group of 16. The lower level control of the converter is adjusted so that the linear plot of photo-peak energy vs channel number crosses through zero energy at channel 4. This places the 1.46-Mev peak of potassium-40 comfortably within the upper limit of the analyzer. We shall refer to the resultant energy span from 25 kev to 1.575 Mev (channels 8 - 255) as the standard calibration. The following section refers to data over this energy range.

Figure 41 compares the backgrounds of the $4 \times 1\frac{1}{2}$ -inch crystal within the iron cave at Argonne and in the tunnel. The upper spectrum was obtained at Argonne with the crystal facing upward, i.e., looking at a rather large fraction of the $1/4$ -inch Pb shielding, and represents a total of 200 ± 0.45 counts per minute. With the crystal inverted, resting on its face against the floor of the cave, the spectrum is modified as indicated by the dotted line, the total count being reduced to 178 ± 0.94 c/m. We are unable to state at this time the cause of this difference and shall disregard the higher reading in the discussions to follow.

Fig. 41. Background spectra, $4 \times 1\frac{1}{2}$ -inch crystal.

The lower solid line represents the mean of two underground runs, one facing upward, the other down. The total counting rates in each case were 120 ± 1.1 and 125 ± 0.71 . The errors quoted are standard deviations as calculated from total number of events recorded. Since the two tunnel runs were made fourteen days apart, there was a greater chance of a small error in the calibration, as compared to the consecutive runs at the laboratory; hence it is believed that there is no statistically significant difference for these two positions in the tunnel runs. The elimination of essentially all of the cosmic-ray mesons and associated secondaries is seen to reduce the background count by at least 55 c/m (31%).

Table 35 summarizes these and additional runs in the tunnel together with comparable data from our laboratory under the same shielding arrangements.

Table 35
(4" x 1.5" crystal)

Shielding	In tunnel		At ANL	
	Total time, min	Counts/min	Total time, min	Counts/min
Fe cave only	125	121.6 ± 1.0	200	178 ± 0.94
Fe plus 1 inch Hg*	50	129.3 ± 1.6	1000	177 ± 0.42
Fe cave, face down on 2" Bi bricks	200	125.5 ± 0.71	200	172 ± 0.93
Fe cave, plus 2" of Bi all around	80 200	112.5 ± 1.2 110.2 ± 0.75	2040	156 ± 0.28

*The mercury shielding consisted of three separate stainless steel casings filled with reagent grade, triply-distilled mercury: a cylinder $8\frac{1}{4}$ inches long and $6\frac{1}{2}$ inches I.D., a $6\frac{1}{2}$ -inch diameter end plug, and a similar plug with a $2\frac{3}{4}$ -inch diameter opening to pass the neck of the phototube housing.

The presence or absence of activity within or upon the interior walls of the cave may be deduced from the observed effects of additional close fitting shielding about the crystal, provided that the additional shielding is known to be clean, or its contribution accounted for. In this case, when the crystal was surrounded by one inch of mercury, an increased background resulted. The difference was too slight to yield a statistically meaningful difference spectrum. Moreover, further experiments showed

the Hg shields to be contaminated with radium; in addition, the extent of possible radon or thoron contamination was not appreciated when these tunnel measurements were made. The data, however, are consistent in indicating that the difference (~ 45 c/m) between the readings at Argonne and in the tunnel is fairly independent of the type of shielding (Hg or Bi) immediately surrounding the crystal. The greater deviation (57 c/m) obtained when no inner shielding was added to the iron of the cave remain at present unexplained since it cannot be ascertained whether it is due to spurious surface radioactive contamination or represents true secondary interaction between cosmic-ray components and Fe only instead of Fe + Hg (or Fe + Bi). The fact that, percentagewise, the decrease due to earth shielding is roughly equal in both instances (68.3 and 71.1%) would suggest that we are observing secondary cosmic-ray interactions with materials of different Z; this impression is supported by the sharpness of the positron annihilation line (0.511 Kev) in the difference spectrum obtained with bismuth lining (Fig. 42). A close analysis of the situation awaits reproducibility of some observations obtained at the Argonne National Laboratory site.

Fig. 42. Net difference spectra due to cosmic rays, $4 \times 1\frac{1}{2}$ -inch crystal (ANL minus tunnel)

Data obtained with the 7×3.5 -inch crystal sizes are in some ways comparable although significant differences do exist. Background spectra of the $7 \times 3\frac{1}{2}$ -inch crystal (Fig. 43) again reveal an over-all reduction, typical figures being 555 c/m at ANL vs 380 c/m in the tunnel, with a large proportion of the difference again being in the 511-keV region. (Figure 44) Percentagewise (68.5%) this reduction is similar to that obtained with the 4×1.5 inch crystal. As before, with no close-fitting heavy Z shield, a pronounced 609 keV radium contribution is evident.

The significance of the net reduction by 175 counts per minute may be evaluated by comparison of coincidence rates at the two locations. The plastic detector discriminator bias was adjusted to give a slight increase

in counting rate with a $0.1 \mu\text{c Co}^{60}$ source placed at its center. Contrasted to the 50 ± 5 counts per minute which were observed in coincidence at the Laboratory, in the tunnel the figure was only $1.2 \pm 0.1 \text{ c/m}$. Roughly speaking, this means that the total count contributed by the nonionizing component is roughly three times that of the ionizing component.

Fig. 43. Background in $7 \times 3\frac{1}{2}$ -inch crystal under various shielding conditions.

Fig. 44. Net cosmic-ray spectrum, $7 \times 3\frac{1}{2}$ -inch crystal.

Table 36 again summarizes data obtained with different shielding arrangements around the 7" crystal.

Table 36
(7" x 3½" crystal)

Shielding	In tunnel		At ANL	
	Total time, min	Counts/min	Total time, min	Counts/min
Fe cave only	270	381 ± 1.2 (mean of 5 runs)	1375	555 ± 0.65 (mean of 4 runs)
Face down on 1" Hg, 1½" Hg in case	80	268 ± 1.8	1350	413 ± 0.6
Face down on 2" Bi, plus 1½" Hg in casing	70	268 ± 2.0	970	404 ± 0.65
Surrounded by 2" Bi, close-packed	80	272 ± 1.9	2590	427 ± 0.4
Surrounded by 2" Bi, plus 1½" Hg in casing			960	379 ± 0.6

The spectrum of the approximate 110 c/m removed by additional shielding is shown in Fig. 45.

Fig. 45. Net reduction in count due to additional shielding.

The annular space containing the mercury around this crystal was kept sealed, thus preventing buildup of radon daughters within. Effectiveness of the mercury shielding is demonstrated by the additional decrease in

counting rate by some 29%. Since the surface contamination problem was recognized when these runs were made, greater precautions were taken in handling and scrubbing the bismuth bricks. The virtually identical count rates and spectra with 1/2 inch of mercury and with 2 inches of bismuth (Table 36, lines 3 and 4) must indicate that either the two shields are identically contaminated, or - much more probable - contain no activity detectable within the uncertainties of this experiment.

Evaluation of the contribution from K^{40} in the 5-inch photomultiplier tube was obtained by counting a second unbased tube separated from the crystal face by a quartz blank. A preliminary breakdown of the total background observed at the Laboratory is as follows:

Cosmic rays, charged particle interactions	55 c/m
Cosmic rays, "soft" or cascade component	120 c/m
Fe and Pb shield	110 c/m
5" phototube, and preamplifier	100 c/m
K^{40} in crystal, (2 ppm estimated)	30 c/m
Residual, unaccounted for (0.0165 c/m/gm NaI)	140 c/m
Total	555 c/m

A description of the scintillation spectra at higher energies awaits further analysis of the collected data.

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LOW BACKGROUND STUDIES USING THE BIM 5-INCH BY 4-INCH NaI CRYSTAL

P. F. Gustafson,* L. D. Marinelli, and S. S. Brar*

A number of background runs have been made using the BIM 5" x 4" NaI crystal under various shielding conditions. The spectral region considered extends from 0.13 to 1.5 Mev. The crystal, canned in electrolytic copper by the Harshaw Chemical Company, was optically coupled to a DuMont 6364 photomultiplier tube. The necessary electronic circuitry was soldered directly to the pins at the phototube base to decrease contact "noise." The ceramic base itself was not removed, and thus a portion of the background counting rate may be due to activity contained therein.

The background data consist of runs made in the basement of Building D202 outside the steel room, and inside the steel room (9" shielding) with and without additional shielding of 1 inch of mercury. Similar readings were taken in the lead cave (4" shielding). In addition, data taken in the Chicago water tunnel inside a steel cave (8" shielding) with and without the mercury shield are also included. Table 37 indicates the counting rate from the total crystal and counting rate per gram of NaI under various shielding conditions. The latter figure is computed on the basis of the 5" x 4" NaI crystal weighing 4.72 kg, its volume and NaI density of 3.67 g/cm³.

Table 37

Background counting rate of BIM 5" x 4" NaI crystal under
various shielding conditions
0.13 to 1.5 Mev

Shielding	Background counts per minute	
	Total crystal	Per gram
None, crystal outside BIM steel room	16,059	3.402
BIM steel room	606	0.128
BIM steel room + mercury shield	371	0.079
BIM lead cave	711	0.151
BIM lead cave + mercury shield	401	0.085
Chicago water tunnel steel cave	297	0.063
Chicago water tunnel steel cave + mercury shield	278	0.059

* Biological and Medical Research Division.

As seen from the data, the background in the steel room is about 100 c/min less than that prevailing in the lead cave when no mercury shield is used. However, the use of mercury shielding yields counting rates more nearly equal. This may imply the removal of the major portion of the background which is due to radioactivity in the surroundings external to the detector itself. This is further borne out by the tunnel observations, where the background with comparable steel shielding is decreased by some 300 c/min with only steel, and 100 c/min (approximately) with the addition of mercury shielding. The background reduction in the tunnel is due in part to reduction of cosmic ray flux, and to the lower level of environmental radioactivity found in limestone.

The small reduction incurred by the use of mercury in the tunnel may be partly explained on the basis that the BIM 5" x 4" crystal has a fairly high potassium content ($2.6 \mu\mu\text{c K}^{40}$ in the total crystal) and contamination with an unknown emitter, which gives rise to the 3.2-Mev line. This line stood out very sharply in the spectra taken in the tunnel and thus is even more strongly suspected to be caused by an internal source. It is, therefore, possible that counts in the region 0.13 to 1.5 Mev may also come from the radioelements producing the 3.2-Mev line.

We should like to express our appreciation to H. May and O. J. Steingraber for extending the opportunity of making runs with the BIM 5" x 4" crystal in the tunnel.

MEASUREMENT OF SOIL RADIOACTIVITY AND CALCULATION OF THE DOSE THEREFROM

P. F. Gustafson*

A spectrometric method, using NaI crystal, for the measurement of radioactivity in soil has been described in previous ANL Reports.^(1,2) Some radiation dose values were also reported.⁽²⁾ This report will present the methods of dose calculation in greater detail, as well as activity and dose values for specific soil samples.

In order to calculate the dose rate at any point in air arising from natural and fission radioactivity present in and on the ground, it is necessary to utilize two different mathematical approaches. On the one hand, the natural activity is uniformly distributed throughout the soil at least down to depths of several feet; on the other hand, fission product radioactivity, is primarily present on the surface with relatively slight penetration in depth due to lack of solubility in water and leaching.

Part I. Calculation of Dose from Natural Radioactivity.

The energy absorbed per gram of soil from the uranium, thorium, and potassium contained therein will be the same as that released per gram of soil at all points well beneath the surface. At the surface, however, energy absorption in soil will be, to a good first approximation, one-half that prevailing at lower levels. This is due to the fact that the direct radiation from soil at any surface point is coming essentially from one-half the solid angle considered for a deep point since the radioactivity content of air is several thousandfold less than that of soil. Although scattering by the air of the radiation originating in the earth would be expected to differ from the corresponding scatter by the soil above a point at a considerable depth, the correction will be small because the energy of almost all the radiation is well above the K absorption limits of the elements involved, and the mass of air above the earth is thick enough to react with essentially all of the radiation coming from the earth.

In a given small mass of air at the point of interest, the energy absorption will be equal to that in a corresponding mass of surface soil, but multiplied by the ratio $(\sigma_a + \tau)_{\text{air}} / (\sigma_a + \tau)_{\text{soil}}$. This ratio was obtained for each gamma-ray energy present in the decay of the uranium and thorium series and of K^{40} . For the purpose of finding $(\sigma_a + \tau)$ for soil, soil was assumed to be composed of equal parts of C, O, N, Na, K, P, S, Mg, Ca, and Si; although this is not the true composition of soil, the values of $(\sigma_a + \tau)$ are fairly constant above 0.2 Mev; in fact, the ratio is

* Biological and Medical Research Division.

essentially unity for photons above 0.24 Mev. In order to compare our dose equations directly with those of Hultqvist,⁽³⁾ the abundances of the various gamma rays from U, Th, and K⁴⁰ were taken directly from Hultqvist's article.

Expressing the measured concentrations, s_i , of U, Th, and K in g/g, the dose in $\mu\text{r}/\text{hour}$ using the above procedure is given by:

$$I_U = 0.78 s_U$$

$$I_{\text{Th}} = 0.38 s_{\text{Th}}$$

$$I_K = 1.62 \times 10^{-4} s_{K^{39}}$$

These equations are to be compared with those of Hultqvist, which are:

$$I_U = 0.76 s_U$$

$$I_{\text{Th}} = 0.36 s_{\text{Th}}$$

$$I_K = 1.57 \times 10^{-4} s_{K^{39}}$$

Part II. Calculation of Dose from Fission Radioactivity.

Calculations of dose from an infinite plane have been made by Dunning⁽⁴⁾ with the assumption that fallout sources consist of uniform, non-self-absorbing surface layers, with no distribution in depth. The dose in r/hour is then given by the following expression:

$$D = 1.75 C E \ln \left[\frac{h^2 + x^2}{h^2} \right]$$

where C is the activity in curies $/\text{m}^2$, E the gamma-ray energy, h the height above the plane, and x is the radius of the finite plane under consideration. Dunning evaluates the logarithm for 0.75 Mev and h equal to one meter as 8.63.

This expression has been modified in two ways; 1) the effect of air absorption has been considered in evaluating the logarithm, 2) the effect of the depth distribution of fallout as regards both absorption and dose build-up has been taken into account. A numerical integration using concentric rings made it possible to determine the asymptotic value of $\ln \left[\frac{h^2 + x^2}{h^2} \right]$

for various gamma-ray energies. Values of the logarithm calculated in this manner are shown in Table 38.

Table 38

Values of $\ln \left[\frac{h^2 + x^2}{h^2} \right]$ as a function of energy

E (Mev)	$\ln \left[\frac{h^2 + x^2}{h^2} \right] *$
0.14	7.1
0.50	8.0
0.66	8.2
0.75	8.3
1.48	9.0
2.18	9.4

* where h equals one meter

A study of the distribution in depth of fission products from fallout made at the Argonne site in July 1957 showed that 77% of the activity was present at a depth of 0 to 3.8 cm, 16% at 3.8 to 7.6 cm, and the remaining 7% at 7.6 to 11.4 cm. To simplify the calculation, the activity was assumed to be concentrated one-third of the way down in each layer, i.e. at 1.27, 5.08, and 8.89 cm respectively. Absorption and build-up factors were then determined using this modified distribution pattern. The absorption coefficients used were taken from those for concrete given in NBS Circular 583.⁽⁵⁾ The composition of concrete is similar to that of soil, and the linear absorption coefficients were found using the measured density of soil of 1.3 g/cm³. Similarly the build-up factors were determined from the data of Wyckoff et al.⁽⁶⁾ using an equivalent thickness of soil instead of concrete. One further effect was considered: that of backscattering. Spectral data using a Cs¹³⁷ source, with and without backscatterer of soil, indicated an increase in counting rate of less than 10 percent. In terms of dose, the increase would be even less than this value.

Introducing these modifications into Dunning's equation, the expression for dose from fission products on the ground becomes;

$$D = 1.75 C E \ln \left[\frac{h^2 + x^2}{h^2} \right] F_A F_B$$

where the first terms have the same significance as before, and F_A is an attenuation factor due to absorption in soil, and F_B is the dose build-up factor in soil. The results using the modified form for dose calculation differ from those of Dunning by only a few percent in all cases except that of Ce¹⁴⁴. Here the absorption in soil reduces the dose value of Ce by approximately 40 percent, and changes the total dose by <5%.

Part III. Radioactivity Measurements and Dose Calculations of Soil Collected at Argonne.

A series of soil samples collected from the Argonne site have been analyzed by gamma-ray spectrometry for natural and fission activity. The average values of the concentration of uranium, thorium, and potassium are indicated in Table 39 along with the doses arising from such concentrations according to Hultqvist's method. These samples were collected between May 1957 and September 1958. Sampling was done over a known area and down to a definite depth, thus making it possible to express the measurements in terms of activity per unit area. The first three samples are from the top $4\frac{1}{2}$ inches of soil, the last three from the top 6 inches. The activity due to $\text{Zr}^{95}\text{-Nb}^{95}$, Cs^{137} , $\text{Ru}^{103,106}$, and $\text{Ce}^{144}\text{-Pr}^{144}$ expressed in mc/km^2 are shown in Table 40. The change in activity due to the various isotopes reflects the frequency and magnitude of weapons tests. Note that Cs^{137} is rising, and in fact has almost doubled during the interval from October 1957 to September 1958. Comparison with the *in vivo* measurement of Cs^{137} in humans by Miller et al.⁽⁷⁾ indicates a greater increase in deposition of this isotope on the ground than occurs in the case of body content over essentially the same time interval. From May 1957 to July 1958, Cs^{137} in soil goes up by a factor of 1.77, whereas the body burden increases by a factor of 1.44. This lag between physical deposition and incorporation into the food cycle is to be expected.

Dose calculations using the modified form presented in Part II are recorded in Table 41. In order to establish an average dose due to fission products each reading in Table 41 is multiplied by the time interval between that reading and the next one. This procedure, although still not yielding the true average value, is more likely to give values that are too high than if an average of the six readings were taken. Thus the result may be considered an approximation to an upper limit of dose due to the fission products measured.

On the basis of the data presented in Tables 39 and 41, and adding $3.42 \mu\text{r}/\text{hour}$ from cosmic radiation at the latitude of Chicago, we arrive at a dose rate of $12.6 \mu\text{r}/\text{hour}$ arising solely from natural sources. The contribution to the dose from fission activity ranges from a low of 1.31 to $3.10 \mu\text{r}/\text{hour}$ occurring in July and September 1958, respectively, with a weighted average from May 1957 to September 1958 of $2.25 \mu\text{r}/\text{hour}$. Since these measurements do not include I^{131} , $\text{Ba}^{140}\text{-La}^{140}$ or any other fission products of less than 30-day half-life, the actual dose rate would presumably be even higher. In any event, the value of $2.25 \mu\text{r}/\text{hour}$ amounts to an increase of 18 percent over the natural dose rate for the Chicago area. For the sake of comparison, an increase of 15 mr/year over that present from natural sources has been suggested by the International Committee on Radiation Protection (ICRP) as the value to be allowed for planning purposes in the development of nuclear energy programs.⁽⁸⁾

Table 39

Concentration of thorium, uranium, and potassium in
soil and the dose therefrom at
Argonne National Laboratory

	Thorium	Uranium	Potassium-39
Concentration, g/g of soil	$8.03 \pm 0.21 \times 10^{-6}$	$4.06 \pm 0.06 \times 10^{-6}$	$2.02 \pm 0.01 \times 10^{-2}$
Dose Rate,* $\mu\text{r}/\text{hour}$	2.89 ± 0.07	3.08 ± 0.04	3.17 ± 0.02

* Calculated according to Hultqvist's equations

Table 40

Fission product radioactivity on the ground at
Argonne National Laboratory

Isotope	Activity in mc/km^2					
	1957			1958		
	May	July	October	April	July	September
Zr ⁹⁵ -Nb ⁹⁵	69.4	75.1	88.6	77.0	36.6	121.5
Cs ¹³⁷	13.4	13.1	14.3	17.4	23.0	31.8
Ru ¹⁰⁶	67.5	69.5	69.5	65.5	58.2	81.2
Ru ¹⁰³	67.5	77.1	92.5	50.1	13.4	31.2
Ce ¹⁴⁴ -Pr ¹⁴⁴	92.5	100.1	106.0	96.4	83.0	104.8

Table 41

Dose from fission product activity on the
ground at Argonne National Laboratory

Isotope	Dose in $\mu\text{r}/\text{hour}^*$					
	1957			1958		
	May	July	October	April	July	September
Zr ⁹⁵ -Nb ⁹⁵	1.36	1.47	1.74	1.51	0.78	2.38
Cs ¹³⁷	0.11	0.10	0.11	0.14	0.18	0.25
Ru ¹⁰⁶	0.18	0.18	0.18	0.17	0.15	0.21
Ru ¹⁰³	0.42	0.49	0.58	0.32	0.08	0.20
Ce ¹⁴⁴ -Pr ¹⁴⁴	0.05	0.06	0.06	0.05	0.05	0.06

* According to the method described in Part II.

Part IV. Measurement of Cs^{137} in Soils from Nevada and Utah

A group of some 40 soil samples from Nevada and Utah which were collected before and after the Plumbob Series in 1957 have been analyzed by gamma-ray spectrometry for Cs^{137} as well as other fission products. Collection was made by the Environmental Radiation Group under Dr. Kermit Larson of the Atomic Energy Project at UCLA.

These were so-called agricultural soil samples, meaning that they were taken from land under cultivation. This factor of physical mixing of soil by plowing may account, in part at least, for some of the spread between samples: in particular it should account for the fact that in a few cases there actually appears to be less Cs^{137} present in the soil after the tests than there was before. Due to the age of the pre-Plumbob samples, with subsequent decay of the bulk of the fission products, no values other than those for Cs^{137} will be reported here. Some of the soils collected close to the test site still (18 months post-Plumbob) contain too much Zr^{95} - Nb^{95} to make an accurate determination of Cs^{137} possible. These samples are therefore not included in the present report.

Results for various localities for the top one inch are shown in Table 42, and for the top six inches of soil in Table 43. Also included in Table 42 is a value for Cs^{137} in soil collected at Belle Fourche, South Dakota, which received a rainout from one particular shot. It should be noted in Table 43 that the Cs^{137} content of soil at Argonne collected in October 1957 is approximately the same as for areas much closer to the test site. This phenomenon may have a twofold explanation: 1) There is a considerable degree of fractionation between Cs and other fission products close to the point of detonation due to the radioactive precursors of Cs^{137} . (This is illustrated by the ratio of Zr^{95} to Cs^{137} activity, which runs as high as 200 to 1, namely, larger than indicated by the fission yields of these two isotopes.) 2) The greater rainfall in the Chicago area increases the amount of Cs^{137} fallout from both continental and worldwide testing.

It is a pleasure to acknowledge the assistance of Dr. G. M. Dunning of the AEC Division of Biology and Medicine in initiating this cooperative effort.

Table 42

Pre- and post-Plumbob agricultural
soil samples
0 to 1 inch depth

Location	Cs ¹³⁷ activity in 10 ⁻¹⁰ curie/kg	
	Pre-Plumbob sample	Post-Plumbob sample
Alamo, Nev.	2.10	2.01
Belle Fourche, S. Dak.	---	8.62*
Delamer	1.69	0.59
Enterprise, Utah	1.90	2.00
Lund (II), Nev.	2.14	5.57
Milford, Utah	5.53	4.59
St. George (II), Utah	2.66	2.81

* From area of known heavy fallout from a single shot.

Table 43

Pre- and post-Plumbob agricultural
soil samples
0 to 6 inch depth

Location	Cs ¹³⁷ activity in 10 ⁻¹⁰ curie/kg	
	Pre-Plumbob sample	Post-Plumbob sample
Alamo, Nev.	1.11	1.59
Beaver, Utah	2.36	1.56
Caliente, Nev.	0.54	1.16
Cedar City, Utah	0.80	0.96
Eureka, Nev.	1.21	1.19
Lund (II), Nev.	2.17	1.21
Mesquite, Nev.	1.21	1.21
St. George (I), Utah	1.44	1.16
St. George (II), Utah	0.70	1.20
Tempiute, Nev.	2.90	3.96

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MEASUREMENT OF Cs^{137} AND K IN GRAIN

P. F. Gustafson* and S. S. Brar*

As a preliminary step toward correlating the Cs^{137} content of soils with that found in the food cycle and that actually incorporated in man, measurements have been made on grain samples. Both oats and barley collected in the Matanuska Valley, Alaska in 1957 have been assayed by gamma-ray spectrometry for Cs^{137} and potassium. These samples were supplied by Dr. Edward M. Scott of the Department of Health, Education, and Welfare at the suggestion of Dr. Paul Pearson of the Division of Biology and Medicine of the AEC. The results of the analysis are shown in Table 44. In 1956, Anderson et al.,⁽¹⁾ found an average value of Cs^{137} in flour and cereals of 2.2×10^{-11} curie/kg and a concurrent potassium content of 1.1 g per kg.

Table 44

Cs^{137} and K in oats and barley from the
Matanuska Valley, Alaska in 1957

Grain	Cs^{137} , c/kg	Potassium,* g/kg
Edda barley	5.9×10^{-11}	5.79
Swedish oats	1.7×10^{-11}	5.77

*Contents reported in WADC Tech. Report 52-301, pp. 116-121, are 1.23 and 3.68 g/kg of pearled barley and rolled oats.

In cooperation with Dr. A. G. Norman of the University of Michigan, the build-up of Cs^{137} in certain grains which have been grown in the same locality will be investigated. This yearly crop analysis will start with grains harvested in 1954. Soil content of Cs^{137} during these years will also be assayed and correlated with the uptake in the plant.

Reference

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THE EFFECTS OF METEOROLOGICAL VARIABLES UPON THE VERTICAL AND TEMPORAL DISTRIBUTION OF ATMOSPHERIC RADON

H. Moses, A. F. Stehney, and H. F. Lucas, Jr.

I. Introduction

On May 20-21, 1958, a series of twenty-seven consecutive hourly measurements of radon concentrations were made at four different levels of the Meteorology Tower. These measurements, together with the associated meteorological data obtained by the Meteorology Group, probably represent the best data of this type available. A report on this work was made in the Semiannual Report of the Radiological Physics Division for January-June, 1958.(1)

During the past six-month period two additional runs of this type were made on July 16-17 and August 27-28, 1958. These measurements showed a number of features not evident in the previous run concerning the distribution of radon in time, space, and the relationship to meteorological variables. The results of this work are presented in this report.

II. Experimental Procedure

Essentially the same experimental procedure was used for the runs of July 16 to 17 and August 27 to 28, 1958 as was used for that of the previous run made on May 20 to 21, 1958. Twenty-minute samples were obtained for each hour beginning at ten minutes before the hour and ending at ten minutes past the hour. In the second series the runs started at 9:50 a.m. CST on July 16 and ended on July 17 at 12:10 p.m. CST, thus providing 27 consecutive hourly samples. The run of August 27 to 28 was shorter in duration. It started on August 27 at 15:50 p.m., and ended on August 28 at 8:10 a.m., thus providing 17 consecutive samples. Measurements during the July run were made at the following four levels: 1/8 inch, 3.17 feet, 18.75 feet and 131 feet above ground. During the August run measurements were made at 1/8 inch, 3.17 feet, 78 feet, and 131 feet above ground.

The air samples were obtained by drawing air through one-fourth- and three-eighths-inch diameter copper tubing attached to the Meteorology Tower and leading to an instrument shack located at its southeast leg. The radon in 50 to 100-liter samples of air from each level was adsorbed on a charcoal trap cooled by a dry ice bath, using the system previously described (2,3). The methods of analysis and the accuracies of the system are the same as in the previous runs.

III. Discussion of Data

A. Comparison of Three Runs

In Figure 46a-c are shown the hourly values of radon concentration for the three experiments. The data for the first run are repeated both for the sake of completeness and for comparison with other data. Although the meteorological conditions during the first two runs were similar in a number of respects, some striking differences in the radon concentration distributions with time and height appeared. During the third run the differences in the radon distribution and weather variables were markedly different from those of the other two.

Fig. 46a

Fig. 46b

Fig. 46c

1. Statistical Summary

Table 45 is presented to summarize a number of parameters pertinent to each of the experiments. These are as follows:

a. Maximum Radon Concentration ($\mu\mu\text{c}/\text{liter}$) and Time of Occurrence

At corresponding levels the maxima were greatest during the May run. The maximum value of $3.93 \mu\mu\text{c}/\text{liter}$ measured at the 1/8-inch level during the July run represents 39% of the maximum permissible concentration of $10 \mu\mu\text{c}/\text{liter}$.⁽⁴⁾ Even during the August run when diffusion was relatively greater, the surface value of $1.07 \mu\mu\text{c}/\text{liter}$ represented about 11% of the maximum permissible concentration.

Little significance can be attached to the times of occurrence of the maximum values other than that they occurred during the dark hours, except at the 131-foot levels during the July and August runs, and at the 78-foot level during the August run. The exceptional cases all occurred at 0600 CST. The maximum at the upper levels occurring shortly after sunrise is of significance and will be discussed below.

b. Minimum Radon Concentration ($\mu\mu\text{c}/\text{liter}$) and Time of Occurrence

The values and time of occurrence for the August run are not comparable with those of the other runs since the measurements began at 1600 CST. With the exception of the 1/8-inch level during the July run, the other minima ranged from about .03 to .05 $\mu\mu\text{c}/\text{liter}$. These all occurred somewhere between 1000 and 1200 CST.

c. Maximum and Minimum Radon Concentration Ranges ($\mu\mu\text{c}/\text{liter}$) and Ratios

The data showed that during clear nights when a strong inversion was present, the radon concentrations increased by a factor of 15 to 20 as compared with the daytime value. With a weak inversion the concentrations were approximately doubled.

d. Maximum Build-up and Decline Rates ($\mu\mu\text{c}/\text{liter}/\text{hour}$)

During the first run the maximum build-up rate substantially exceeded the decline rate, but the two rates were comparable during the other runs. The horizontal advection of a pronounced radon concentration discontinuity during the first run may account for this difference.

e. Maximum Gradients ($\mu\mu\text{c}/\text{liter}/\text{hour}$)

As is to be expected, the maximum gradients within the lowest layers are much more pronounced than those in the upper layers. The last two runs show that from the surface to three feet the radon concentration gradient was about 200 times as large as near the top of the tower.

Table 45

Statistical summary - radon concentration measurements

	May 20-21, 1958 Tower level				July 16-17, 1958 Tower level				August 27-28, 1958 Tower level			
	3 17'	18.75'	78'	131'	1/8"	3.17'	18.75'	131'	1/8"	3.17'	78'	131'
Maximum concentration, $\mu\mu\text{c}/\text{liter}$	0 960	0 850	0 650	0 445	3.93	0 928	0 582	0 276	1 07	0 488	0 383	0 355
Time of maximum, CST	0300	0300	0200	0200	2400	2100	0300	0600	2300	0100	0600	0600
Minimum concentration, $\mu\mu\text{c}/\text{liter}$	0 053	0 039	0 039	0 039	0 223	0 047	0 038	0 034	0 484	0 181	0 174	0 159
Date	5/20	5/20	5/20	5/20	7/17	7/17	7/17	7/17				
Time of minimum, CST	1000	1100	1000	1000	1000	1200	1000	1000	1600	1600	1600	1600
Range max. min., min.	0.907	0.811	0.611	0.406	3.71	0 881	0 544	0 212	0 523	0 307	0 209	0 196
Ratio max/min	18.1	21.8	16 7	11.4	17.6	19.7	15 3	8.1	2.2	2.7	2.2	2 2
Max. build-up rate $\mu\mu\text{c}/\text{liter}/\text{hr}$	0.492	0 416	0 436	0 365	1.97	0 647	0 184	0 234	0 30	0 101	0 055	0 044
Hours, CST	01-02	01-02	01-02	01-02	23-24	23-24	02-03	05-06	22-23	22-23	22-23	03-01
Max decline rate, $\mu\mu\text{c}/\text{liter}/\text{hr}$	0.218	0 229	0 168	0 146	2.00	0 314	0 190	0 157	0 25	0 119	0 042	0 032
Hours, CST	06-07	06-07	06-07	07-08	04-05	24-01	06-07	07-08	23-24	01-02	07-08	06-07
Gradient calculations												
	May 20-21, 1958 Tower level				July 16-17, 1958 Tower level			August 27-28, 1958 Tower level				
	3.17'-18 75'	18.75'-78'	78'-131'	1/8"-3.17"	3.17'-18.75'	18.75'-131'	1/8"-3.17'	3.17'-78'	78'-131'			
Maximum gradient, $\mu\mu\text{c}/\text{liter}/\text{ft}$	0.0094	0.00359	0.00447	0.95	0.0382	0.00476	0.206	0.00196	0.00109			
Time of maximum gradient, CST	0200	0300	0400	2400	2400	0300	2300	0100	0700			

2. Additional Features Shown by Radon Graphs

Besides the information characterizing the three runs shown in Table 45, a number of other features appeared. These may be summarized as follows:

a. Advective Effects

In the May run a sharp rise in radon concentration occurred at all levels from 0100 to 0200 CST. The fact that it occurred at all levels simultaneously and was associated with a sharp change in wind direction are strong points of evidence indicating that the rise was due to air with a higher radon concentration brought in over the station. There is some evidence for a similar effect in the July run at 2300 to 2400 CST, but the effect is not as clear cut.

b. Phase Shift in Concentration Maxima

In both the July and August runs there is a tendency for high concentrations to show up first at the surface and work their way up, thus appearing later at the 3.17 foot and 18.75 or 78 foot levels. It is of interest that during the July run concentrations at the 131-foot level did not increase until 0600 CST. The rise at this level occurred after sunrise with the increased vertical turbulent transport.

c. Diurnal Variation Patterns

The May and July runs were made during sunny days and relatively clear nights. Consequently, strong convection during the day transferred the radon upward at a rapid rate so that low concentrations were observed. During the night the atmosphere became very stable as a result of strong nocturnal cooling, thus suppressing vertical mixing. Therefore, high radon concentrations developed in the lower layers. During the August run radiational heating and cooling were smaller; consequently the amplitude of the diurnal variation of radon concentration was markedly reduced.

IV. Meteorological Relationships

A. Wind

Wind speed and direction were measured at five levels above ground: 79", 18.75', 37.5', 75', and 150'. The plots of wind speed and direction shown in Figures 47 and 48 are based on 20-minute averages taken during the radon observation periods. Only the direction at 75 feet is shown as being representative of the air flow over the station.

An examination of the hour-to-hour variations of wind speed in the lower layers shows a gross pattern of high values during the daytime and low values at night. However, a number of apparently minor fluctuations are superimposed on the gross pattern. An analogous pattern is observed for other meteorological variables such as temperature. Stability and radon concentration exhibit a similar pattern but with a 180-degree phase shift, i.e., higher values occur at night. One is tempted to relate the minor fluctuations of one of these variables with the others. An examination of the data with this in mind provides a number of instances where a change in one variable corresponds to a change in the other or where secondary peaks or valleys of the two traces correspond. Many instances of such correspondence appear when the radon and wind data are compared. However, more data than are available are necessary to establish the relationship on a sound basis.

FIGURE (47a)
HOURLY VALUES OF WIND SPEED AT INDICATED LEVELS
METEOROLOGY TOWER - MAY 20-21, 1958

FIGURE (47b)
HOURLY VALUES OF WIND SPEED AT INDICATED LEVELS
METEOROLOGY TOWER - JULY 16-17, 1958

FIGURE (47c)
HOURLY VALUES OF WIND SPEED AT INDICATED LEVELS
METEOROLOGY TOWER - AUGUST 27-28, 1958

It is of interest to point out the following points of correspondence:

1. Minima in the 79" level wind speed correspond to maxima in radon concentration at the 1/8" level. Examples: 1) July 16, 2100 CST. 2) July 17, 0400 CST. 3) August 27, 2300 CST.

2. Since the gross patterns for radon concentrations and wind speed are out of phase, it is to be expected that falls in one variable should correspond to rises in the other. However, it is possible to point to exceptions to this. Example: May 20, 0700-0900 CST.

It is not surprising to find high radon concentrations with low wind speeds since under these conditions mechanical turbulence is also low, with the result that the vertical turbulent exchange coefficients are small, thus allowing an accumulation of radon near the ground.

3. The degree of terrain roughness differs with direction from the station; consequently the turbulent structure and diffusion rates may differ for winds from various directions. For this reason a change in wind direction may bring in air with differing amount of radon, thus causing a variation in radon concentration. Examples 1) May 20, 0100-0200 CST. 2) July 16, 1600-1800 CST. 3) August 27, 2100-2400 CST. (The pronounced wind shift of July 16 was characteristic of a frontal passage.)

The wind direction for each of the three runs was different: west northwest for the May run, northwest up to 1700 CST and then east for the July run, and south for the August run.

B. Stability

Figure 49 shows the hourly values of stability for the following atmospheric layers. 5.5-15.2' 5.5-34.0' 5.5-72.2', and 5.5-144'. The stability values represent four-minute averages of the temperature difference between the top and the bottom of each of the above layers.

An examination of the radon data and the stability traces brings to light a number of interesting relationships. These may be summarized as follows:

1. Variation of Radon Concentration with Stability

Examination of the gross features of the stability and radon curves shows that marked similarity exists. In general, when the stability is high, the radon concentrations are high and vice versa. It may be noted that on the day when the radon concentration was highest, May 21, the stability was greatest; similarly, the night which had the weakest nocturnal inversion, that of August 28, exhibited the lowest radon concentrations.

FIGURE (49a)
 HOURLY VALUES OF ATMOSPHERIC STABILITY AS DENOTED BY TEMPERATURE DIFFERENCE
 BETWEEN TOP AND BOTTOM OF INDICATED LAYERS
 METEOROLOGY TOWER - MAY 20-21, 1958

FIGURE (49b)
 HOURLY VALUES OF ATMOSPHERIC STABILITY AS DENOTED BY TEMPERATURE DIFFERENCE
 BETWEEN TOP AND BOTTOM OF INDICATED LAYERS
 METEOROLOGY TOWER - JULY 16-17, 1958

FIGURE (49c)
 HOURLY VALUES OF ATMOSPHERIC STABILITY AS DENOTED BY TEMPERATURE DIFFERENCE
 BETWEEN TOP AND BOTTOM OF INDICATED LAYERS
 METEOROLOGY TOWER AUGUST 27-28, 1958

2. Radon Concentration Rise with Completion of Lapse-Inversion Transition

In the transition period between lapse and inversion conditions, the thermometers located between 5.5 and 144 feet may show a decrease of temperature with height for some layers, while others may show a simultaneous increase. In all three runs a sharp rise in radon concentration occurred when all the layers showed an inversion simultaneously for the first time, i.e., the transition was complete. Note the rise in radon concentration at 2000 CST during the May and July runs and 2200 CST during the August run.

3. Decline of Radon Concentrations with Morning Break-up of Inversion

Examination of the charts illustrates quite well that when the inversion breaks up, the radon concentrations fall because of the increased vertical transport, which causes the distribution of radon over a thicker layer. However, in the transition process from inversion to lapse conditions, the concentrations at the 131-foot level increased shortly after the inversion started to break up. This may be explained by the passage of air with higher radon concentration from the lower layers through the 131-foot level. Such a radon peak occurred at 0600 on all the runs.

C. Temperature

The temperature readings shown in Figure 50 represent 20-minute averages taken during the same interval the radon measurements were taken. An examination of the charts showing hourly values of temperatures shows that a striking inverse relationship existed between the pattern of radon concentrations, especially at the $1/8''$ and 3.17' levels and the temperature curves. As examples sharp falls in temperature corresponded to sharp rises in radon concentrations, periods of temperature minima corresponded to periods of radon maxima; and the larger the daily range (maximum minus minimum) of temperature the larger the range of radon concentration. In fact, for the three sets of data the curves of hourly temperatures served as an index of the variation in radon concentrations.

An interesting chain of events takes place, with feedback at the various steps, to produce the observed changes in temperature. The radiational cooling at night, which causes the temperature to fall in the lowest atmospheric layers, produces stability, which in turn suppresses vertical motions and reduces the downward transport of momentum. As a result, the wind speed becomes lower, and vertical turbulent transport or mixing is reduced further, thus favoring a strengthening of the inversion. This process results in a reduction of the turbulent exchange coefficients and the accumulation of radon in the lower layers. Thus we see that the same

Fig. 50

factors which operate to reduce the air temperature result in an increase in radon concentration. An analogous line of reasoning may be employed to explain temperature rises which result in instability, increased turbulent exchange coefficients, and consequently a reduction in the radon concentration near the surface. It is, therefore, reasonable that the radon concentration curves and temperature curves bear strong points of resemblance since both are produced by the same driving forces. A similar resemblance is evident in the stability and ambient temperature charts.

D. Soil Temperature

Soil temperature data are based on two-minute averages taken either during the period of the radon observation or within five minutes.

Only the readings for the ten- and twenty-nine foot levels were made outside of the radon measurement period. The soil temperature data for the July and August runs are shown in Fig. 51.

Fig. 51

The temperature of the upper layers of the soil may contribute to the distribution of radon in two ways:

1. The rate of release of the formed radon in the soil is affected by temperature since the soil aeration is affected by this variable.⁽⁵⁾

2. The heat flux from soil to air is a function of the temperature distribution within the soil among several other parameters. Since the amount of heat released by the soil affects the air temperature, it is part of the chain of events which determines the rate of diffusion of radon within the lower layers of the atmosphere.

Soil temperatures near the surface were lowest during the May run; the values during the July and August runs were comparable. In the May and August runs the air temperature at 5.5' above the surface was lower than the 1-cm soil temperature during the night, but higher during the day. In the July run the 1-cm soil temperature was higher during the entire run. The effect of the soil temperature distribution upon the observed radon concentrations is not apparent. More data and statistical analyses are necessary to bring out the soil temperature effects.

E. Humidity

Dew point temperature data for the 5.5- and 131-foot levels are plotted in Figure 52. These values are based on the averages of two two-minute intervals taken during the radon measurement period.

The air during the May run was quite dry, with dew points ranging from the middle twenties to the middle thirties. In the July run dew points ranged from the upper forties to the lower sixties, and in the August run dew points were mostly in the sixties. There appears to be no marked similarity between the shape of the dew point traces and radon concentration curves.

For a full understanding of the factors which contribute to the concentration of radon at a given level, information concerning the distribution of moisture with height is of importance. The flux of radiation from above is a function of the water vapor pressure. Since the vapor pressure is a function of the dew point temperature, the rate of cooling, and consequently the establishment of a strong inversion, and its effect upon the turbulent exchange coefficient are dependent upon the dew point temperatures in the lower layers of the atmosphere. The high moisture values found during the August run no doubt contributed to the reduced rate of nocturnal cooling in the lower layers, thus weakening the inversion and resulting in smaller radon concentrations.

F. Pressures

Hourly values of atmospheric pressure are shown in Figure 53. The pressure graph shows a rise after a weak frontal passage in the July run, but remains relatively steady during the August run. The effect of pressure appears to be undetectable in these data.

G. Solar Radiation and Net Radiation Flux

The charts showing solar radiation for July and August runs are shown in Figure 54. It may be noted that during the July experiment the days were mostly sunny. However, the variations from hour to hour do indicate that some clouds were present. Not enough of the experiment during August was made during daylight to show a significant portion of the solar radiation curve.

FIGURE(52)
HOURLY VALUES OF DEW POINT TEMPERATURE AT INDICATED LEVELS
METEOROLOGY TOWER

FIGURE(53)
HOURLY VALUES OF ATMOSPHERIC STATION PRESSURE
METEOROLOGY LABORATORY

FIGURE(54)
SOLAR RADIATION INTENSITY ON A HORIZONTAL SURFACE BASED ON
20 MINUTE AVERAGES TAKEN DURING RADON MEASURING PERIODS
METEOROLOGY LABORATORY

The net radiation flux charts (Fig. 55) for both the July and August runs indicate that there was an increase in the net radiation flux upward after 2300 CST. This was associated with an increase in stability and radon concentrations during the very early morning hours. The first portion of the net radiation flux chart for August was omitted because a few sprinkles of rain occurred. Rain wets the radiometer plate, thus causing spurious readings.

V. Summary and Conclusions

Study of these radon measurements with the associated meteorological data brings to light data which may be of use to scientists working with radon and also provides information on the rates of diffusion in the lower layers of the atmosphere.

1. The radon measurements provide information concerning the heterogeneity of the radon distribution horizontally. This heterogeneity was brought out clearly in the May experiment. When the wind direction changed, thus changing the immediate trajectory near the measuring point, the radon concentrations increased markedly.

2. The data also revealed the fact that large changes of radon concentration occurred from one hour to the next. Although large changes were more likely to occur at the lower levels, large hourly changes did occur at the 131-foot level. In addition to advection as a cause for large hourly changes, a sudden clearing at night allows a rapid inversion build-up and may also result in an accelerated radon increase in the lower layers.

3. On the very stable night of July 16-17 with relatively light winds, the large accumulation of radon did not reach the 131-foot level. Typically low daytime concentrations were observed at this level until after sunrise.

4. All three sets of data showed a peak in radon concentration at the 131-foot level at 0600 CST - shortly after sunrise. The most obvious explanation is that with the onset of daytime heating, turbulent vertical transport increased and brought up the radon-laden air. The knowledge that the radon concentrations may increase at a level 100 feet above the ground shortly after sunrise as contrasted with a sharp decrease at the lower levels may prove useful to those conducting scientific work, where the radon background may be a factor, in laboratories located in the upper stories of tall buildings.

5. The data also bring out rather interesting information concerning the ratios of daytime to nighttime radon concentrations. At about 19 feet, the height of the second or third story of a conventional building, the radon concentrations at night were larger by a factor of twenty as compared with the daytime values when the skies were clear. During cloudy conditions the nighttime values were only about twice as large.

6. The graphs showing the hourly values of ambient air temperature served as a rough index to the variation of radon concentration. One must not conclude from this that the radon concentration is a function of the ambient air temperature. Rather, as explained above, the same factors which contribute to rises or falls of the surface air temperature (provided the station remains in the same air mass) contribute to changes in the radon concentration. One might suspect that within the same air mass under dry conditions the daily range of temperature would correlate highly with the daily range of radon concentration at a given level near the ground.

7. The relationships between the radon concentrations and the other meteorological variables pertinent to the assessment of the diffusive capacity of the atmosphere, such as wind and stability, indicate that radon is an excellent tracer for studying the diffusion process in the atmosphere.

To determine adequately the turbulent exchange coefficients for momentum which previous investigators^(6,7) have shown to be equal to those for matter such as radon, would require better anemometers than the Aerovanes we are using and measurements at several different levels closer to the ground. Anemometers such as those manufactured by C. F. Casella and Co., Ltd. of London, England, have been obtained and will be used in future work of this type.

Preliminary calculations of the exchange coefficients using our present instrumentation have enabled us to calculate the vertical flux of radon. If it is assumed that the radon flux is constant with height and, during the period under consideration, the horizontal advection is negligible, then one can easily calculate the rate of radon emanation from the ground. These calculations agreed with published values of $4.0 \times 10^{-5} \mu\mu\text{c}/\text{cm}^2/\text{sec}$ as given by Israel.⁽⁸⁾

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ARGONNE METEOROLOGICAL MODEL TOWING TANK

Paul Frenzen

Introduction

Studies of turbulent diffusion in the lee of a moving grid in the Argonne towing tank have been extended to the unstable fluid case during the period covered by this report. Previous reports in this series⁽¹⁻⁴⁾ have detailed the methods of particle photography and Lagrangian analysis employed, and have described the manner in which the data are treated to compensate for decay of the turbulence field before Lagrangian autocorrelation functions are computed. Results of the current unstable trials are reported here in conjunction with those previously described for neutral and unstable fluid stratification; all results are collected in Fig 56 and Table 46 below.

FIGURE 56
LAGRANGIAN AUTOCORRELATION FUNCTIONS IN STABLY, NEUTRALLY, AND UNSTABLY STRATIFIED WATER, DETERMINED FROM DECAY CORRECTED PARTICLE VELOCITIES MEASURED IN THE LEE OF A RECTANGULAR GRID, $\bar{U} \approx 25$ CM/SEC, GRID BAR = 1 CM, GRID MESH = 5.1 CM, $d\theta/dz = +42$ DEG/CM (STABLE), $+0.03$ DEG/CM (NEUTRAL), AND -0.01 DEG/CM (UNSTABLE)

Table 46

	Fluctuations mm/sec		Rel. energies	Scale times, ξ sec		Scale lengths l_1 , mm		Diffusion coefficients, mm ² /sec		
	(1)	(2)		(4)	(5)	(6)	(7)	(8)	(9)	(10)
	$\sqrt{u^2}$	$\sqrt{w^2}$		T_0	t_1	$\sqrt{u^2}t_1$	$\sqrt{w^2}t_1$	K_x	K_z	K_x/K_z
Stable										
Horizontal	8.0		1.53	1.28	0.49	3.9		31.4		4.61
Vertical		6.5		0.45	0.16		1.0		6.8	
Neutral										
Horizontal	11.7		1.52	1.40	0.49	5.7		67.1		1.40
Vertical		9.5		1.19	0.53		5.0		47.8	
Unstable										
Horizontal	10.5		1.28	1.77	0.54	5.7		59.6		1.30
Vertical		9.3		1.60	0.53		4.9		45.8	

Apparatus

Experiments under conditions of thermal instability were made possible by the installation of a heat exchanger covering almost the entire bottom of the thirty-foot towing tank. Sheets of corrugated aluminum roofing material, laid end to end and sealed with synthetic Thiokol rubber were wedged tightly against the aluminum bottom of the tank to form nine parallel water channels. Inlets and drains were installed in the ends, and a lead-covered, electrical heating cable was laid in the bottom of each channel. The unit can maintain the tank in a more or less steady condition of either stability or instability by circulating cold water through the channels in the one case, or by supplying up to two kilowatts of uniformly distributed electrical heat in the other.

Experimental Conditions

Except for the unstable vertical temperature gradients generated by heating from below, all conditions of the experiment were essentially the same as those of the neutral and stable runs described previously.⁽¹⁻⁴⁾ The fluid depth remained fourteen inches, and the rectangular grid (1-cm bars, 5.1-cm apart) was driven at an average speed of 25.5 centimeters

per second, within one per cent of the speeds in the earlier runs. Timing of the camera start was arranged so that the first particle displacements observed corresponded to average velocities centered four seconds after the passage of the virtual origin* of the decay turbulence field.

Statistical independence of the data was insured by measuring only one trajectory from each experimental run. In addition, a degree of objectivity in tracer selection was attained by choosing for measurement the one legible trajectory nearest the center of the field in each photograph, a procedure which also minimized errors due to parallax.

Twenty-three trajectories from as many runs were selected and measured in this way. Nineteen of them lasted the full twenty-four seconds of the experimental period; the remaining four were nineteen, twenty-one, twenty-two, and twenty-three seconds long. Trajectories shorter than the length of the experiment are caused either by the tracer particle's delayed appearance in or premature disappearance from the field of illumination.

Experimental runs occupied a period of four days, during which period as many separate batches of tracer were employed. This led to some variation in the effective vertical mean velocity since it was quite difficult to reproduce tracer of the same buoyancy for each series of runs. It will be necessary to include photographs of the undisturbed tracer before each run in order to evaluate the buoyancy contribution to the vertical mean flow in future experiments.

Temperatures were measured with five mercury-in-glass thermometers mounted near the test section wall as described in the previous report. In addition, a similar thermometer was placed in thermal contact with the aluminum bottom. These wall-mounted thermometers appeared to be particularly inadequate in this case of continuous mixing due to thermal instability. Though considerable vertical exchange could be seen taking place in the "resting" fluid immediately preceding each run, the thermometers indicated average lapse rates through the test section ranging from a stable $+0.005$ deg/cm to an unstable -0.003 deg/cm. Observations of the gradient prior to all runs averaged very nearly isothermal, but the most frequent observation (about half of the total) showed an unstable lapse rate of the order of -0.001 deg/cm. Horizontal space averaging in the main body of the test section is evidently required.

Powerstats supplying current to the heating cables were set at 100 volts for all unstable trial runs. The slow increase in mean water temperature recorded during the trials (approximately 0.5 deg C per hour)

*The virtual origin is that point where the decaying turbulence would have had infinite energy; in these experiments it lies in the neighborhood of ten mesh lengths behind the grid.

would indicate that the tank received a net 1.1 kilowatt input at this setting, about 80% of that supplied. The thermometer on the tank bottom read about 0.8 degree C warmer than the mean fluid temperature throughout the runs.

Analysis

In computing the longitudinal component of the autocorrelation function $R(\xi)^*$ an estimate of the mean flow was determined by averaging all velocity measurements taken at similar times in the twenty-three experiments. As mentioned in earlier reports in this series, the reproducibility of the mean, longitudinal "surging" flow under similar experimental conditions makes this procedure feasible. On the other hand, vertical mean flow components, because of the changing tracer buoyancy, were largely undetermined. They were, however, kept within ± 1 mm/sec by discarding batches of tracer which rose or sank at greater speeds. For purposes of the analysis of these trails it was necessary to neglect their effect in the calculation of the vertical autocorrelation function, assuming them to be zero.

A method of "block-averaging" (see below) the autocorrelation values over a range of corrected times was introduced in the presentation of the results of the unstable fluid experiments. Some of the data from the neutrally and stably stratified runs (specifically, the vertical components) were reworked in the same way. Figure 56 plots these block-averaged values along with the previously determined, non-block-averaged, horizontal autocorrelation functions of the neutral and stable cases.

From the twenty-three by twenty-four (number of trajectories by number of observation times) input matrix of the unstable fluid experimental series, the GEORGE computer program derived over two hundred fifty correlation values, each of these corresponding to (with a few exceptions due to missing entries) twenty-three pairs of velocity measurements, one pair from each trajectory. Within these there are almost fifty cases of repeated corrected-time (ξ) intervals, eighteen consisting of three or more sets of twenty or more data pairs. These arise from velocity pairs that are common multiples of one another, and which are, therefore, assigned the same corrected time by the second Batchelor correction.**

$$* \text{where } R(\xi) = \frac{u(t) \cdot u(t + \xi)}{\sqrt{u(t)^2} \sqrt{u(t + \xi)^2}}$$

**The Batchelor corrections⁽³⁾ are two in number; the first restores the magnitude of the decaying fluctuation velocity by multiplying by the square root of the observation time, while the second compensates for the increasing scale of turbulence by redefining time intervals in the experiment, dividing them by their times of observation. This last, dt/t , integrates to $\ln t$.

In the twenty-three, twenty-four-second long trajectories of these trials, the correlations of the velocities at time four, with those at time eight, five with ten, etc., gave ten autocorrelation estimates from some two hundred data pairs all corresponding to an identical corrected time lag of 0.69 sec., numerically equal to $\ln 2$. Curves drawn for earlier reports of this series, (e.g., the horizontal neutral and horizontal stable curves, Figs. 56a and 56c) have been principally based upon these repeated time interval values.

In the first tenth of a second of corrected time, averages were taken of the six computed correlation values corresponding to values of ξ between 0.060 and 0.079 seconds and also of the seven values between 0.080 and 0.099 seconds. Correlations calculated for ξ 's less than 0.060, eleven in number, appeared to scatter badly and were ignored. The two average correlations thus obtained, each now resulting from between 130 and 160 data pairs, were arbitrarily assigned ξ times in the center of the blocks over which they were averaged. Distributions of ξ were such that these central times were almost always within ± 0.01 corrected seconds of the arithmetic mean; they never differed by more than ± 0.02 corrected seconds from this figure. Data between 0.10 and 0.70 seconds were grouped in blocks 0.05 sec long; between 0.70 and 1.30 seconds, the blocks were 0.10 sec long, while between 1.30 and 1.80 they were further increased to 0.20 sec in length. Increasing the width (i.e., duration) of the blocks with increasing time lag values kept the number of data pairs in each more nearly constant. In this way, the two hundred sixty-eight correlation values computed from the twenty-three by twenty-four matrix of measured velocities were collected into twenty-three block-averaged values. The number of data pairs represented by each point were increased from the aforementioned twenty or so to between one hundred thirty and four hundred.

Experimental Results

In Fig. 56 the horizontal and vertical autocorrelation functions obtained in the unstable fluid trials are plotted along with those previously evaluated under stable and neutral conditions. A distinguishing feature of the unstable fluid correlation traces is their undulating character. This is especially apparent in the curve of the longitudinal component (Fig. 56e) at medium and late lag times. Evidently this is associated with the strengthening of certain turbulence spectrum components through the addition of thermal convective eddies created by heating the fluid from below. It should be noted that any new wave-length peaks that might result in the cosine transform of this autocorrelation curve (i.e., in the distorted, model power spectrum) have to be interpreted in the light of the intervening Batchelor decay corrections.

As noted in the preceding report, the autocorrelation curves are better compared in detail through a number of parameters which may be derived from them.

Complete explanations of the entries in Table 46 can be found in the preceding report of this series, but briefly they are as follows:

Columns 1 and 2: the decay-corrected, root mean-square velocity fluctuations.

Column 3: the ratio of the horizontal to the vertical turbulent energies.

Column 4: the intercept of the autocorrelation curve with the $R(\xi) = 0$ axis.

Column 5: the Lagrangian scale time, the area under the $R(\xi)$ curve.

Columns 6 and 7: the Lagrangian scale length, column 1 or 2 multiplied by column 5.

Columns 8 and 9: the virtual diffusion coefficients, column 1 or 2 multiplied by column 6 or 7.

Column 10: the ratio of the horizontal to the vertical diffusion coefficient.

Once again it should be noted that the relative magnitudes of entries in the foregoing table can be given greater weight than the individual values of the parameters themselves because of the, as yet, uncertain influence of the mean flows.

Decay-corrected fluctuations observed in the unstable fluid (columns 1 and 2) appear to be the same order of magnitude as those found in the neutrally stratified fluid. However, this may be in error due to the timing of the photos as described below. The unstable fluid's average horizontal fluctuation was proportionally somewhat smaller than its average vertical fluctuation, this leading to a ratio of horizontal to vertical turbulent energies (column 3) which indicates a closer approach to isotropy than in either of the other two cases.

The "time to zero," T_0 , of both the horizontal and vertical components of the Lagrangian autocorrelation function were significantly delayed in the unstable fluid experiments. However, in both curves the delay in crossing the $R(\xi) = 0$ axis is associated with a more rapid fall from larger values of $R(\xi)$ in the earlier portion of the curve, resulting in a tendency to preserve the areas under the curves. Column 5 shows very similar Lagrangian scale times for all cases except the vertical component in the stably stratified fluid.

Lagrangian scale lengths (columns 6 and 7) measured in the unstable case were quite similar to those found under conditions of neutral fluid stratification. Similarly, the diffusion coefficients (columns 8 and 9) and their ratio (column 10) in the unstable and neutral fluid experiments appear to have been much alike.

Since one would have expected larger coefficients of diffusion in the unstable fluid, especially in the vertical direction, it seems likely that the results here in the form of the magnitudes of these coefficients are incorrect. Part of the error arises in the magnitudes of the turbulent fluctuations after they are corrected for decay. In the neutral fluid experiments, some of the first trials carried out in this study, the automatic camera-starting arrangement had not yet been installed. The manual starting technique then employed gave only an approximate estimate of the time of the first velocity measurements and hence only an approximate base for the velocity decay corrections. Entries in columns 1 and 2 for the neutral fluid experiments may well have been somewhat too large as a result. The neutral series will be rerun in order to take advantage of several such recent refinements in techniques of experiment and analysis.

Conclusion

In the three sets of experiments described in this and the previous report, experiments under conditions ranging from extreme stability through neutral stratification to moderate instability, several trends can be seen in the data of Table 46.

A degree of anisotropy (favoring the horizontal) apparently maintained through stable and neutral conditions, was significantly shifted toward isotropy with the introduction of heating from below. However, this may change to anisotropy, favoring the horizontal in the stable case, coming isotropic in the neutral case, and then to anisotropy, favoring the vertical in the unstable case, when timing of the photographic observations is improved and better allowance is made for the mean flow components.

T_0 , the intercept of the autocorrelation curve with the ξ axis, monotonically increased for both components as the stability of the fluid decreased. The greatest change observed was the virtual tripling of T_0 between conditions of stability and neutrality. On the other hand, the Lagrangian scale time t_1 (as measured by the area under the $R(\xi)$ curve) exhibited a tendency to be conserved in every case except that of the vertical component in stable conditions where it was found to be less than a third of the other observations. The Lagrangian scale length showed more or less the same variation.

The ratios of horizontal and vertical diffusion coefficients under conditions of neutral and unstable fluid stratifications were found to be approximately the same; however, these findings are subject to the same restrictions involving phototiming and mean flow influence that were noted for the isotropy estimates above. Under stable conditions, this ratio was, again, more than tripled.

Future experiments, using the heat exchanger to control the temperature of the lower boundary, will investigate these parameters over a range of stability conditions. A thermopile probe, capable of determining horizontally averaged temperatures, will be employed and further efforts will be made to evaluate and compensate for the mean flows.

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PROFILE SIMILARITY AND THE RATIO OF TURBULENT TRANSFER COEFFICIENTS

W. C. Swinbank*

The concept of the influence of buoyant motion in distinguishing the vertical turbulent transfer of heat from that of other properties in the atmosphere was introduced and discussed by Priestley and Swinbank.⁽¹⁾ One of the predicted consequences of this discrimination was that the transfer mechanism for heat would differ from that for other properties which do not influence the motion, such as horizontal momentum (frictional drag) and water vapor.

Since that time a number of experimental investigations (e.g. Pasquill,⁽²⁾ Swinbank,^(3,4) and Rider⁽⁵⁾) designed to test the prediction by means of various measuring techniques have established its essential validity. There has been, nevertheless, considerable reluctance to accept the prediction and its corroboration, more especially at levels near the ground. This has been based mainly on derived relationships among the transfer coefficients which result from claimed similarity of profile form and which are considered to be difficult to reconcile with the supposed consequences of buoyancy, (e.g. Rider and Robinson⁽⁶⁾). The evidence produced by Rider and Robinson was not convincing, appeal being made to visual assessment of observations selected by the authors. Comparison of profiles, even when attempted numerically, is notoriously difficult. Rider's 1954 observations are much better and bring strong support for the claim of similarity. Also, examination of the O'Neill (Lettau and Davidson⁽⁷⁾) and Aspendale (Swinbank loc. cit.) data reveals no systematic dependence of the ratio $\frac{\partial\theta/\partial z}{\partial u/\partial z}$ on the height.

Rider, despite the consistent differences he found experimentally between the transfer coefficients for heat and momentum, prefers to accept his claimed similarity of profiles as evidence for identity of the transfer mechanism, and Sheppard⁽⁸⁾ has also recently repeated this implied contradiction.

In point of fact, profile shape can provide information only on the mechanical component of turbulent transfer. The latter comprises the whole of the transfer of momentum and water vapor whereas, as proposed by Priestley and Swinbank, a further component, convective or buoyant turbulence, reinforces the flux of heat in unstable conditions. Profile shape can tell us nothing about the latter, and this point of view will be sustained in the following. It will be shown that the apparent conflict can be avoided by one assumption - that the component of heat flux due to buoyancy is constant with height. This hypothesis appears more acceptable on closer examination.

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First let us summarize the observations which appear to stand in need of reconciliation:

1. There is a layer of air some meters, or tens of meters, deep above the ground wherein the fluxes of the various properties are constant with height; that is to say, when there is horizontal uniformity, the layer is in a steady state for these variables. In the case of heat there is radiative as well as the turbulent transfer under consideration. Strictly, therefore, the steady state requires constancy with height of the combined transfers. But the radiative flux is comparable with the turbulent flux under conditions of strong isolation, and its divergence with height is small. Thus the radiative flux leaving the ground could not exceed by more than 10 per cent that passing through a higher level 7°C colder, even if the air were a full and strong absorber. The difference in fact will be very much less and we may assume approximate constancy with height of both fluxes.

2. The transfer coefficient for heat exceeds that for momentum (and water vapor) according to the degree of instability, as shown by the results of the direct measurements mentioned above.

3. The profiles of temperature and momentum are similar, i.e.

$$\partial\theta/\partial Z = \alpha \frac{\partial u}{\partial Z}$$

where α is independent of Z .*

The flux equations may be written

$$H = c_p \rho K_H \partial\theta/\partial Z$$

$$\tau = -\rho K_M \partial u/\partial Z$$

whence integration for two successive layers (flux constant with height) and division yields

$$\frac{\overline{\overline{K_H}}_2}{\overline{\overline{K_M}}_2} \bigg/ \frac{\overline{\overline{K_H}}_1}{\overline{\overline{K_M}}_1} = \frac{u_3 - u_2}{u_2 - u_1} \cdot \frac{\theta_2 - \theta_1}{\theta_3 - \theta_2}$$

where the bar signifies a mean value through the layer indicated.

Similarity of profile requires the expression on the right to be unity, whence $K_H = a K_M$ at all heights where a is independent of height and may differ from unity. A value of a other than unity and constant with height is the consequence of profile similarity that some (e.g. Sheppard loc. cit.) have found difficult to accept. The difficulty stems from the presumption that, if buoyancy is to affect the transfer of heat differently from

*Symbols used, see p. 193.

that of momentum, the difference must increase with height. Since profile similarity shows this not to be so, it is concluded that a is unity, which is in conflict with directly measured values of K_H and K_M .

It is, however, intuitively obvious that profile shape cannot distinguish between convective and mechanical turbulence, and this can be demonstrated by use of the mixing-length model. This, though now unfashionable, remains useful conceptually.

Since we are concerned with profile shape we must introduce height derivatives beyond the first:

$$\frac{H}{c_p \rho} = \overline{wT''} + \overline{w\ell} \frac{\partial \bar{\theta}}{\partial Z} + \frac{\overline{w\ell^2}}{2} \frac{\partial^2 \bar{\theta}}{\partial Z^2} + \dots$$

$$-\frac{\tau}{\rho} = \overline{w\ell} \frac{\partial \bar{u}}{\partial Z} + \frac{\overline{w\ell^2}}{2} \frac{\partial^2 \bar{u}}{\partial Z^2} + \dots$$

Symbols Used

- Z - height above ground
- θ - potential temperature
- α - constant
- u - horizontal wind velocity
- w - vertical wind velocity
- ρ - air density
- c_p - specific heat of air at constant pressure
- H - vertical flux of sensible heat by turbulence
- τ - vertical flux of momentum by turbulence (= horizontal drag)
- K_H - coefficient of turbulent transfer of sensible heat
- K_M - coefficient of turbulent transfer of momentum
- a - constant
- ℓ - mixing length
- T'' - temperature anomaly of eddy at its level of origin

Here $\overline{wT}$ is the flux of heat due to buoyancy alone defined by Priestley and Swinbank (loc. cit.) (Note that the terms $\overline{w\ell^2}$ etc. will be non-zero due to the influence of the boundary.) From these expressions it follows that, if H and τ are constant with height and the profiles are similar, $\overline{wT}$ must also be independent of height

This necessary consequence of height-constancy of the fluxes and similarity of profiles is not difficult to substantiate further. For if we regard the heat flux as composed of convective and mechanical components, the latter may remain constant or it may increase or decrease with height. Since the mechanical turbulence alone maintains the flux of momentum constant, it is reasonable to expect the same of heat, whence the constancy of convective heat flux follows.

It should be mentioned that the foregoing discussion is not designed to establish a difference in transfer mechanism between heat and momentum, but to show that such difference as determined by separate measurements of K_H and K_M is not inconsistent with profile similarity. It would indeed appear, if the reasoning presented here is correct, that similarity of profile form is to be expected. An experimental program designed to test this prediction is in course of preparation.

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CONVECTIVE TURBULENCE WIND TUNNEL PROJECT

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Introduction

Wind tunnel scale model experiments have been applied to many atmospheric pollution problems which involve diffusion of stack gases and the aerodynamic influence of buildings and topography. The influence of thermal properties of the earth's surface and the resulting convective turbulence and air currents have been omitted. These studies have treated atmospheric motions as aerodynamic phenomena of the type encountered in various areas of fluid mechanics research and have used conventional equipment.

Future development of wind tunnel scale model experimentation for meteorological and air pollution applications must include thermal influences of the earth's surface. This introduces new scaling criteria and requires new features in test equipment. New problems arise in instrumentation and control.

The work performed at New York University under Argonne sub-contract No. 31-109-38-743 has the objective of investigating the application of wind tunnel scale model experiments to atmospheric phenomena which involve air motions caused by thermal characteristics of the earth's surface. Field experiments conducted at Argonne National Laboratory on smoke diffusion from the experimental smoke stack⁽¹⁾ provide data useful for correlation with wind tunnel experiments. The New York University 3½ by 7-foot wind tunnel developed primarily for air pollution studies has features needed for the desired scale model experiments. The general program of this project is to develop the needed additional equipment and techniques and conduct scale experiments for correlation with field results.

The program of experiments as outlined in the project proposal begins with the simplest type of experiments which will yield valuable data and then progress to more complicated cases in which specific features of the Argonne site are included. This report deals with the simplest form of model arrangement.

Two series of experiments were conducted in late 1957 showing the need for improvements in equipment.⁽¹⁾ Experimental techniques were developed, but the test results were not satisfactory. The results are omitted from this report since the second series is a good deal better and covers conditions tested in the first. The second series consisted of fifteen runs.

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A discussion of the scaling criteria used in these experiments may be found elsewhere.^(1,2)

Apparatus

The $3\frac{1}{2}$ by 7-foot wind tunnel is of the open return type with the air intake within the laboratory building and exhaust out through the roof. Three main sections of the wind tunnel are (1) the upstream air conditioning section, (2) test section and (3) air driving equipment and exhaust.

The upstream section has the purpose of producing for introduction to the test section an airstream which is of uniform temperature and velocity and remains constant with time. The entire section is surrounded with an insulated enclosure which houses thermal apparatus and air straightening equipment. Thermostatically controlled electric heaters at the intake to the enclosure keep the incoming air at constant temperature. Heaters within the enclosure keep the inside temperature constant day and night. Mixing fans prevent thermal stratification. Air straightening equipment consists of an expanding duct with fine mesh screens across the airstream to reduce turbulence followed with a contraction cone which tends to produce constant velocity distribution. The contraction cone terminates at and joins with the upstream end of the test section.

The test section is basically a horizontal rectangular duct 7 feet wide, $3\frac{1}{2}$ feet high, and 40 feet long. One foot aft of the beginning of the test section, a vertical grid of electric heating wires spaced 10 per inch is stretched horizontally across the air stream. Each wire has a separate control, thus providing the means for introducing a vertical gradient of heat input to the air stream as it moves into the test section. A short distance downwind of the heating grid are vertical arrays of hooks, one on each wall, for placing strings across the air stream. These provide the means for controlling the initial velocity profile. The floor and ceiling of the test section are of steel sheet construction and are enclosed on the outside with heating chambers in which electric heating elements and mixing fans control the temperature of floor and ceiling. The left wall of test section (looking upstream) has a series of windows beginning ten feet from the upstream end. The right wall is of plywood construction. The walls are not temperature controlled. The test section framework is of structural steel. Two rails on the ceiling support a survey carriage which, by remote control, positions instrument probes in three dimensions.

Downstream of the test section is located the fan which moves the air stream. It is driven with a variable-speed electric motor for air-speed control. The exhaust air is conveyed to the roof where it is further controlled with adjustable exhaust louvers.

Various auxiliary equipment is used in the experiments. An air supply system and smoke generators provide an oil fog smoke in controlled and measured rates for the smoke diffusion experiments. Helium is introduced with the smoke for gas density control. Temperatures in the test section are measured with thermocouples and read on a Leeds and Northrup Speedomax Indicator. A vertical rake of thermocouples is attached to the survey carriage and may be positioned laterally and longitudinally. A single thermocouple and the velocity probe are mounted on the vertically moving part of the carriage and may be positioned vertically as well as laterally and longitudinally. Both the rake and movable thermocouple are used for measurement of temperature profiles. The movable velocity probe is the sensing element of the hot wire anemometer. It is used for point-by-point measurements of velocity profiles. The velocity level of stream is kept constant by measurement at a point near the upstream end. A Hastings heated thermopile anemometer is used.

For the purpose of the second series of tests in this project two 4 x 5 Crown Graphic cameras with 135-mm focal length lenses are used. They are positioned as shown in Fig. 57. Four electronic flash lamps are used to illuminate the smoke plume. Two are located in the lower corner of the test section nearest the side camera and the other two in the upper corner farthest from the side camera. To improve the contrast between smoke plume and background, the floor and far wall in the view of the cameras and ceiling over the plume are covered with black flock paper.

The model of the Argonne experimental smoke stack is located 28 feet 6 inches downstream of the beginning of the test section. Its position relative to the cameras is shown in Fig. 57. Model data are given in Table 47. No other modeling elements representing the Argonne installation are included in this series.

In future series of tests it is planned to produce nonuniform surface temperatures by placing rubber heating pads on the floor. In order that the same surface configuration be included in the present series, the rubber heating pads were placed but not heated. In order to further roughen the floor surface as an aid to increasing the boundary layer thickness and turbulence, roughness strips were also placed on the floor. The pattern of rubber pads and roughness strips is shown in Fig. 57. The strips are $1/4$ inch thick by one inch wide. The rubber pads are $3/8$ inch thick with a ribbed surface running cross stream. The roughness pattern extended up to the heating grid, a distance of approximately 27 feet. The strips were not used downwind of the stack and the pads were covered with flock paper under the smoke plume.

Fig. 57. Wind tunnel test section

Table 47

Model characteristics

Linear Scale = 1/96

	Prototype	Model
Stack height	110 ft	13.75 in.
Stack inside dia. at exit	17.5 in	0.182 in.
Stack gas temp.	140°F	Helium/air ratio to give same density ratio as prototype for gas temp. of 140°F and ambient air temp. of 80°
Stack gas ejection speed	40 ft/sec	4.08 ft/sec
Time between exposures for multiple exposure photographs	15 sec	1.5 sec
Number of exposures per photograph	16	16

Test Procedure

Each test was conducted at constant wind speed and constant thermal conditions to the extent that constancy of conditions could be maintained. Model test speeds of about 1 to 3 ft/sec were used; these correspond to prototype speeds of about 9.8 to 29.4 ft/sec.

Since thermal controls for the floor and ceiling have heating only - no cooling - thermal conditions obtainable depend to some degree on the initial surface temperatures when a test is started. The airstream temperature profile was varied by the application of heat to the floor, ceiling, and heating grid. In five of the runs the heating grid was used in addition to floor and ceiling controls. In the remaining, only floor and ceiling temperature was controlled. Ceiling temperature was always kept above the temperature of the adjacent airstream to maintain a stable top layer.

A rectilinear coordinate system (X, Y, Z) is used to identify locations within the tunnel. The origin is taken as the base of the stack on the tunnel floor; Z is the distance above the floor, X the distance downstream, and Y is the crosswind distance (Fig. 57). Temperature profiles at location

$X = 0$, $Y = +31$ in. were measured with the vertically movable thermocouple while at the remaining locations the profile was measured with the thermocouple rake.

Point-by-point velocity measurements were made with the hot wire anemometer. Velocity measurement for holding constant velocity level was indicated on the Hastings heated thermopile anemometer. The Hastings anemometer is more sensitive than the hot wire anemometer at the low air speeds of these experiments. The large probe of the Hastings anemometer made it unsuited for point-by-point measurements particularly near the floor and regions of appreciable velocity gradient. Measurement of these low speeds with the hot wire is difficult, and significant inaccuracies sometimes occur.

No attempt was made to force an initial velocity profile at the beginning of the test section as is planned for later experiments.

Multiple exposure photographs of the smoke plume issuing from the model of the Argonne experimental smoke stack were taken at the beginning and end of each run to provide the record needed for obtaining the size and shape of plume outline. Side and top views were taken simultaneously with the two cameras located as shown in Fig. 57. The electronic flash lamps were fired at 1.5-sec intervals for 16 flashes with the shutters of the two cameras open. Thus each photograph shows 16 images and was used to find the envelope of the fluctuating plume.

The elapsed time of each test run including taking of the photographs and various test measurements was approximately one hour. Adjustment of various thermal controls and bringing of surface and air temperatures to desired values took longer, sometimes most of a day. A run would not be started until it appeared that equilibrium had been achieved. It was difficult to hold conditions constant, and some variation occurred during the test runs.

Test Results

Velocity and temperature measurements taken at various locations for two of the runs (number 8 and 11) are shown in Figures 58-61. All of the velocity measurements are shown in logarithmic coordinates and three runs are shown in rectilinear coordinates in Figure 62.

Diffusion characteristics of the model smoke plume are given as horizontal and vertical plume half widths at downwind distance X equivalent to 100 ft in the prototype. This distance was chosen because the corresponding prototype data were available. In the calculations performed

Fig 58

TEMPERATURE PROFILE - RUN 8

Fig. 60

TEMPERATURE PROFILE - RUN 11

Fig 59

VELOCITY PROFILE - RUN 8

Fig 61

VELOCITY PROFILE - RUN 11

Fig 62
VELOCITY PROFILES

for the prototype a virtual origin (generally upstream of the stack) was used. This virtual origin was determined by requiring the plume outline to plot as a straight line on logarithmic coordinates except for regions close to the stack. This gave effectively a point source origin and removed the initial plume expansion due to jet action and other actions not solely due to atmospheric turbulence. For reasons discussed below it was felt that this series of wind tunnel tests did not give plume outline sufficiently well defined to warrant the analysis used in the prototype. An approximate virtual origin was found by fairing to an upstream point the plume outline downstream of the initial expansion.

Downwind distances to location of plume width measurement in the model experiments (equivalent to 100 ft prototype) were measured from this virtual origin and are essentially similar to those of the prototype. Plume half-widths found in model experiments are given in Table 48. Photographs of model smoke plumes are shown in Figs. 63 and 64.

Table 48

Test results
(at prototype scale)

Run No.	Temp. gradient, °C/100 m	Wind profile parameter n	Hor. plume half-width,* meters	Vert. plume half-width,* meters
1	-1.66	0.35	4.18	3.94
2	-1.49	0.28	2.46	3.22
3	-1.44	0.28	2.04	3.05
4	+1.86	0.52	3.33	3.40
5	+0.65	0.45	1.96	2.50
6	-0.77	0.35	2.27	2.50
7	-1.25	0.33	5.11	8.78
8	-1.11	0.33	6.48	8.42
9	-2.12	0.48	1.90	3.40
10	+4.07	0.53	2.73	2.33
11	+1.07	0.48	1.87	2.50
12	-1.25	0.39	2.09	2.87
13	-0.87	0.33	2.04	2.50
14	-1.11	0.44	2.03	2.87
15	-1.95	0.35	2.57	3.40

*At downwind distance $x = 100$ ft (30.48 meters).

Overhead Photograph

Side Photograph

Fig. 63

SMOKE PLUME PHOTOGRAPHS - RUN #7

Overhead Photograph

Side Photograph

Fig 64

SMOKE PLUME PHOTOGRAPHS - RUN #11

Model plume half-widths and half-width ratios are plotted against temperature gradient in Figs. 66-68. These charts also show corresponding data obtained from the Argonne field measurements for model-prototype comparisons. Prototype data are taken from measurements made at Argonne. Temperature gradient in the prototype is based on the temperature difference between 144-ft and 5.5-ft levels at the Meteorology Tower. Determination of temperature gradients in the model experiments is based largely on the lowest foot of boundary layer. This corresponds roughly to 100 ft in the prototype. Preference is given to measurements on the test section centerline which is the location of the stack. Locations near the tunnel walls and higher levels show, in most cases, considerably different values. The straight line in Figs. 58-61 were used for calculation of the gradients in Table 48 shown as prototype equivalents.

Figure 65 shows comparison of wind profile characteristics of model vs. prototype as related to temperature gradient. The wind profile parameter n used in this comparison is defined by the power law for expression for velocity variation with height

$$V/V_0 = (Z/Z_0)^{n/2-n} \quad (10)$$

where V is airspeed and the subscript " 0 " indicates reference level. Reference level need not be identified for n is based on the curves shown in Figs. 59 and 61 where logarithmic coordinates are used in plotting height vs. velocity. Emphasis is placed on centerline locations and the lowest foot of the boundary layer.

Discussion

Comparison of model results with prototype field experiments in Figs. 65-68 show limited agreement and significant differences. The relatively small number of model experiments compared with the prototype leaves some aspects inconclusive. Wind and temperature profile characteristics presented in Fig. 65 show the model results to fall generally in the region covered by the prototype experiments and give a similar trend.

Plume-width characteristics in Figs. 66 and 67 show the model plume to be narrower than prototype although there is some indication of similar trends with temperature gradient. Plume-width ratio in Fig. 68 shows the horizontal width in relation to the vertical to be less in model than in the prototype.

Differences in configurational features between model and prototype such as lack of certain topographical features may account in part for differences in results between model and prototype. It is believed that the

Fig. 65.
WIND PROFILE
PARAMETER VS TEMPERATURE GRADIENT

Fig. 66.
PLUME HORIZONTAL HALF WIDTH VS
TEMPERATURE GRADIENT AT 100 FT DOWNWIND

Fig. 67.
PLUME VERTICAL HALF WIDTH VS
TEMPERATURE GRADIENT AT 100 FT DOWNWIND

Fig. 68.
PLUME WIDTH RATIO VS TEMPERATURE
GRADIENT AT 100 FT DOWNWIND

differences are, however, due primarily to features in the model velocity and temperature profiles. It will be noted that the velocity and temperature profiles show in many cases distinct discontinuities at the top of the boundary layer. Boundary layer height varies between 10 and 15 inches. Plume height at the higher air speeds was close to stack height, namely, 13.75 inches and higher at lower air speeds. Thus the plume is, unfortunately, at or near the top of the boundary layer. Part or all of the plume may, in some cases, be above the boundary layer where there is less turbulence and less diffusive action. This may account in part for the lower plume widths in the model.

A peculiar characteristic is noted in some of the velocity profiles, particularly at low speeds. There is a pronounced change in the slope of the profile near the top of the boundary layer. Figure 62 is presented to emphasize this characteristic. It may seem that a layer of low-eddy viscosity is needed to explain this marked change in slope since it produces a high local velocity gradient. It is believed that the explanation lies, however, in another direction. It will be noted that in cases of irregularity in velocity profile, peculiar temperature characteristics occur. This is emphasized in some cases of unstable gradients as, for example, in run No. 8 (Fig. 58). The gradient towards the sides near the floor is stable but the central region is unstable. It may appear that this is caused by nonuniform floor temperature distribution (and some may occur) but special experiments conducted after this series showed it is due to secondary thermal circulation. Since thermal control is achieved by raising the temperature of the air stream and various surface temperatures above ambient room temperature, wall temperature (which is uncontrolled) tends to be lower than the airstream. This causes the layer adjacent to the wall to be of lower temperature and, therefore, greater density. It tends to fall and flow across the floor towards the center. Thus stable gradients may be found near the sides even though test conditions are set for an unstable gradient. This secondary circulation has undoubtedly some effect on the velocity profiles since the infusion of air from the wall boundaries will modify momentum characteristics of the floor layer. The fact that the profile irregularity tends to occur at low speed is further indication since thermal effects tend to be more pronounced at low speeds.

Asymmetric thermal characteristics tend to make the secondary circulation more complicated. Since one side of the test section is of plywood and the other of glass, the differences in heat transfer rates contribute to the temperature differences between the two sides. The figures show differences in air temperature. The overhead photographs show cross-flow at plume level. This is apparent in Fig. 63 (run 7), one of the extreme cases.

Smoke plume photographs were taken at the beginning and end of each run to determine constancy of test conditions. In all cases there was little or no difference. While velocity and temperature were not remeasured at the end of each test run, it is believed that in some cases there were significant changes in test conditions even though not evident from the photographs. The changes which occur in velocity and temperature profile in moving from one location to another may be in part caused by change in test conditions. Run #2 on Fig. 62 shows distinct differences in shape of velocity profile when moving 25 inches downstream. It is believed that this change is due primarily to change in test conditions rather than change in position.

Conclusions

Comparison of scale model results with those of the prototype show some agreement in wind profile shape as identified by the power law equation and some similarity in vertical plume width variation with temperature gradient. Plume widths are generally lower in the model.

The floor boundary layer was not sufficiently high to envelop the plume. This may be a significant factor in the low plume widths found in the model.

Secondary thermal circulation caused by the test section walls caused peculiar features in velocity and temperature profiles. These made evaluation of the profiles difficult and probably introduced undesirable flow conditions.

The lighting used for plume photographs was not sufficiently uniform to give a well-defined plume outline over the length of the plume. Variation of plume width with downwind distance could not be evaluated.

Plans and Recommendations

The lighting system must be improved to give the needed uniformity in illumination. It is planned to install tubular flash lamps which will accomplish this objective.

A higher boundary layer is needed for better plume diffusion. It is planned to experiment with methods of increasing the boundary layer before the next series of diffusion experiments is conducted. This will involve control of the velocity profile at the beginning of the test section and possibly modification of floor roughness.

Methods of eliminating secondary circulation by modifying thermal characteristics of the walls will be investigated.

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SOIL TEMPERATURE STUDIES

J. E. Carson* and H. Moses

I. Introduction

The program of analyzing soil temperature data and studying the effect of meteorological parameters on thermal conditions in the soil has continued. In a previous report⁽¹⁾ the instrumentation for measuring the soil temperature was described, and the results of studies on the diurnal, ten-day, and monthly cycles of soil temperature at various depths were presented. In this report results of studies on the extreme soil temperature values and day-to-day and hour-to-hour variations are discussed. In addition, a detailed case study of the effects of meteorological variables during a prolonged dry period upon the soil temperature is presented.

II. Extremes of Air and Soil Temperature

The air temperature data used in this study were obtained by means of a sensitive aspirated thermopile mounted 5.5 feet above the ground and recorded on a Brown Electronik Potentiometer. The highest and lowest temperatures for each calendar day are based on five-minute averages. Maximum and minimum soil temperatures are the highest and lowest of the hourly readings. Since extreme values of soil temperature may fall between the regular observation times, this procedure often underestimates values of extremes and the daily range, which is defined as the difference between the highest and lowest hourly soil temperature readings.

Many factors influence the magnitude of the daily range of soil temperature in the upper layers. It is relatively large on a clear sunny day with dry soil and is very small or zero under a snow cover. A large daily range also often accompanies frontal passages with strong advective temperature changes.

The daily maximum and minimum temperatures at the ten-centimeter depth for the year 1955 are shown in Fig. 69. During December, January, and February when the upper soil layer is usually saturated with moisture and is often frozen or covered with snow, day-to-day fluctuations of the maximum and minimum soil temperatures are quite small. No such quiet period is found in the air temperature record, but during these three months the daily range of air temperature is also quite low. The large number of points at the freezing point of water reflects the effects of the heat of fusion of water and a snow blanket.

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Fig. 69

An examination of similar charts for the other depths within the soil indicates a rapid decrease of the daily temperature range with depth in the soil. At the 50-centimeter depth the daily range is usually less than $1\frac{1}{2}^{\circ}\text{F}$ during the winter months. During the rest of the year the daily range is usually 1°F or less. However, values larger than 1°F are not infrequent.

The annual progression of monthly average maximum and minimum and daily range of soil and air temperatures are shown in Figs. 70 a-e. It is of interest to note that February 1954 was an extremely mild month averaging 10°F above normal, while March 1954 was 2°F cooler than normal. This deviation from the normal pattern showed up as an irregularity in the air temperature curve. However, this variation did not show up at any of the soil temperature levels. Another abnormality is the very warm summer of 1955. The high daily maximum temperatures, however, did show up in both the air and soil records.

MONTHLY AVERAGE TEMPERATURE MAXIMA MINIMA AND RANGES FOR AIR AND SOIL
JAN 1953 - DEC 1955

An examination of the monthly mean temperature range for air shows that two maxima occurred, one in the spring and the other in the fall. This phenomenon appears in the data of the five-year climatological summary for the period of July 1949-June 1954.⁽²⁾ During 1953 this pattern is also found at the 1-centimeter level but is not clearly shown in the other two years. At the lower levels the greatest ranges occurred during the summer months and the smallest during the coldest months. Note that the amplitude of the monthly mean daily range curve is greater at the 1-centimeter level than it is for air, but that the range values themselves are greater in air.

Of practical importance is information on the freezing of the soil at various depths. Table 49 presents data on the number of days on which the soil temperature was recorded as 32°F or lower during each month in the three-year period under investigation. Temperatures of 32°F or lower occurred only during the four months of January, February, March, and December at the levels measured. January and February showed the greatest frequency at the 1-, 10-, and 20-centimeter levels. At the 20-centimeter level during February there were over twice as many cases as in January. This, in all probability, is due to the phase displacement of the temperature wave. Of interest is that no freezing temperatures occurred at 50-centimeters (about 20 inches) or below.

Table 49

Number of days with minimum soil temperature less than or equal to 32°F

	1cm				10cm				20cm				50cm
	1953	1954	1955	Total	1953	1954	1955	Total	1953	1954	1955	Total	None
January	30	31	19	80	31	23	18	72	6	19	5	30	
February	28	17	22	67	28	19	24	71	21	18	24	63	
March	8*	2	0	10	10*	0	1	11	0	0	0	0	
April	0	0	0	0	0	0	0		0	0	0		
November	0	0	0		0	0	0		0	0	0		
December	12	2	29	43	4	0	26	30	0	0	16	16	

*The soil temperature data for March 7-8, 1953 were missing. Since the air temperature was below 32°F all day, it was possible that the soil temperatures at the 1- and 10-centimeter levels also fell below this value.

III. Interdiurnal Temperature Variability

Interdiurnal variability of any climatic element is defined as the change from one day to the next and is a useful index of the restlessness of the climate. The quantity used can be the value at a selected hour of the day or it can be the daily average. For this study the twenty-four-hour

change in the air and soil temperatures from midnight to midnight was used. A positive value of the interdiurnal temperature variability (IDTV) indicates a rise in temperature. The interdiurnal temperature variability of the air and the soil temperatures at the 1-, 10-, 20-, and 50-centimeter depths were calculated and monthly averages of the absolute value were determined by means of the following formula:

$$IDTV = \frac{\sum |d|}{N}$$

where $|d|$ equals the absolute value of the twenty-four hour change and N equals the number of days in the month.

Figure 71 shows the monthly average values of the interdiurnal temperature variability in the air and at the 1-, 10-, 20-, and 50-centimeter depths within the soil for each month from January 1953 through December 1955, the period of investigation of this soil temperature study. Figure 72 shows the three-year monthly average values.

Fig. 71

Fig. 72

The annual cycle of the interdiurnal soil temperature variability in Fig. 72 shows two maxima, one in spring and the other in the fall and two minima, the main minimum occurring in February and the secondary in midsummer, July and August. The annual cycle in air is less regular and shows a distinctive maximum in winter and a minimum during August. These charts also demonstrate the expected rapid decrease of interdiurnal temperature variability with depth within the soil. Figure 71 reveals considerable variability in these quantities for a given month from one year to the next. For example, the value for air during April of 1954 is twice as large as the average variability during April of 1953 and 1955. These variations from one year to the next are indicative of the unsettled nature of our climate. An important exception to the rule of decreasing interdiurnal temperature variability with depth occurs during the winter of 1954-1955, when the value of the quantity at ten centimeters is equal to or greater than the value at one centimeter.

IV. Hour-to-Hour Temperature Changes

The monthly average change of temperature from one hour to the next of the air and the soil at 1-, 10-, and 20-centimeter depths has been computed. Since the diurnal cycle of soil temperature is almost completely damped out in the first fifty centimeters of soil, no such curve is possible for this level and greater soil depths.

Figures 73 a-d show the monthly average diurnal cycle of one-hourly temperature change in the air and soil for four months typical of each of the seasons, January, April, July, and October of 1955. Similar charts have been prepared for the other months of the three-year period studied.

January is normally the coldest month in this area and the curves in Fig. 73a indicate very little warming and cooling of the soil. The air, however, does show appreciable warming and cooling rates.

April, a typical spring month, (see Fig. 73b) shows much greater warming and cooling rates within the soil than January. The curves show the characteristic phase shift and amplitude reduction with depth, however, the deviation from a simple sinusoidal pattern in the air and at the 1-centimeter level is pronounced. This, of course, shows the influence of harmonics on the fundamental temperature wave.

The conditions for July, normally the warmest month at Argonne, are shown in Fig. 73c. As expected, the amplitudes of these curves are greater than those of April. Of interest is the fact that the maxima of the air, 1-centimeter and 10-centimeter curves are displaced in time, but the minima are not.

THE AVERAGE ONE-HOUR TEMPERATURE CHANGES IN AIR AND SOIL
FOR THE HOURLY PERIOD ENDING AT THE INDICATED TIME

Fig. 73

The curves for October, a typical fall month, are shown in Fig. 73d. The amplitudes are smaller than those of July, and the maxima occur somewhat closer together. The minima for air and the 1-centimeter soil level occur at the same time, but those of the other two levels appear later.

A case study showing the response of soil and air temperature to intense daytime solar heating and strong nighttime outgoing radiation for the period of August 20-27, 1953, is shown in Figs. 74a-b. The soil during this period was quite dry since the last previous rain of consequence occurred on August 12, when 0.18 inch was recorded. August 20-27 represents a warm but not extremely hot period.

An examination of Fig. 74a-b shows that the daily maximum and minimum temperatures of the air and soil down to 50 centimeters increased steadily during this period. The daily air temperature maximum

CASE STUDY OF SOIL AND AIR TEMPERATURE VARIATIONS DURING SUNNY DRY PERIOD
AUGUST 20-27, 1953

Fig. 74

rose 7.8°C from the beginning to end of the period, while the daily soil maxima at 1 and 10 centimeters increased by only about half as much, 3.9°C and 3.8°C respectively. However, at the end of the period the daily minimum temperatures of the air and the 1- and 10-centimeter soil levels were higher by only 7.1, 3.5, and 3.3°C , respectively, thus indicating an increase in the daily ranges. The diurnal effect upon the 50-centimeter and lower levels was quite small as may be seen from the diagrams.

A plot of the hourly values of solar radiation on a horizontal surface is also shown in Fig. 74a-b. The data show that the days were quite sunny. However, some cloudiness did occur during the afternoon of August 23, 1953. Note the effect of the reduced solar radiation in reducing the air temperature and soil temperatures at the 1- and 10-centimeter levels.

The classical phase shift and amplitude reduction with depth is clearly depicted in the figures. However, the data show a tendency for the daily maximum 1-centimeter soil temperature to occur one hour before that of the air.

Another interesting feature shown by this study is the time variation of the difference between the air and 1-centimeter soil temperature. At the beginning of the period the air was colder than the soil throughout the entire day. At the end of the period the air was warmer for as much as twelve hours of the day. Attention is called to the higher air temperature on August 25, 1953, as compared with that of the previous day. This warming was associated with a change in wind direction to southwest from a direction with an easterly component.

A point to bear in mind is that the air is mobile while the soil is stationary. The modification of the soil temperature distribution depends on the solar heating and radiational cooling at the station as well as continual heat transfer between air and soil. On the other hand, the air temperature arriving at the station is more a function of conditions many miles upwind in its trajectory.

This case study shows graphically the effect of strong solar heating and nocturnal cooling under dry conditions on the soil temperature structure and the relationship between air and soil temperature.

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