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THE ABSORBANCE CHANGE OF IRRADIATED ORGANIC SOLVENTS. PART V.

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

In Part IV of this series[†] it was shown how the absorbance produced in aerated xylene by a Co^{60} gamma-ray dose differed from the absorbance produced in air-free xylene by a similar dose. In order to ascertain to what extent the absorbance produced in irradiated xylene can be used as a measure of dose, measurements of the absorbance produced by gamma-rays under both aerated and air-free conditions were undertaken for a wide range of doses.

The xylene was always doubly distilled. Each vial was filled to the same height and was irradiated under identical conditions. To aerate the xylene air was bubbled through it before introducing it through a small opening at the top of the vial. This opening was maintained patent during irradiation to allow air to diffuse inward from the surface of the liquid.** Curve I of Fig. 1 is a plot of the absorbance observed at 3438 Å as a function of the dose in joules/cm³; the radiation dose rate was 10.4 joules/cm³/hr or 1.2×10^6 r/hr. Curve II is a similar plot for $\lambda = 3555$ Å. The absorbance produced in aerated xylene is relatively stable over a period of a few weeks and the reproducibility of the observations seems to be within $\pm 10\%$. If it were possible to determine what causes the points in Fig. 1 to deviate from the slope at the origin which is shown by the dotted line, this accuracy might conceivably be improved.

In the second case, the vial containing the xylene was placed on a vacuum line and the air pumped off. Then a cycle of freezing, thawing, and freezing again with periodic pumping was carried out, after which the vial was pulled off the vacuum line. A plot of the absorbance produced at 3555 Å versus dose is shown in Fig. 2. In sharp contrast to the case of aerated solutions, the reproducibility for the air-free solutions is not better than $\pm 30\%$. By repeating the absorbance measurements of the same irradiated sample, one noted that the absorbance produced by radiation decreases with time after exposure. The absorbance at 3555 Å produced by a dose of 250 joules/cm³ decreasing to 1/2 of its original value in approximately 28 days. This decrease in the absorbance with time is probably due to oxygen bleaching

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[†]Part I. ANL-5518, p. 8 (1956)
Part II. ANL-5596, p. 3 (1956)
Part III. ANL-5679, p. 5 (1957)
Part IV. ANL-5755, p. 44 (1957)

**It has been pointed out by Dr. E. J. Hart of this Laboratory that this air phase may introduce an error due to ozone formation and reaction with the solutions.

Fig. 1. Effect of gamma radiation on aerated xylene: Curve I is a plot of the absorbance at 3438 Å. Curve II is a similar plot but at a wave-length of 3555 Å. The dose is given in joules/cm³ where one joule/cm³ equals approximately 1.2×10^5 r. The dashed line is a straight line extension of the initial slope.

Fig. 2. The curve in this figure is a plot of absorbance at 3555 Å versus dose for air-free xylene. One-mm quartz cells were used.

which may also be responsible for the erratic results. The rate of decrease of the absorbance as measured at 3555 Å appears to be dependent on the total dose. A larger dose means a shorter time for the absorbance to decrease to $1/2$ its original value.

In conclusion, it may be stated that it is possible to use the absorbance produced in aerated xylene for the purposes of dosimetry under those conditions where a $\pm 10\%$ error is a tolerable uncertainty. The range of such a dosimeter would be from a dose of 3×10^5 r to more than 8×10^7 r.

The authors would like to thank D. E. J. Hart and his group for their cooperation in the use of the Co⁶⁰ source, Mr. W. L. Buck for the use of his Beckman spectrophotometer, Dr. L. Basile for the use of his 1-mm quartz cells, and Mr. L. D. Marinelli and Dr. E. J. Hart for their helpful suggestions.

THE EFFECT OF MASSIVE Co^{60} DOSES ON THE LIGHT OUTPUT OF A SCINTILLATING SOLUTION

I. B. Berlman

Some information on the deleterious effects of radiation on the luminescence of organic crystals and plastic scintillators has recently been published.⁽¹⁻⁴⁾ It is of more than casual interest to compare some of our observations of the harmful effects of ionizing radiation on the light output of a liquid scintillation solution with the following published results on solid scintillating systems.

Black⁽¹⁾ irradiated various organic crystals with a large alpha-emitting source and measured the resultant integrated light output. He found that the total light output decreased with dose and claimed that this decrease followed the quenching formula:

$$L = \frac{L_0}{1 + AN} \quad (1)$$

where L_0 is the original measured luminescence, N is the number of alpha particles/cm² and A is a constant of the material. From the value of N where $L = L_0/2$, $E_{1/2}$ (the dose in ergs/cm³ required to reduce the emission by $\frac{1}{2}$) is determined. Stated differently, if $\frac{L_0 - L}{L}$ is plotted versus N , then $E_{1/2}$ is that value of N where $\frac{L_0 - L}{L} = 1$. When he used beta particles as his ionizing radiation, he obtained no measurable decrease in the luminescent properties of anthracene after a dose of 1.5×10^8 ergs/cm³.

Hinrich⁽²⁾ irradiated polystyrene samples containing various activators with 50-kv X-rays. He states that the light decreases according to the formula

$$L = (L_0 - L') e^{-AN} + L' \quad , \quad (2)$$

where L' is a limiting intensity reached after considerable irradiation. The other symbols are the same as in Equation 1. From one of his graphs it is possible to compute $E_{1/2}$ for a plastic composed of polystyrene plus an unspecified amount of diphenylbutadiene.

Rosman and Zimmer⁽³⁾ irradiated various plastic scintillators with alpha particles and measured $E_{1/2}$. They claim that plastic scintillators do not follow Eq. 1 as is postulated for organic single crystals but that they follow rather the sum of two exponentials.

Schulman, et al.⁽⁴⁾ studied the radiation effects of Co^{60} gamma rays and 2-Mev electrons on anthracene and naphthalene crystals. They confirm Eq. 1 and determine the $E_{\frac{1}{2}}$ values for anthracene and naphthalene. No dose-rate dependence was found from 5×10^4 to 4×10^5 r/hr for Co^{60} and to 1.6×10^8 r/hr for 2-Mev electrons. It is of importance to note that they obtained the same $E_{\frac{1}{2}}$ value for anthracene using 50-kv X-rays as Black⁽¹⁾ obtained using alpha particles. The values of $E_{\frac{1}{2}}$ obtained by these authors are collected in Table 1.

Table 1

Scintillator	Dose for $E_{\frac{1}{2}}$, ergs/cm ²	Type of radiation	Author
Anthracene	1.3×10^8 1.4×10^8	Alpha particles Co^{60} gamma rays	Black ⁽¹⁾ Schulman, et al. ⁽⁴⁾
Naphthalene	1.7×10^8 0.8×10^8	Alpha particles Co^{60} gamma rays	Black ⁽¹⁾ Schulman, et al. ⁽⁴⁾
Terphenyl	5.5×10^8	Alpha particles	Black ⁽¹⁾
Stilbene	7.0×10^8	Alpha particles	Black ⁽¹⁾
Polystyrene + $\frac{1}{2}\%$ TPB*	$\simeq 10^9$	50-kv X-rays	Hinrich ⁽²⁾
Polystyrene + 3% PT [†] + 0.02% TPB	6×10^9	Alpha particles	Rosman and Zimmer ⁽³⁾
Polystyrene + 1.5% TPB	9×10^9	Alpha particles	Rosman and Zimmer ⁽³⁾
Polystyrene + 1.5% TPB	1.8×10^{10}	Beta particles	Rosman and Zimmer ⁽³⁾

*Tetraphenyl butadiene

†Terphenyl

The author's interest in the effects of ionizing radiation on a liquid scintillation system has been stimulated by two facets of the problem: firstly, by the desire to determine at what radiation dose can absorption and luminescence changes be detected, and once having determined this lower limit, to investigate the rate of change of these effects with dose; secondly, by the prospect of throwing some light on the underlying mechanisms by observing the influence of certain parameters, such as scintillator

concentration or impurity concentrations (like oxygen) on the radiation effects. The facility with which these parameters can be varied in a liquid solution cannot be readily matched in the use of crystals or plastics. This report deals only with some of the light output measurements, whereas the preceding article deals with some absorption studies.

The measurements in this laboratory are being made on the organic scintillating solution composed of xylene as the solvent and diphenyloxazole (PPO) as the scintillator. The damage to the solution is caused by massive Co^{60} gamma-ray doses, and the subsequent light output measurements are made with a curium alpha source.* The experimental techniques herein reported are designed to minimize absorption effects on the measurements of radiation damage to the luminescent solution. This is accomplished by studying very thin layers of solution, namely, thicknesses somewhat larger than the range of the curium alpha particle. By a simple modification of a Teflon cell previously reported,⁽⁵⁾ a wide range of solution thicknesses can be tested. The cell, shown in Fig. 3, is provided with a central narrow steel rod, at one end of which a thin film of curium is deposited, whereas at the other end a 1/32-inch threaded screw is soldered. This arrangement permits motion of the source up or down in the solution for measurable distances. By moving the source to within 1/4 turn of its closest approach to the glass plate, the amount of solution under observation is reduced to only 0.008 inch. This thickness is about four times the range of the alpha particles. A photomultiplier tube, in conjunction with a 20-channel analyzer, is used to measure the light flashes due to the alpha particles. In this fashion a sharply peaked spectrum of pulses is obtained, and the position of the peak of the pulse spectrum is a measure of the light output of the solution.

The xylene used in preparing the solution was always doubly distilled under vacuum, while the PPO was used as received from the manufacturer.** Experiments were conducted on both aerated*** and air-free solutions containing either 2 g/l, 10 g/l or 15 g/l of PPO. Furthermore, experiments were performed by irradiating the xylene and then adding the PPO. This latter type of experiment is unique to solutions and cannot easily be done with plastic and organic scintillators. A plot of $\frac{L_0 - L}{L}$ vs dose for a sample of aerated xylene to which 2 g/l of PPO were added after

* Curium is used as an alpha source rather than polonium because it is less toxic since it is not a bone seeker.

** Pilot Chemical, Inc., Waltham, Massachusetts.

*** During irradiation the surface of the liquid was open and in contact with the air phase above it. Dr. E. J. Hart has subsequently pointed out this arrangement complicates matters due to the presence of ozone, with the end result that the data, while reproducible, may be peculiar to this type of experiment.

irradiation is shown in Fig. 4 (Curve I). Note that a straight line may be drawn to a dose of 8×10^9 ergs/cm³; this can be interpreted to mean a linear increase in the number of quenching molecules with dose. The value of $E_{\frac{1}{2}}$ obtained from this figure is approximately 9×10^8 ergs/cm³. Curves II and III of the same figure are similar lines drawn for 10 g/l and 15 g/l of PPO added to irradiated xylene, respectively. It is to be noted that the slope of the line decreases with an increase in PPO concentration and that $E_{\frac{1}{2}}$ correspondingly increases. The value of $E_{\frac{1}{2}}$ for 10 g/l and 15 g/l of PPO from each of these curves is 2.2×10^9 and 2.6×10^9 ergs/cm³, respectively; these values of $E_{\frac{1}{2}}$ and others mentioned below have been entered in Table 2 for purposes of comparison.

Table 2

Data from Co⁶⁰ gamma-ray irradiation

Solution	Condition	PPO concentration	When PPO added	$E_{\frac{1}{2}}$, Ergs/cm ³
Xylene + PPO	Aerated	2 g/l	Before irradiation	7×10^8
"	"	2 g/l	After irradiation	9×10^8
"	"	10 g/l	"	2.2×10^9
"	"	15 g/l	"	2.7×10^9
"	Air-free	2 g/l	Before irradiation	3.4×10^9
"	"	2 g/l	After irradiation	7.4×10^9

On irradiating aerated solution of xylene plus 2 g/l of PPO and plotting $\frac{L_0 - L}{L}$ vs dose one obtains Curve I shown in Fig. 5. A plausible explanation for its departure from linearity at high doses is the decrease in the number of PPO molecules available for the scintillation process. The value of $E_{\frac{1}{2}}$ obtained from this curve is equal to approximately 7×10^8 ergs/cm³.

The irradiation of xylene plus 2 g/l of PPO in an evacuated vial leads to results shown by Curve II in Fig. 5. Although the points are more difficult to reproduce for the air-free solutions than for the aerated solutions, Eq. 1 seems to hold for this particular case (Curve II) in the range of doses shown. The value of $E_{\frac{1}{2}}$ equals approximately 3.4×10^9 ergs/cm³.

Fig. 3. A modified Teflon cell for the measurement of α -ray scintillations in xylene solutions.

Fig. 4. Plots of $\frac{L_0 - L}{L}$ vs dose in joules, cm^3 for irradiated xylene (aerated) to which various PPO concentrations have been added afterwards. One hour of irradiation is equivalent to 1.04×10^8 ergs cm^3 , and one joule equals 10^7 ergs. Curve I is for 2 g/l of PPO, Curve II is for 10 g/l of PPO, and Curve III is for 15 g/l of PPO.

Fig. 5. Curve I is a plot of $\frac{L_0 - L}{L}$ vs dose for an irradiated (aerated) xylene plus 2-g/l PPO solution. Curve II is a similar plot for a solution of xylene plus 2 g/l of PPO in an evacuated vial.

Fig. 6. A plot of $\frac{L_0 - L}{L}$ vs. dose for air-free xylene to which 2 g/l of PPO were added after irradiation of the xylene.

By irradiating xylene under vacuum alone and then adding unirradiated PPO (2 g/l) one obtains the curve in Fig. 6. On extrapolation of this curve, $E_{1/2}$ becomes roughly equal to 7.4×10^9 ergs/cm³.

If PPO is irradiated in powder form with a dose of 3.5×10^9 ergs/cm³ and then 2 g/l of this irradiated PPO are added to unirradiated xylene, there is only a slight measurable decrease in the light output from that obtained if fresh PPO is added to the unirradiated xylene. At dosages smaller than 2×10^9 ergs/cm³ there is no measurable difference in the light output between irradiated xylene plus fresh PPO (2 g/l) and irradiated xylene plus separately irradiated PPO (2 g/l). Measurements were not made at larger dosages. In conclusion, it may be stated that the results shown here indicate that $E_{1/2}$ values are dependent on the scintillator concentration and the presence of air. As would be expected, on the theory of competing mechanisms, it was found that the higher the scintillator concentration the higher the $E_{1/2}$ value.

For the purposes of discussion, only the vacuum irradiated sample of Table 2 should be compared to the plastic samples and organic crystals of Table 1, since they are all characterized by a lack of air. As for plastics, the value of $E_{1/2}$ (340 joules/cm³) for the solution is considerably higher than for the organic crystals (10-70 joules/cm³), indicating greater radiation stability on the part of the former. The paucity of data allows one only to state at present that the radiation resistance of the solutions in vacuum is of the same order of magnitude as that of plastics when gamma rays are used.

It has also been shown that Eq. 1 holds for irradiated xylene (aerated) to which scintillator has been added afterward. This can be interpreted to mean that the number of stable quenching molecules produced by radiation increases linearly with dose. Eq. 1 seems to hold also for the case of vacuum irradiated PPO (2 g/l) plus xylene over the range of doses shown in Fig. 5. Finally it is believed that the deviation of aerated irradiated PPO (2 g/l) plus xylene from Eq. 1 is due to the decrease in effective PPO concentration with dose.

The author would like to thank Dr. E. J. Hart and his group for their generous cooperation in the use of the Co⁶⁰ source, Dr. A. F. Stehney for the use of his chemical balance, and Mr. L. D. Marinelli and Dr. E. J. Hart for their helpful suggestions.

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DRIFT VELOCITY OF ELECTRONS

J. C. Bowe

A. Introduction

Drift velocities of electrons were computed for nitrogen, helium, neon and argon over a range of E/p_0 values corresponding to the energy range for which elastic collision cross sections were available. For this calculation, the energy distribution function $f(\epsilon)$ and the transport cross section, $\sigma_t(\epsilon)$ are required. The most recent formulation leading to $f(\epsilon)$ in high frequency ac fields and dc fields has been presented in an intensive paper by T. Holstein⁽¹⁾ using the method of Boltzmann. This approach leads to an integrodifferential equation defining $f(\epsilon)$, and the solution is approximated by the first two terms of an expansion into spherical harmonics in velocity space, i.e.

$$f(v) = f_0(v) + (v_x/v) f_1(v) \quad (1)$$

where f_0 is a spherically symmetric function and v_x is the component of velocity in the direction of the field. The conditions of validity of this solution are established in Holstein's paper.

The average velocity of drift, $\overline{W}$, in the direction of the electric field is then given by

$$\overline{W} = \frac{\int_0^\infty v_x f(v) d\vec{v}}{\int_0^\infty f(v) d\vec{v}}$$

After substituting Eq. (1) into this expression, we obtain

$$\overline{W} = \frac{\int_0^\infty v_x f_0(v) d\vec{v} + \int_0^\infty \left(\frac{v_x^2}{v}\right) f_1(v) d\vec{v}}{\int_0^\infty f(v) d\vec{v}} \quad (2)$$

where the integral with f_0 is zero due to spherical symmetry. Now, f_0 and f_1 are related by

$$f_1 = \left(\frac{-eE}{\sigma_t(v)m v} \right) \frac{\delta f_0}{\delta v} \quad (3)$$

where e and m are the electronic charge and mass, respectively, E the electric field strength and $\sigma_t(v)$ the transport cross section. The expression for $f_0(\epsilon)$ in terms of the energy variable $\epsilon = mv^2/2$, is given by

$$f_0(\epsilon) = A \exp. \left[\frac{-6m}{M(eE)^2} \cdot \int_0^\epsilon \sigma_t^2(\epsilon) \epsilon d\epsilon \right] \quad (4)$$

where M is the atomic mass and A is the normalizing constant. Using (4), Eq. (3) becomes

$$f_1(\epsilon) = \frac{6m \sigma_t(\epsilon)}{MeE} \cdot \epsilon f_0(\epsilon) \quad (3')$$

By substituting (3') into Eq. (2), we obtain

$$\overline{W} = \frac{24\pi m}{MeE} \int_0^\infty v v_x^2 \sigma_t(\epsilon) \epsilon f_0 dv \quad (5)$$

using the relations

$$d\vec{v} = 4\pi v^2 dv$$

and

$$\int_0^\infty f(v) d\vec{v} \approx \int_0^\infty f_0(v) d\vec{v} = 1 \quad (6)$$

By replacing v_x^2 with $v^2/3$ in (5) and converting to the energy variable ϵ we obtain

$$\overline{W} = \left(\frac{16\pi}{mMeE} \right) \int_0^\infty \sigma_t(\epsilon) \epsilon^2 f_0(\epsilon) d\epsilon \quad (7)$$

which is the expression used in the computation of drift velocity.

B. Computation of the Energy Distribution

The procedure used to compute values of the distribution function $f_0(\epsilon)$ is outlined as follows. Values of the integral $\int_0^\epsilon \sigma_t^2(\epsilon) \epsilon d\epsilon$, which appears in the exponent of f_0 , were determined graphically by means of a planimeter to an average accuracy of about 1% over a range of energies for each gas. These were used to evaluate f_0/A for each E/p_0 (see Eq.4) to an accuracy which varies from a small fraction of a per cent at low energy to as much as 15% at high energy. The relative error in f_0/A varies as the

magnitude of the exponent multiplied by the fractional error associated with the exponent. The normalizing constant A was determined from Eq. (6) by performing the necessary graphical integration, i.e.

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

The computational error of about 1% was associated with A. The values of $f_0(\epsilon)$ presented in Tables 3 to 6, therefore have a computational error which varies from about 1% at low energy to as much as 16% at high energy. The last digit in each of the tabulated numbers is unreliable. The dimensions of $f_0(\epsilon)$ are (gram/electron-volt)^{3/2}.

Table 3

Nitrogen

 f_0 in units of 10^{-42}

Energy, ev	E/p_0					
	0.01	0.02	0.04	0.06	0.08	0.10
0.001	103	53.2	26.6	18.0	13.0	10.2
0.002	103	53.2	26.6	18.0	13.0	10.2
0.003	103	53.2	26.6	18.0	13.0	10.2
0.004	103	53.2	26.6	18.0	13.0	10.2
0.005	103	53.2	26.6		13.0	10.2
0.006	103	53.2	26.6	18.0	13.0	10.2
0.007	103		26.6		13.0	10.2
0.008	103	53.2	26.6	18.0	13.0	10.2
0.009	103				13.0	10.2
0.01	103	53.1	26.6	18.0	13.0	10.2
0.012	103	53.1	26.6	18.0	13.0	10.2
0.014	103	53.1			13.0	10.2
0.016	103	53.1	26.6	18.0	13.0	10.2
0.018	102	53.0		18.0	13.0	10.2
0.02	102	53.0	26.6	18.0	13.0	10.2
0.025	101	52.8	26.5	18.0	13.0	10.2
0.03	98.9	52.6	26.5	17.9	13.0	10.2
0.04	93.4	51.9	26.4	17.9	13.0	
0.05	85.2	50.7	26.3	17.9	12.9	10.1
0.06	74.2	48.9	26.0	17.8	12.9	10.1
0.08	46.9	43.6	25.3	17.6	12.8	10.1
0.09	33.2	40.0	24.8	17.4	12.8	10.0
0.1	19.8	35.2	24.0	17.2	12.7	9.99
0.15	0.39	13.2	18.9	15.4	11.9	9.60
0.2	0.00023	2.10	11.8	12.5	10.6	8.92
0.25		0.084	5.3	8.08	8.7	7.84
0.3		0.000071	1.62	5.1	6.4	6.5
0.35		0.0000008	0.30	2.41	4.2	4.95
0.4			0.044	1.01	2.60	3.6
0.45			0.00540	0.40	1.56	2.61
0.49			0.0012	0.20	1.03	2.03
0.5						1.30
0.55						0.87
0.6						0.64
0.65						0.27
0.7						0.081

Table 4
Helium
 f_0 in units of 10^{-42}

Energy, ev	F/P_0										
	0.01	0.025	0.05	0.1	0.2	0.5	1.0	2.0	2.5	3.0	4
0.001	810	204	211								
0.002	805										
0.003	796										
0.004	784										
0.005	769	202	210	25.6	9.01	36.5					
0.006	751										
0.007	730										
0.008	707										
0.009	682										
0.01	654	197	209	25.5	9.00	36.5					
0.012	595										
0.014	532										
0.016	467										
0.02	342	178	203	25.4	8.99	36.5					
0.025	211										
0.03	116	149	195	25.1	8.96	36.5					
0.035	57										
0.04	26	117	183	24.7	8.93	36.5					
0.05	3.7	86	170	24.2	8.89	36.4	0.814	0.258	0.00948	0.106	0.0241
0.06	0.35	59	154	23.7	8.83	36.4					
0.07	0.0203	37.5									
0.08	0.0008	22.3	121	22.3	8.70	36.3					
0.09		12.4									
0.1		6.4	89	20.6	8.53	36.2	0.813	0.258	0.0379	0.106	0.0241
0.115		2.1	67								
0.12		1.40	61								
0.15		0.084	31	15.7	7.98						
0.17			17.1								
0.2		0.00020	6.7	10.8	7.26	35.3	0.808	0.258	0.152	0.106	0.0241
0.22			3.2								
0.25		0.000023	0.95	6.6	6.43						
0.3			0.089	3.7	5.54	33.8	0.799	0.257			
0.4			0.00021	0.80	3.80	31.8	0.787	0.256			
0.5				0.116	2.34	29.4	0.772	0.255	0.164	0.106	0.0240
0.6				0.011	1.29	26.7	0.754	0.254			
0.7				0.00065	0.64	23.9	0.733	0.252			
0.8					0.21	21.0	0.710	0.250			
0.9					0.11	18.1	0.784	0.247			
1.0					0.041	15.4	0.656	0.245	0.160	0.104	0.0238
1.1					0.013		0.627	0.242			
1.2					0.0038		0.597	0.239			
1.3							0.566	0.236			
1.4							0.534	0.233			
1.5						5.2	0.501	0.229	0.153	0.101	0.0234
1.6					0.000048		0.469	0.225			
1.7							0.437	0.221			
1.75							2.6				
1.8							0.405	0.217			
1.9							0.374	0.213			
2.0						1.15	0.343	0.208	0.144	0.0966	0.0229
2.25						0.39	0.270	0.196			
2.5						0.151	0.207	0.183	0.133	0.0913	0.0222
3						0.012	0.110	0.157	0.120	0.0851	0.0213
3.5						0.0061	0.053	0.130	0.107	0.0784	0.0203
4.0							0.023	0.106	0.094	0.0716	0.0193
4.5							0.0097	0.085	0.082	0.0650	0.0183
5							0.0038	0.068	0.070	0.0586	0.0173
5.5							0.0015	0.053	0.060	0.0527	0.0163
6.0							0.00055	0.042	0.052	0.0473	0.0153
7								0.038	0.038	0.0381	0.0135
7.5							0.000030	0.0292			
8								0.0157	0.0276	0.0306	0.0120
8.5										0.0275	0.0113
9										0.0278	0.0106
10							0.00000022	0.0060	0.0119	0.0209	0.0094
12								0.0023	0.0080	0.0130	0.0074
15								0.00056			
16								0.00034	0.0023	0.0055	0.0046
20								0.000064	0.00081	0.0027	0.0030
25								0.000028	0.0012	0.00196	0.00196
30								0.000124	0.00072	0.00145	0.00145
40								0.000046	0.00037	0.00099	0.00099
50									0.00024	0.00079	0.00079
60									0.000018	0.00018	0.00068
70										0.00016	0.00062
80										0.00015	0.00057
85									0.000011	0.00012	0.00056

Table 5
Neon
 f_0 in units of 10^{-42}

Energy, ev	E/p ₀							
	0.02	0.1	0.2	0.4	0.6	0.8	1.0	1.5
0.05	10.1							
0.1	9.05	1.01	0.402	0.163	0.0970	0.0676	0.0520	0.0318
0.2	7.38	1.00	0.401	0.162	0.0970	0.0676	0.0520	
0.3	5.66							
0.4	3.84	0.975	0.399	0.162	0.0969	0.0676	0.0520	0.0318
0.5	2.40	0.957	0.397	0.162	0.0969	0.0675		
0.6	1.18	0.930	0.394	0.162	0.0968	0.0675	0.0520	0.0318
0.8	0.179	0.863	0.387	0.161	0.0966	0.0674	0.0520	0.0318
1.0	0.018	0.786	0.378	0.160	0.0963	0.0673	0.0519	0.0318
1.25	0.00036	0.673	0.363	0.159	0.0959	0.0671		
1.50		0.554	0.346	0.157	0.0954	0.0669	0.0517	
1.75		0.433	0.325	0.151	0.0947	0.0667		
2.0		0.321	0.302	0.151	0.0940	0.0664	0.0515	0.0317
2.5		0.152	0.250	0.145	0.0920	0.0656		
3.0		0.059	0.197	0.136	0.0896	0.0646	0.0506	0.0314
3.5		0.0180	0.147	0.127	0.0867	0.0634		
4.0		0.0043	0.103	0.116	0.0833	0.0620	0.0492	0.0311
4.5		0.00078	0.067	0.104	0.0795	0.0604		
5.0		0.00010	0.041	0.092	0.0752	0.0586	0.0475	0.0306
5.5		0.000010	0.0226	0.080	0.0705	0.0564		
6.0			0.0125	0.068	0.0659	0.0544	0.0453	0.0299
6.5			0.0061	0.057	0.061	0.0520		
7.0			0.0028	0.0471	0.056	0.0496	0.0427	0.0292
7.5			0.00114	0.0376	0.051	0.0468		
8.0			0.00046	0.0300	0.0457	0.0443	0.0397	0.0282
9.0			0.000052	0.0175	0.0360	0.0387	0.0364	0.0272
10.0			0.0000041	0.0092	0.0271	0.0329	0.0329	0.0260
11.0				0.0044	0.0195	0.0274	0.0292	0.0246
12.0				0.0019	0.0133	0.0222	0.0254	0.0232
13.0				0.00069	0.0085	0.0172	0.0217	0.0215
14.0				0.00019	0.0049	0.0126	0.0177	
15.0				0.000060	0.0029	0.0094	0.0148	
15.9				0.000016	0.00165	0.0068	0.0120	0.0166
17						0.0045	0.0092	
18					0.00050	0.0035	0.0078	0.0137
20					0.000091	0.00135	0.0042	0.0105
22					0.000011	0.00041	0.00198	0.0075
24					0.0000010	0.000106	0.00084	0.0051
26						0.000024	0.00033	0.00335
28						0.000047	0.000119	0.00214
30						0.0000009	0.000041	0.00132
32							0.0000109	0.00075
34							0.0000031	0.00043
36							0.00000079	0.00023
38								0.000124
40								0.000063
42								0.000031
44								0.000016
46								0.0000076
48								0.0000038
52								0.0000009
56								0.00000023
0.02	6.51*							
0.04	6.51							
0.06	6.50							
0.08	6.48							
0.1	6.45							
0.15	6.33							
0.2	6.11							
0.3	5.32							
0.4	4.16							
0.5	2.88							
0.6	1.79							
0.7	0.92							
0.8	0.416							
0.9	0.161							
1.0	0.051							
1.25	0.0013							

*The transport cross sections, as determined from microwave experiments⁽⁵⁾ were used to compute the values in this column

Table 6
Argon

f_0 in units of 10^{-42}

Energy, ev	E/P ₀									
	0.005	0.01	0.1	0.2	0.4	0.6	0.8	1.0	1.2	1.5
0.02	12.5	6.24	0.867	0.546	0.326	0.242	0.196	0.166	0.145	0.123
0.04	12.5	6.23	"	"	"	"	"	"	"	"
0.06	12.4	6.22	"	"	"	"	"	"	"	"
0.08	12.3	6.20	"	"	"	"	"	"	"	"
0.1	12.1	6.18	"	"	"	"	"	"	"	"
0.15	11.4	6.09	"	"	"	"	"	"	"	"
0.2	10.2	5.93	"	"	"	"	"	"	"	"
0.25	8.69	5.69	"	"	"	"	"	"	"	"
0.3	6.86	5.36	"	"	"	"	"	"	"	"
0.35	4.94	4.91	"	"	"	"	"	"	"	"
0.4	3.19	4.33	"	"	"	"	"	"	"	"
0.45	1.81	3.84	"	"	"	"	"	"	"	"
0.5	0.78	3.12	0.865	"	"	"	"	"	"	"
0.55	0.36	2.56	"	"	"	"	"	"	"	"
0.6	0.119	1.95	"	0.546	"	"	"	"	"	"
0.65	0.031	1.39	"	"	"	"	"	"	"	"
0.7	0.0062	0.93	"	"	"	"	"	"	"	"
0.75	0.00096	0.58	"	"	"	"	"	"	"	"
0.8	0.00011	0.34	"	0.546	"	"	"	"	"	"
0.85	"	0.18	"	"	"	"	"	"	"	"
0.9	"	0.084	"	"	"	"	"	"	"	"
0.95	"	0.036	"	"	"	"	"	"	"	"
1.0	"	0.029	"	"	"	"	"	"	"	"
1.25	"	0.000012	0.822	0.539	0.324	0.241	"	"	"	"
1.5	"	"	0.761	0.528	0.321	"	0.195	0.166	"	0.0718
1.75	"	"	0.669	0.512	0.316	0.239	"	0.165	0.144	0.122
2.0	"	"	0.53	0.483	0.310	0.237	0.194	0.164	"	"
2.25	"	"	0.380	0.444	0.301	0.234	0.192	"	"	"
2.5	"	"	0.236	0.394	0.289	0.230	0.190	"	"	"
3.0	"	"	0.123	0.335	0.230	0.207	0.180	0.157	0.140	0.120
3.5	"	"	0.032	0.135	0.176	0.184	0.168	0.142	0.130	0.114
4.0	"	"	0.000048	0.046	0.121	0.156	0.153	0.114	0.111	0.104
4.5	"	"	"	0.101	0.067	0.120	0.132	"	"	"
5.0	"	"	"	0.030	0.030	0.084	0.108	0.075	0.084	0.0860
5.5	"	"	"	0.0098	0.051	0.082	0.082	0.0361	0.0502	0.0622
6.0	"	"	"	0.026	0.022	0.056	0.056	0.0119	0.0232	0.0339
7.0	"	"	"	0.0035	0.0002	0.0035	0.0032	0.0023	0.0074	0.0183
8.0	"	"	"	"	"	0.00016	0.00018	0.00027	0.0016	0.0069
9.0	"	"	"	"	"	0.000009	0.000009	0.000019	0.00027	0.0102
10.0	"	"	"	"	"	"	"	0.0000010	0.000033	0.00058
11.0	"	"	"	"	"	"	"	"	"	"
12.0	"	"	"	"	"	"	"	"	"	"
12.5	"	"	"	"	"	"	"	"	"	"

In these calculations, the following $\sigma_t(\epsilon)$ data were used: for nitrogen, the microwave values of Phelps et al.,⁽²⁾ over the energy interval 0 to 0.5 ev; for helium, Barbieri's⁽³⁾ values from 1.8 to 19 ev, the Graham-Ruhlig⁽⁴⁾ values above 19 ev, and the extrapolated value of 22.9 cm^{-1} from 0 to 1.8 ev; for neon, Barbieri's values from 1 ev to 16 ev, the Graham-Ruhlig values above 16 ev, and our extrapolated curve below 1.8 ev which intercepted the zero energy axis at $\sigma_t = 5.2 \text{ cm}^{-1}$; for argon, Barbieri's values from 1.1 ev to 12.5 ev and our extrapolated curve below 1 ev which intercepted the axis at $\sigma_t = 1.25 \text{ cm}^{-1}$. All $\sigma_t(\epsilon)$ values used were at 0°C . and 1 mm Hg pressure.

C. Computation of $\bar{W}$

Values of $\sigma_t(\epsilon)\epsilon^2 f_0(\epsilon)$ were plotted against ϵ for each E/p_0 , and the area under the curve (see Eq. 7) was used to compute $\bar{W}$. These values are tabulated in Tables 7 to 10.

Table 7

Nitrogen

Calculated Values

E/p_0 , volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	A (gram/ev) ^{3/2} in units of 10^{-42}
0.01	0.055	103
0.02	0.070	53.2
0.04	0.089	26.6
0.06	0.107	18.0
0.08	0.119	13.0
0.10	0.131	10.2

Table 8

Helium

Calculated Values

E/p_0 , volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	A (gram/ev) ^{3/2} in units of 10^{-42}
0.01	0.086	812
0.025	0.135	204
0.05	0.190	72
0.1	0.270	25.6
0.2	0.381	9.01
0.5	0.594	2.27
1.0	0.843	0.815
2.0	1.27	0.258
2.5	1.56	0.166
3.0	2.03	0.106
4.0	5.71	0.0241

Table 9

Neon

Calculated Values

E/p_0 , volts/cm/mm Hg	$\bar{W}$, cm/ sec	A (gram/ev) ^{3/2} in units of 10^{-42}
0.02*	0.169	6.51
0.02	0.162	10.7
0.1	0.328	1.02
0.2	0.448	0.402
0.4	0.600	0.163
0.6	0.708	0.097
0.8	0.784	0.068
1.0	0.872	0.052
1.5	1.02	0.032

* The microwave cross sections (5) were used in this calculation.

Table 10

Argon

Calculated Values

E/p_0 volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	A (gram/ev) ^{3/2} $\times 10^{42}$
0.005	0.097	12.6
0.01	0.121	6.24
0.1	0.214	0.867
0.2	0.234	0.546
0.4	0.289	0.326
0.6	0.327	0.242
0.8	0.353	0.196
1.0	0.372	0.166
1.2	0.392	0.145
1.5	0.417	0.123
2.0	0.452	0.0989
3.0	0.523	0.0718

All integrations were performed with a planimeter, and readings reproducible to within 1% were obtained. The over-all accuracy of the calculations is estimated at 5%.

D. Experimental Values of $\bar{W}$

Experimental values of $\bar{W}$ for nitrogen and the noble gases, presented in Report ANL-5679, are presented here in tabulated form in Tables 11 to 16. The error assigned to each value is that associated with the time measurement.

E. Drift Velocity in the Region of Inelastic Collisions

Calculation of $\bar{W}$, taking into account inelastic collisions, leads to differential equations for the distribution function, $f(\epsilon)$, which are difficult to handle. In addition, one is required to make assumptions concerning the excitation cross sections since these data required for the computation are lacking. For these reasons, work on this problem has been postponed.

In the last semiannual report (ANL-5755) it was indicated that information concerning the excitation $\sigma(\epsilon)$ could perhaps be extracted from the experimental $\bar{W}$ curves. It seems clear that only information concerning some kind of an average behavior could be inferred, but due to the mathematical complexity it is not evident how best to proceed.

Table 11

Nitrogen

Experimental values

$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 653$ mm Hg		$p_0 = 653$ mm Hg	
0.0138	0.126 ± 0.001	0.640	0.538 ± 0.011
0.0335	0.212 ± 0.003	0.664	0.552 ± 0.012
0.0417	0.222 ± 0.004	0.690	0.560 ± 0.012
0.0586	0.267 ± 0.004	0.697	0.565 ± 0.013
0.0695	0.278 ± 0.005	0.755	0.596 ± 0.014
0.0864	0.291 ± 0.006	0.814	0.618 ± 0.015
0.0973	0.292 ± 0.006	0.871	0.629 ± 0.016
0.114	0.305 ± 0.007	0.930	0.670 ± 0.018
0.125	0.310 ± 0.007	0.988	0.690 ± 0.019
0.142	0.321 ± 0.007	1.047	0.711 ± 0.020
0.153	0.328 ± 0.008	1.047	0.721 ± 0.021
0.186	0.346 ± 0.005	1.102	0.740 ± 0.022
0.235	0.364 ± 0.005	1.161	0.762 ± 0.023
0.242	0.376 ± 0.006	1.220	0.804 ± 0.025
0.269	0.392 ± 0.006	1.279	0.814 ± 0.026
0.298	0.405 ± 0.007	1.338	0.855 ± 0.029
0.325	0.417 ± 0.007	1.395	0.870 ± 0.030
0.369	0.430 ± 0.007	1.452	0.885 ± 0.031
0.391	0.446 ± 0.008	1.511	0.930 ± 0.034
0.425	0.456 ± 0.008	1.570	0.930 ± 0.034
0.452	0.465 ± 0.009	1.625	0.952 ± 0.036
0.480	0.484 ± 0.009	1.682	0.988 ± 0.039
0.480	0.475 ± 0.009	1.742	1.020 ± 0.041
0.507	0.487 ± 0.009	1.801	1.031 ± 0.042
0.551	0.505 ± 0.010	1.860	1.078 ± 0.045
0.580	0.520 ± 0.011	1.918	1.102 ± 0.048
0.581	0.517 ± 0.011	1.975	1.102 ± 0.048
0.608	0.529 ± 0.011	2.035	1.133 ± 0.051
0.636	0.536 ± 0.011		

Table 12

Helium

Experimental values

$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 615$ mm Hg			
0.0031	0.0298 ± 0.0002	0.311	0.431 ± 0.004
0.0055	0.0397 ± 0.0003	0.314	0.451 ± 0.005
0.0080	0.0504 ± 0.0005	0.373	0.475 ± 0.005
0.0129	0.0616 ± 0.0005	0.417	0.498 ± 0.006
0.0154	0.0789 ± 0.0007	0.513	0.546 ± 0.007
$p_0 = 248$ mm Hg			
		0.543	0.579 ± 0.008
		0.594	0.585 ± 0.008
0.0194	0.0860 ± 0.0009	0.619	0.596 ± 0.008
0.0254	0.106 ± 0.001	0.665	0.616 ± 0.009
0.0314	0.123 ± 0.001	0.695	0.626 ± 0.009
0.0374	0.141 ± 0.002	0.741	0.654 ± 0.010
0.0435	0.153 ± 0.002	0.770	0.665 ± 0.011
0.0495	0.164 ± 0.002	0.816	0.689 ± 0.011
0.0555	0.174 ± 0.002	0.846	0.705 ± 0.012
0.0616	0.186 ± 0.003	0.891	0.720 ± 0.012
0.0676	0.198 ± 0.003	0.925	0.731 ± 0.013
0.0832	0.223 ± 0.004	1.010	0.774 ± 0.014
0.113	0.259 ± 0.005	1.040	0.794 ± 0.015
0.159	0.313 ± 0.008	1.085	0.814 ± 0.016
0.189	0.336 ± 0.009	$p_0 = 112$ mm Hg	
0.234	0.377 ± 0.003		
0.234	0.380 ± 0.003	1.113	0.846 ± 0.028
0.265	0.408 ± 0.004	1.282	0.900 ± 0.032

Table 13

Neon

Experimental values

$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 118$ mm Hg			
0.0142	0.162 ± 0.003	0.335	0.555 ± 0.010
0.0258	0.188 ± 0.004	0.391	0.589 ± 0.014
0.0375	0.214 ± 0.005	0.495	0.640 ± 0.016
0.0492	0.238 ± 0.007	0.553	0.677 ± 0.018
0.0608	0.260 ± 0.008	0.655	0.715 ± 0.020
0.0725	0.282 ± 0.009	$p_0 = 169$ mm Hg	
0.0842	0.310 ± 0.011		
0.0958	0.334 ± 0.013	0.749	0.817 ± 0.013
0.108	0.348 ± 0.014	0.791	0.867 ± 0.015
0.119	0.371 ± 0.017	0.859	0.917 ± 0.017
0.131	0.388 ± 0.018	0.901	0.996 ± 0.020
0.174	0.447 ± 0.008	0.968	1.024 ± 0.020
0.233	0.493 ± 0.009	1.010	1.076 ± 0.022

Table 14

Argon

Experimental values

E/p_0 volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	E/p_0 volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 577$ mm Hg			
0.0065	0.0991 ± 0.001	0.606	0.324 ± 0.008
0.0093	0.166 ± 0.002	0.659	0.330 ± 0.009
0.0122	0.123 ± 0.002	0.684	0.334 ± 0.009
0.0152	0.130 ± 0.002	0.693	0.334 ± 0.009
0.0180	0.133 ± 0.001	0.724	0.338 ± 0.009
0.0208	0.133 ± 0.001	0.752	0.338 ± 0.009
0.0237	0.141 ± 0.002	0.756	0.344 ± 0.009
0.0266	0.146 ± 0.002	0.789	0.344 ± 0.010
0.0294	0.151 ± 0.002	0.818	0.346 ± 0.010
0.0323	0.154 ± 0.002	0.820	0.350 ± 0.010
0.0509	0.177 ± 0.002	0.888	0.353 ± 0.010
0.0692	0.192 ± 0.003	0.957	0.363 ± 0.011
0.0835	0.200 ± 0.003	1.025	0.368 ± 0.011
0.102	0.215 ± 0.004	1.093	0.374 ± 0.011
0.116	0.223 ± 0.004	1.161	0.375 ± 0.011
0.135	0.231 ± 0.004	1.231	0.384 ± 0.012
0.149	0.238 ± 0.004	1.298	0.385 ± 0.012
0.167	0.246 ± 0.005	1.368	0.391 ± 0.012
0.182	0.249 ± 0.005	1.435	0.394 ± 0.012
0.221	0.264 ± 0.006	1.503	0.406 ± 0.013
0.256	0.268 ± 0.006	1.572	0.413 ± 0.014
0.289	0.274 ± 0.006	$p_0 = 94.8$ mm Hg	
0.321	0.282 ± 0.006		
0.354	0.290 ± 0.007		0.445 ± 0.015
0.373	0.302 ± 0.007		0.470 ± 0.018
0.443	0.304 ± 0.007		0.530 ± 0.011
0.476	0.306 ± 0.008		0.576 ± 0.013
0.508	0.313 ± 0.008		0.635 ± 0.016
0.541	0.318 ± 0.008		0.701 ± 0.020
0.573	0.322 ± 0.008		0.746 ± 0.022
			0.820 ± 0.027

Table 15

Krypton

Experimental values

$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 302$ mm Hg		$p_0 = 302$ mm Hg	
0.0113	0.0896 ± 0.0018	0.834	0.222 ± 0.005
0.0162	0.0967 ± 0.0020	0.860	0.226 ± 0.006
0.0212	0.107 ± 0.003	0.896	0.228 ± 0.006
0.0262	0.112 ± 0.003	0.921	0.230 ± 0.006
0.0361	0.116 ± 0.003	0.965	0.233 ± 0.006
0.0410	0.119 ± 0.003	0.984	0.234 ± 0.006
0.0460	0.122 ± 0.003	1.048	0.239 ± 0.006
0.0510	0.123 ± 0.003	1.087	0.242 ± 0.006
0.0560	0.125 ± 0.003	1.110	0.246 ± 0.006
0.0691	0.131 ± 0.002	1.148	0.252 ± 0.007
0.0940	0.140 ± 0.002	1.172	0.252 ± 0.007
0.132	0.150 ± 0.002	1.245	0.262 ± 0.007
0.157	0.156 ± 0.003	1.270	0.266 ± 0.008
0.195	0.163 ± 0.003	1.309	0.270 ± 0.008
0.220	0.167 ± 0.003	1.331	0.273 ± 0.008
0.258	0.172 ± 0.003	1.370	0.281 ± 0.009
0.283	0.175 ± 0.003	1.392	0.282 ± 0.009
0.320	0.180 ± 0.003	1.432	0.289 ± 0.009
0.346	0.183 ± 0.004	1.459	0.294 ± 0.009
0.422	0.190 ± 0.004	1.459	0.297 ± 0.009
0.446	0.192 ± 0.004	1.499	0.299 ± 0.009
0.485	0.192 ± 0.004	1.521	0.308 ± 0.010
0.509	0.198 ± 0.004	1.56	0.312 ± 0.011
0.560	0.201 ± 0.004	1.585	0.324 ± 0.011
0.572	0.204 ± 0.004	1.700	0.348 ± 0.013
0.611	0.209 ± 0.005	1.830	0.369 ± 0.008
0.636	0.208 ± 0.005	1.960	0.391 ± 0.009
0.674	0.211 ± 0.005	2.090	0.429 ± 0.011
0.699	0.213 ± 0.005	2.090	0.425 ± 0.011
0.736	0.217 ± 0.005	2.221	0.440 ± 0.011
0.761	0.218 ± 0.005	2.355	0.486 ± 0.013

Table 16

Xenon

Experimental values

$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec	$\frac{E}{p_0}$ volts/cm/mm Hg	$\bar{W}$, cm/ μ sec
$p_0 = 267$ mm Hg		$p_0 = 267$ mm Hg	
0.0293	0.0745 ± 0.001	1.085	0.171 ± 0.003
0.0349	0.0790 ± 0.001	1.155	0.175 ± 0.003
0.0406	0.0802 ± 0.001	1.225	0.179 ± 0.003
0.0406	0.0815 ± 0.001	1.298	0.186 ± 0.004
0.0462	0.0854 ± 0.001	1.410	0.199 ± 0.004
0.0518	0.0871 ± 0.002	1.480	0.209 ± 0.005
0.0574	0.0884 ± 0.002	1.482	0.209 ± 0.005
0.0631	0.0909 ± 0.002	1.552	0.218 ± 0.005
0.0777	0.0943 ± 0.002	1.622	0.228 ± 0.005
0.106	0.0996 ± 0.002	1.629	0.228 ± 0.005
0.106	0.101 ± 0.002	1.695	0.238 ± 0.006
0.148	0.105 ± 0.002	1.768	0.250 ± 0.007
0.177	0.110 ± 0.002	1.775	0.250 ± 0.007
0.220	0.113 ± 0.003	1.924	0.275 ± 0.008
0.248	0.117 ± 0.003	2.070	0.300 ± 0.010
0.291	0.119 ± 0.003	2.220	0.321 ± 0.011
0.318	0.123 ± 0.003	2.370	0.349 ± 0.005
0.361	0.125 ± 0.003	2.580	0.376 ± 0.006
0.389	0.127 ± 0.003	2.663	0.395 ± 0.006
0.475	0.133 ± 0.002	2.810	0.421 ± 0.007
0.547	0.137 ± 0.002	2.960	0.441 ± 0.008
0.618	0.142 ± 0.002	3.110	0.465 ± 0.009
0.688	0.145 ± 0.002	3.260	0.490 ± 0.010
0.760	0.149 ± 0.002	3.410	0.510 ± 0.010
0.832	0.154 ± 0.003	3.550	0.536 ± 0.011
0.941	0.160 ± 0.003	3.700	0.555 ± 0.012
1.012	0.164 ± 0.003	3.850	0.575 ± 0.013
1.083	0.168 ± 0.003	3.990	0.600 ± 0.014

Some consideration has been given to the problem of measuring, by a straightforward method, the absolute excitation $\sigma(\epsilon)$ of the individual levels in the region of the threshold energy. That these measurements are not available is an affirmation of their difficulty. In addition to the more usual problems associated with scattering experiments, difficulties involving the production and detection of low energy (~ 1 to 20 eV) electrons are encountered. Analysis of the inelastically scattered electrons, which constitute about 1% of the total scattered current, presents additional difficulties. However, because of the importance of these measurements, both theoretically and experimentally, further study, directed to overcome the experimental difficulties, is planned.

F. Publication of Results

Publication of a paper in the open literature describing our experimental $\bar{W}$ results has been postponed so as to include the results of our calculations. A revised manuscript is now in the final stages of preparation.

G. Transport Cross-Section Data

The transport cross-section data used in our calculations are presented in Figs. 7 to 10 to provide a convenient reference and to facilitate their comparison with other data. It should be pointed out that the $\sigma_t(\epsilon)$ computations of Graham and Ruhlig were made using a set of phase shifts determined from experimental data, including the scattering data used by Barbieri. Evidently, the relative weighting of the scattering measurements in the determination of the phase shifts was such as to result in the $\sigma_t(\epsilon)$ differences shown in the figures.

For our calculations, we plotted Barbieri's $\sigma_t(\epsilon)$ values and drew an average curve through them. These curves were extrapolated to zero energy as shown in the figures. At higher energy, the Graham-Ruhlig $\sigma_t(\epsilon)$ values were used.

The $\sigma_t(\epsilon)$ values of Graham and Ruhlig and of Brown et al., which are presented in the figures, were obtained from their published curves. An unspecified error is associated with the process of transferring such data.

For helium, best agreement between experimental and calculated $\bar{W}$ up to $E/p_0 = 1.3$ was obtained by assuming $\sigma_t = 28 \pm 2 \text{ cm}^2$ in the energy range 0 to 4 eV. The circled points in Fig. 8 were computed by us, using the Westin⁽⁶⁾ phase shifts, and thereby served as a check on the Graham-Ruhlig computation. The cross sections of Brown et al.,⁽⁷⁾ for helium, gave $\bar{W}$ values about 20% higher than experiments.

Fig. 7. Transport cross-section data determined from microwave experiments by Brown et al. (2) for nitrogen.

Fig. 8. Comparison of transport cross-section data used in $\bar{W}$ calculations for helium. The value of $\sigma_t = 28 \text{ cm}^{-1}$ gave best agreement with experimental $\bar{W}$. The circled points were computed by author from Westin (6) phase shifts.

Fig. 9. Comparison of transport cross-section data used in $\bar{W}$ calculations for neon.

Fig. 10. Comparison of transport cross-section data used in $\bar{W}$ calculations for argon.

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REPORT ON A STUDY OF LIGHT PIPE DESIGNS FOR A SCINTILLATION SPECTROMETER

R. Grismore

A. Introduction

The purpose of the study was to determine the type and design of the light piping system which will most efficiently collect the gamma-ray excited fluorescent scintillations from a $1\frac{1}{4}$ -inch thick NaI(Tl) crystal 8 inches in diameter and transfer them to a $4\frac{1}{2}$ -inch diameter multiplier photocathode. Achievement of satisfactory energy resolution requires minimal loss of photons in the collection and transfer processes since gamma radiations with energies as low as 17 kev are to be detected. Uniformity of optical efficiency for all possible positions of occurrence of a scintillation throughout the crystal is also necessary for good energy resolution. In the latter regard it is to be noted that especial effort must be made to prevent the falling off of this efficiency in the outer zones of the crystal which are of relatively great importance because of the large fraction of the total volume which they represent.

B. Discussion of Various Systems Described in the Literature

Garwin⁽¹⁾ describes an arrangement in which a disc-shaped scintillator is viewed edge on through a light pipe with polished reflecting walls. The transition from the long, thin, rectangular area of the scintillator edge to the circular area required by the photomultiplier is made by "adiabatically" tapering the walls of the light pipe, that is by tapering them slowly and maintaining the cross-sectional area constant. It is demonstrated in Garwin's article that under these conditions a light ray will pass through the pipe without changing its angle of inclination to the axis. As a result, all light entering the pipe at an angle of inclination to the axis less than the critical angle of the pipe to air surface will be transmitted through the pipe by total internal reflection. A uniformity of optical efficiency to 1 per cent over the volume was achieved by a scintillation counter which Garwin built according to this principle. However, theory shows that a Lucite light pipe of this design would collect only approximately 25 per cent of the total light emitted, even with an aluminum reflector at the far edge of the crystal to return the rays which start out in the wrong direction. The 25 per cent figure can be increased somewhat by wrapping the outside of the light pipe with shiny aluminum foil, but Garwin himself indicates that a diffusely-reflecting pipe may be more desirable when 100 per cent collection of the emitted light is the goal. In addition to its low optical efficiency, the Garwin system has the added disadvantage that it would require a considerable length of light pipe (made of some material with refractive index greater than unity) in order to accomplish the transition from a rectangular area 8 inches by $1\frac{1}{4}$ inch to a $4\frac{1}{2}$ -inch diameter circular area using small angles of taper.

Tove⁽²⁾ describes a series of measurements on the transmission of light through various cylindrical guides, a collecting head being used to transfer the light from the crystal face to the guide. Two different collecting heads were used in these measurements, each of which is supposed to collect all of the light coming across its input surface instead of the 35 per cent collected by the cylindrical guide alone. The first head is a solid of revolution whose surface is defined by the equation of a logarithmic spiral. By properly choosing the constants of the spiral it is possible to ensure that a ray from any part of the crystal will strike any point of the collecting head surface at an angle of incidence greater than the critical and thus will be totally reflected. The second collecting head used in the Tove measurements, devised by Gerholm,⁽³⁾ has a special shape somewhat resembling a prolate spheroid. The method of generation of the surface is not described in the literature. Gerholm finds that this head actually collects about 70 per cent of the light coming from the crystal, and that for a total length of head plus cylindrical light guide of 22.5 cm an efficiency of nearly 60 per cent is realized.

Although Tove used both of the collecting heads described above to transfer light from a crystal face of given area to a light guide of greater cross-sectional area, the idea occurred that it might be possible to adapt one of them to the present reverse situation. However, the minimum length of logarithmic-spiral Lucite collecting head needed to connect an 8-inch diameter crystal with a $4\frac{1}{2}$ -inch diameter photocathode turns out to be 27 inches. The expanding section alone would be 10 inches long, and if used by itself would have to be followed by a length of truncated cone which again would make the total length prohibitive. Similarly, the Gerholm head would be much too long since even in the form used by Tove its length was 2 to 3 times as great as the diameter of the input face.

Thus, it appears that the answer to the present problem does not lie in a light pipe scheme based on the principle of total internal reflection. In fact, workers experienced in the field have recommended that in the case of a large crystal and a small photocathode, light pipes should be as short as possible, rather than long enough to produce a critical angle of reflection at the sides of the cone.⁽⁴⁾ Also, diffusely reflecting surfaces are to be preferred over specularly reflecting ones for the walls of the light pipe in the present case, judging by other experimenters' results.⁽⁵⁾ In the first place, a good diffusely reflecting surface, such as magnesium oxide with quaterphenyl evaporated on it, may be more than 90 per cent efficient,⁽⁶⁾ while the best specularly reflecting ones are limited to 87 per cent or less for the given wave length band.⁽²⁾ In addition, the diffusing action of the former type of surface aids in the collection of light trapped in out-of-the-way corners and in general makes the optical efficiency more nearly equal over the entire scintillator surface.

C. Deductions Concerning the Best Light Pipe Design

The above findings, together with the consideration that the most efficient collection of light from the given crystal can probably be accomplished through one of its large circular faces, leads to the hypothesis that the best light pipe for the present application will probably be an axially symmetric one with a diffusely reflecting surface which leads from one 8-inch diameter face of the crystal to the multiplier photocathode. Several possible shapes which such a system could assume are shown in Fig. 11

Fig. 11. Various Possible Light Pipe Shapes

Mention of the use of truncated cone shaped light pipes (Fig. 11-a) is to be found in the literature,^(7,8) and the observation is made that the efficiency of light collection increases rapidly as the angle of the cone decreases.⁽⁸⁾ However, no data could be found on the effect of varying the wall shape, as in Figs. 11-b through 11-e. Intuition seems to indicate, though, that the straight cone of Fig. 11-a would be the most efficient. Any design which carries the light pipe surface inside the limits of the

straight cone, as in Figs. 11-b and 11-d, would certainly be disadvantageous since this would have the effect of materially decreasing the photocathode surface area visible from points in the critical outer zones of the crystal and so would certainly lower the optical efficiency there. On the other hand, a design such as that of Fig. 11-c or 11-e which stays outside the limits of the truncated cone would improve the transmission in that portion of the pipe near the crystal by decreasing the cone angle but only at the expense of that in another portion farther along where the cone angle must be increased. This would seem undesirable for the following reason. The influence of the walls becomes more important as the diameter of the light pipe decreases since in this case the area presented to the oncoming radiation by walls of a certain inclination in a given increment of pipe length becomes a larger fraction of the total area seen by that radiation. Hence, it would seem most advantageous to keep the cone angle as small as possible in the regions of smaller cross-sectional area, so that designs such as those in Figs. 11-c and 11-e are ruled out. Thus, on the basis of the two arguments the straight truncated cone seems preferable.

The optimum length for the cone will have to be determined by experiment in the absence of a full-scale effort to calculate light pipe transmission. The forward transmission properties of the walls improve as the length is increased owing to the decreased cone angle, but absorption losses also increase. Increasing the length of a diffuse light pipe decreases the non-uniformity of optical efficiency, but on the other hand, one cannot conduct light many diameters in such a pipe.⁽¹⁾ A length of the order of one crystal diameter or slightly less probably would be best.

D. Conclusions

A light pipe of the type shown in Fig. 12 is suggested for the present application. Unless internal lenses are used as suggested by Dr. G. S. Monk,⁽⁹⁾ the light pipe should be made of some material which will provide optical coupling between crystal and photomultiplier.⁽⁴⁾ The use of silicone oil as a light pipe material should be considered as Tove⁽²⁾ found that it allows better transmission than a quartz light pipe.

Fig. 12. Suggested Light Pipe Design

The superiority of the truncated cone shaped light pipe over other possible forms is not proven. The optimum length must also be determined by experiment. Calculation of the transmission properties of various forms and sizes of light pipes might be carried out by Monte Carlo methods on a computer, or possibly by extension of the methods which have been used to calculate the flow of rarefied gases through pipes with perfectly diffusing walls.⁽¹⁰⁾

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VARIABLE ENERGY NEUTRON SOURCE

PART I. EXPERIMENTAL PROGRAM

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A. Introduction,

The development of a simple source of monoenergetic fast neutrons, variable within the energy range of biological interest, would aid in the advancement of the field of neutron dosimetry as well as promote the study of instruments under development in the Radiological Physics Division.

The range of such a source should, of course, cover those energies encountered in most laboratory situations. The approximate energy range of prompt fission neutrons, 50 kev to 17 Mev, would be sufficient. It is not necessary for the source to be continuously variable over this range, but if only a few discrete energy values are available they should be spaced as close as 100 kev apart at the low end where the greatest amount of work needs to be done. The neutrons supplied at any given energy will be sufficiently monoenergetic for the purpose stated if the spread between five per cent intensity points is thirty per cent or less of the peak energy. Only moderate intensity under good geometry conditions is required of the source since most dosimeters are designed to operate at low fluxes. However, it is important that any accompanying gamma radiation be of low intensity. Reasonable initial cost and ease of operation and maintenance are, of course, necessary if the source is to be within reach of small laboratories. Employment of pulsing or coincidence techniques would be undesirable since some dosimeters, such as ionization chambers, are not easily adapted to short-interval operation.

B. The Moderator-Type Source

A group of Russian scientists⁽¹⁾ has built a neutron spectrometer utilizing the fact that a short burst of neutrons introduced into a heavy elastic moderator will develop a comparatively narrow energy spectrum. This spectrum degrades as an entity toward lower energies as time passes. The pulsing techniques employed in the Russian device preclude adopting it as the desired variable energy source per se. However, it seemed possible that the steady-state spectrum emerging from a not-too-thick block of the moderator might still be sufficiently monoenergetic to be useful. Thus, a simple source of monoenergetic neutrons, such as a low-voltage deuteron accelerator with deuterium and tritium targets, plus a moderator whose thickness could be varied, might provide a variable energy neutron source.

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Preliminary calculations indicated that graphite should prove to be a suitable moderator. Although the mass number is not high, the mean fractional energy loss per neutron collision for carbon is only about $1/6$, which is small enough so that the energies should stay reasonably well bunched during moderation. In addition, the comparatively small mass number and neutron scattering length make it possible to obtain the same degree of moderation with a considerably smaller volume than would be required of many other suitable materials (see Eq. 4, Part II of this report). A further advantage of carbon is the absence of nuclear energy levels below 4.4 Mev, which rules out large energy loss by inelastic scattering for D-D neutrons.

Accordingly, it was decided to explore the neutron spectrum emerging from a cubical graphite moderator with a monoenergetic neutron source at the center. A length of ten inches was chosen for the cube half-side, this representing a minimum thickness of graphite of roughly ten mean free paths. The neutrons supplied were of average energy 2.6 Mev from the D-D reaction, or optionally, of average energy 14 Mev from the D-T reaction. The neutron-producing targets were located at the cube center on the end of an extension to the beam tube of the 250-kev Cockcroft-Walton linear accelerator. The energy spread of the neutrons introduced into the graphite was about ± 0.5 Mev in the case of the D-D neutrons, and ± 1.0 Mev in that of the D-T neutrons. The several different methods which were used to measure the neutron spectra emerging from the graphite cube are given in the following sections.

C. Measurements with the Perlow Neutron Spectrometer

A proportional counter telescope-type neutron spectrometer recently developed by Dr. G. J. Perlow⁽²⁾ of the Physics Division was graciously loaned by him for use in measuring the moderated D-D neutron spectrum. It could not be used for the D-T spectrum because its useful range is limited to 50 kev to 1 Mev. The results obtained with this spectrometer, representing 33.5 hours of running time with a deuteron current of 25 microamperes to the deuterium-zirconium target, are shown in Fig. 13. The spectrum is given without the subtraction of any background. It has been corrected for the efficiency of the spectrometer using a smooth curve drawn through experimental efficiency values determined by the Reactor Engineering Division.⁽³⁾ The error bars attached to the points in Fig. 13 indicate their statistical standard deviations only.

The proportional counters of the Perlow spectrometer were operated at a gas pressure of 13.69 cm of Hg (referred to 0°C) and a voltage of 2850 in taking the data for Fig. 13. The counters were placed directly in front of the graphite cube in line with the projection of the accelerator beam tube, the forward counter being 20 inches from the cube face. The source-counter distance was thus equal to the source span. This choice of distance

Fig. 13. Spectrum measured with the Perlow spectrometer for the neutrons emerging from a 20-inch graphite cube enclosing a D-D source.

Fig. 14. Proton recoil spectrum obtained with the "twin" scintillation fast neutron detector from neutrons emerging from a 20-inch graphite cube enclosing a D-D source.

was a compromise between the fundamental requirement that the spectrometer be exposed only to collimated neutrons, and the desire to move it as close to the source as possible to make up for the low efficiency.

D. Measurements with the "Twin" Scintillation Fast Neutron Detector

A second measurement of the moderated D-D spectrum was made using a "twin" scintillation fast neutron detector of the type developed by Berlman and Marinelli.⁽⁴⁾ In this form of spectrometer two similar liquid scintillators are used, one of which is much richer in hydrogen atoms than the other. The two are adjusted to give identical response to gamma rays, the ratio of solute concentrations being varied to match pulse heights and the ratio of scintillator volumes to match counting rates. The difference pulse height spectrum obtained by subtracting the response of the one from that of the other in a mixed radiation field is then a measure of the proton recoil spectrum due to the neutron flux.

Inasmuch as it was desired to measure the graphite-moderated D-D spectrum down to around 100 kev where photomultiplier noise would undoubtedly cause serious distortion, it was decided to view each scintillator with two photomultipliers operating in coincidence. The two outputs were summed, after which the resulting pulse was amplified and fed to an Argonne 256-channel pulse height analyzer. A high degree of stability of optical contact between scintillators and photomultipliers is required to maintain a good match between the two scintillation cells over the long neutron runs. Hence, it was found necessary to immerse the photomultipliers into the scintillation fluids directly. This also improves the light collection efficiency. Each scintillation fluid cell was constructed by bonding a section of glass cylinder between the end windows of two Type 6292 photomultipliers with R-313.* Expansion tubes were provided so that the cells might always be completely full of scintillator. The exterior surfaces of the cells were given 3 coats of Tygon paint to provide suitable reflective surfaces. The hydrogen-rich cell was 37 mm in diameter by 32 mm long and contained a solution of 0.28 g/l PPO (diphenyloxazole) in commercial xylene. The hydrogen-deficient cell was 32 mm in diameter by 31 mm long and contained a solution of 4 g/l PPO in triple-distilled 1,4 bis-(trifluoromethyl) benzene. These two scintillators yielded identical spectra for Cs¹³⁷ gamma rays over long periods of time when exposed in the time ratio of 1.16 for the ordinary xylene to 1 for the fluorinated xylene.

The proton recoil spectrum obtained with this pair of cells for the graphite-moderated D-D source is shown in Fig. 14. Each cell was exposed in turn on the projection of the accelerator beam tube at 40 inches from the

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

graphite cube face. Deuteron currents of 20 microamperes to the neutron-producing target were used, the ordinary xylene counter being exposed for 5 hours and the fluorinated xylene counter for 4 hours and 19 minutes. Sixteen-hour background runs without deuteron current were taken with each cell in position and the correct fraction of each background spectrum was subtracted from the corresponding bombardment spectrum. The net fluorinated xylene spectrum was then subtracted from the net ordinary xylene spectrum to yield the curve of Fig. 14. The data have been combined into 5-channel boxes in order to improve the statistical accuracy. The data from the first 11 channels of the 256-channel analyzer have been discarded since coincidence circuit effects distort this portion of the spectrum. The error bar attached to each point in Fig. 14 represents the result of combining the statistical standard deviations of the corresponding points from the two bombardment runs and two background runs. Photomultiplier voltages in the neighborhood of 1350 were used in these runs.

The relationship between the proton recoil spectrum and the neutron flux spectrum for a device such as the "twin" scintillation spectrometer is

$$n(E) = -k \frac{E}{\sigma(E)} \cdot \frac{dN(E)}{dE} \quad (1)$$

where $n(E)$ is the integrated neutron flux per unit energy interval of neutron energy E falling on the counter, $N(E)$ is the number of proton recoils detected per unit energy interval at proton energy E , $\sigma(E)$ is the neutron-proton elastic scattering cross section at E , and k is a constant which depends on the properties of the spectrometer. The derivation of Eq. 1 depends upon the neutron-proton scattering being isotropic. Unfortunately, the curve of Fig. 14 has not as yet been converted to a neutron flux spectrum since the energy calibration of the spectrometer has not been completed. However, since the neutron spectrum varies essentially as the negative of the slope of the proton recoil spectrum, it is evident that the neutron spectrum derived from Fig. 14 will have, in common with that of Fig. 13, a broad peak below which the spectrum rises toward infinity. In the case of Fig. 14, the separation between the peak and the unlimited rise will be more marked than in Fig. 13. It should be noted that the broad peak in Fig. 14 comes in a pulse height range much below that where the unmoderated D-D peak would be found. The energy calibration of the "twin" scintillation spectrometer is now being carried out with monoenergetic neutron sources so that the conversion of Fig. 14 to a neutron spectrum may be accomplished.

E. Nuclear Emulsion Measurements

In addition to the two previously described measurements, a measurement was also made of the graphite-moderated D-T spectrum by means of a nuclear emulsion. A fresh 200-micron plate of Ilford C-2 emulsion wrapped in 2-mil platinum foil and black paper was exposed for approximately 22 hours in a position in line with the accelerator beam tube projected

and 2 m from the face of the 20-inch graphite cube. During this time the tritium-zirconium target at the cube center was bombarded with deuteron currents ranging from 5 to 15 microamperes. The nuclear plate was given an angle of inclination of approximately 5 degrees relative to a line passing through it and the source center.

The plate was developed by the two-bath method of Blau and DeFelice,⁽⁵⁾ and was impregnated with wood rosin to prevent shrinkage, as suggested by Dr. Roberts⁽⁶⁾ of Northwestern University. The proton track lengths were measured under 970-power magnification with a calibrated eyepiece scale, using azimuthal and dip acceptance angles of ± 10 degrees to insure that the proton energies obtained would give a good measure of the neutron energies causing the recoils. The lengths of tracks which began and ended in the emulsion and met the angular criteria were converted to energies using the range-energy of Beiser.⁽⁷⁾ The numbers of such tracks in each 1-Mev interval were determined, and each number was corrected for the fraction of the tracks which escape from the emulsion at the given energy by means of Rosen's⁽⁸⁾ curve, and for the variation of the neutron-proton elastic scattering cross section with energy.⁽⁹⁾

The resulting neutron spectrum is shown in Fig. 15. It should be emphasized that this is a preliminary spectrum representing only 198 events, and the statistical uncertainties of the points are quite large. Nevertheless, in common with the moderated D-D spectra of Figs. 13 and 14, the nuclear plate spectrum does give evidence of a broad peak at intermediate energies and a rise toward infinity at low energies. In addition, it indicates that a fair number of the D-T neutrons are getting through the graphite with little energy change.

F. Comparison of Theory and Experiment

In Part II of this report a neutron age theory derivation is given for the spectrum emerging from a heavy spherical moderator with a mono-energetic, isotropic, neutron point source at the center. While age theory is admittedly based on very simplified assumptions, it still should give some rough idea of what to expect from the present experimental arrangement at low energies after many scatterings. The approximation should be best for the case of the D-D neutrons where there can be no inelastic scatterings and the elastic scattering is approximately isotropic in the center-of-mass system.

Accordingly, Fig. 16 shows the spectrum calculated from Eq. 2, Part II, for the case of 2.6 Mev neutrons and a 50-cm-diameter sphere, the diameter of the sphere being chosen to coincide with the thickness of the cube used in the experimental arrangement. The age-theory spectrum shows the rise toward infinity at low energies found in all the experimental spectra but gives no evidence of the broad peak at intermediate energies or

Fig. 15. Spectrum measured with 200-micron nuclear emulsion for the neutrons emerging from a 20-inch graphite cube enclosing a D-T source.

Fig. 16. Spectrum calculated by age theory for the neutrons emerging from a 50-cm.-diameter graphite sphere enclosing a 2.6 Mev monoenergetic, isotropic, point source.

of the presence of any appreciable number of neutrons whose energies have been changed very little. In connection with this latter point it must be remembered that age theory is a very poor approximation for a small number of scatterings. The discrepancy between experiment and theory in regard to the existence of the broad peak at intermediate energies may be due to one or a combination of the following reasons: (1) deficiencies of the age theory in the present case, (2) differences between a cubical moderator and a spherical one, (3) rather broad energy spread of the neutrons introduced in the experimental case, and (4) anisotropic angular distribution of the neutrons introduced in the experimental case.

All the evidence thus seems to verify the existence of the unlimited rise in the graphite-moderated spectrum at low energies. A recent moment-method⁽¹⁰⁾ calculation also indicates such a rise for the case of 2-Mev neutrons moderating in air when the observation point is several mean free paths or more from the source.⁽¹¹⁾ According to age theory, the unlimited rise in the spectrum will occur with any heavy-nucleus spherical moderator of sufficient thickness to reduce the average neutron energy to 0.25 or less of its original value, as is seen in Part II of this report. For lesser moderator thicknesses the spectrum will consist of a very broad peak, the energy of maximum amplitude coming somewhat above the average energy. Indeed, if we consider the maximum amplitude value to be the effective energy of the moderated spectrum, age theory indicates that it is only possible to reduce 2.6-Mev neutrons to around 1 Mev and 14-Mev neutrons to around 5.6 Mev by this method without incurring the unlimited rise and then only at great sacrifice with regard to spectrum narrowness.

The suggestion has been made that the low-energy unlimited rise with a thick moderator might be eliminated by forming the moderator in the shape of a long narrow cylinder. Monoenergetic neutrons would be introduced at one end of the cylinder and moderated ones would be observed at the other end, all neutrons emerging from the side walls being eliminated by an absorber. However, a simple calculation shows that with typical parameters only about one properly moderated neutron would emerge from the cylinder face to 50,000 entering the absorber. Thus, the source could only yield low intensity, and an elaborate absorber would be required to eliminate background from the neutrons leaving the cylinder sides.

G. Charged-Particle Reactions-Type Source

In view of the difficulties encountered with the moderator-type source it was decided to survey all known charged particle-neutron reactions to see whether there exist enough suitable ones for a variable energy neutron source composed of a low-voltage accelerator and a set of targets. Such a source would have the advantages of small active volume and narrow, controllable neutron energy spread. However, the requirements are difficult in that with only 250-kev charged particles available most negative Q-value

reactions are eliminated, and a number of reactions with positive ground-state Q-values within fairly closely defined limits must be found to provide the low energy points. In addition, these reactions must provide sufficient intensity, and many reactions have low yields at low bombardment energies. The materials chosen must be capable of being made up in target form, and they must be generally available if we are to achieve our original objectives.

To date, the survey has been concentrated on finding reactions to provide the more difficult low energy points and has covered the Ajzenberg and Lauritsen(12) and Endt and Kluver(13) compilations plus the pertinent American and British journals published since. One salient fact which quickly became apparent is that only in the case of very light charged particles bombarding light-element targets has any appreciable amount of work been done on charged particle-neutron reactions. Even for most reactions in this area the ground state Q-values are not very well known, and the yields have not been measured at all. The reactions uncovered by the survey which appear to hold some promise are listed in Table 17 together with the neutron energy ranges obtainable by the use of the 250 kev Cockcroft-Walton linear accelerator. The latter energy ranges are calculated from the given measured Q-values and take account of the fact that the neutron energy may be varied somewhat by changing the bombardment energy or the angle of observation of the neutrons. The natural abundances of the target isotopes are also indicated in the table; some of them appear to be high enough to indicate use of the elements as they occur in nature. In addition, information is given on the enrichments available.

Table 17

Charged particle-neutron reactions chosen for further study

Reaction	Ground-state Q-value, Mev		Calculated neutron energy range obtainable, Mev	Availability of target isotope	
	Calculated from masses	Measured		Natural isotopic abundance, (19) %	Enriched material from Oak Ridge National Laboratory (19)
$\text{Ne}^{20}(\text{d}, \text{n})\text{Na}^{21}$	0.226	-0.17(14)	0 -0.07	90.92	Not yet available.
$\text{Mg}^{24}(\text{d}, \text{n})\text{Al}^{25}$	0.07	0.07(15)	0.11-0.32	78.60	Mg^{24}O or Mg^{24} available in gram (or smaller) amounts enriched to 98.4-99.6% at \$400-575 per gram.
$\text{B}^{11}(\text{d}, \text{n})\text{N}^{14}$	0.154	-	0.16-0.39	81.02-81.55	B^{11} available in gram (or smaller) amounts enriched to 93.5-99+% at \$7750-16,500 per gram.
$\text{S}^{32}(\text{d}, \text{n})\text{Cl}^{33}$	0.3	0.25(16)	0.28-0.50	95.06	S^{32} available in gram (or smaller) amounts enriched to 97.9-99.1% at \$1700-3400 per gram.
$\text{Si}^{28}(\text{d}, \text{n})\text{P}^{29}$	0.498	0.29(17) -0.80(18)	0.32-0.54	92.27	Si^{28}O_2 or Si^{28} available in gram (or smaller) amounts enriched to 98.1-99.4% at \$900-1500 per gram.
$\text{B}^{10}(\text{d}, \text{n})\text{N}^{13}$	1.068	-	0.98-1.32	18.98-18.45	B^{10} available in gram amounts enriched to 92-96% at \$15 per gram.

H. Conclusions

The prospects for development of a variable energy neutron source based on the moderator principle alone seem rather poor. The energy distributions obtained are too broad for the requirements, and in addition, it is not possible to reduce the input energy to any appreciable degree without introducing the unlimited rise at low energies.

The charged-particle reactions-type source seems to hold some promise, but to be sure, the reactions already found will have to be studied at the low energies available, and perhaps some hitherto unstudied reactions will also have to be investigated. It is planned to use a recently obtained $\text{Li}^6\text{I}(\text{Eu})$ crystal spectrometer in these studies, as well as the "twin" scintillation spectrometer. In case sufficient usable reactions are not available to cover all the desired energies, the combination of a set of targets and a set of thin moderators might prove to be the answer to the problem.

In conclusion, we wish to thank Dr. I. B. Berlman for many helpful ideas on the operation of the "twin" scintillation fast neutron detector, Dr. J. E. Rose for the design of the accelerator beam tube extension and target holder, and Dr. J. H. Roberts of Northwestern University for the techniques and materials which he supplied during the nuclear emulsion phase of the work.

VARIABLE ENERGY NEUTRON SOURCE

PART II. DIFFUSION OF MONOCHROMATIC NEUTRONS BY A SPHERICAL MODERATOR

J. C. Reynolds*

The purpose of this study was to investigate theoretically the possibility of using a moderator to produce a roughly monochromatic distribution of neutrons from a source of monochromatic neutrons of a higher energy. The energy distribution of the neutron radiation from a sphere of moderator with a monochromatic source at its center was calculated by means of neutron age theory. It was found that the distribution per unit energy interval will go to infinity as the energy goes to zero if the ratio of incident energy to average energy of the distribution is greater than about four to one.

Age theory is an approximation which holds only for moderators with heavy nuclei and without appreciable inelastic scattering cross sections.⁽²⁰⁾ If either light nuclei or inelastic scattering are present, the distribution will be broader than that predicted by age theory.

The "age" θ of a neutron is a measure of its energy loss by scattering. It is given by:

$$\theta = \int_0^u du' \frac{\ell^2(u')}{3\xi(1 - \cos \phi)}$$

where $u = \ln \frac{E_0}{E} = \ln \frac{\text{initial energy}}{\text{present energy}}$

l = mean distance between scatterings,

$$\overline{\cos \phi} = \frac{2}{3M}$$

and $\xi = 1 + \frac{(M-1)^2}{2M} \ln \left(\frac{M-1}{M+1} \right)$

$$\cong \frac{2}{M + 2/3} \quad (\text{to within 1\% for } M > 10)^{(21)}$$

where M is the mass of the moderator nuclei. We limit ourselves to the case where ℓ is independent of energy. Then

$$\theta = \frac{M}{6} \frac{M+2/3}{M-2/3} \ell^2 \ln \frac{E_0}{E} \tag{1}$$

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In age theory, the assumption is made that there is a unique relationship between the time since a neutron leaves the source and its energy. If this is assumed, then it is possible to define

$$\psi(\vec{r}, \theta) = - \frac{dE}{dt} N(\vec{r}, E)$$

where $N dE d\vec{r}$ is the number of neutrons in a volume $d\vec{r}$ about $\vec{r}$ with energies between E and $E + dE$. It may be shown that ψ must satisfy

$$\frac{\partial \psi}{\partial \theta} = \nabla^2 \psi + s \delta(\theta)$$

where s represents the neutron source and θ is the age defined above. (22) If we normalize to a unit production of neutrons per second, this becomes

$$\frac{\partial \psi}{\partial \theta} = \nabla^2 \psi + \frac{\delta(r) \delta(\theta)}{4\pi r^2}$$

With the boundary condition $\psi = 0$ on the surface of the sphere, this has a solution of the form (where a = radius of the sphere) (23)

$$\psi(\theta, r) = \frac{1}{2a^2 r} \sum_{m=1}^{\infty} m \sin \frac{m\pi r}{a} e^{-\frac{m^2 \pi^2}{a^2} \theta}$$

Now the rate at which the neutrons radiate from the sphere of moderator is given by:

$$\begin{aligned} R &= \left(- \frac{dt}{dE} \right) \left(- \frac{d}{dt} \int \psi(\vec{r}, \theta) d\vec{r} \right) \\ &= \frac{d\theta}{dE} \frac{d}{d\theta} \int \psi(\vec{r}, \theta) d\vec{r} \\ &= - \frac{M}{6} \frac{M+2/3}{M-2/3} \frac{\ell^2}{E} \frac{\partial}{\partial \theta} \int \psi(\vec{r}, \theta) d\vec{r} \end{aligned}$$

where the integrations are performed over the volume of the sphere. Substituting the solution for ψ , we get

$$\begin{aligned} R &= \frac{M}{6} \frac{M+2/3}{M-2/3} \frac{\ell^2}{E} 2 \sum_{m=1}^{\infty} (-1)^{m+1} \frac{m^2 \pi^2}{a^2} e^{-\frac{m^2 \pi^2}{a^2} \theta} \\ &= \frac{h}{E} F \left(h \ln \frac{E_0}{E} \right) \end{aligned} \quad (2)$$

where

$$F(X) = 2 \sum_{m=1}^{\infty} (-1)^{m+1} m^2 e^{-m^2 X} \quad (X > 0) \quad (3)$$

and

$$h = \frac{\pi^2}{a^2} \frac{M}{6} \frac{M+2/3}{M-2/3} \varrho^2 \quad (4)$$

The entire dependence of R on the moderator properties and the size of the sphere is contained in the single parameter h . For the purpose of calculation, we set $E_0 = 2.6$ Mev, which corresponds to the average energy of neutrons produced by a deuteron-deuteron reaction. Once E_0 is fixed, the average energy of the radiated neutrons, $E_{av} = \int E R dE$, is a function of h only. This relationship was solved by graphical means, giving the following results:

E_{av}	E_{av}/E_0	h
0.065 Mev	0.0250	0.311
0.150	0.0577	0.430
0.3	0.115	0.599
0.6	0.231	0.954
1.5	0.577	2.86

R was then calculated for each value of h . The results are given in Table 18 and Fig. 17. Note that although Fig. 17 uses logarithmic axes, R is a distribution per unit energy, not per unit logarithm of energy.

It may be seen that R goes to infinity as E approaches zero whenever $h < 1$. This critical point corresponds to $E_{av}/E_0 = 0.2466$.

Table 18

Spectra of 2.6 Mev neutrons scattered by a spherically symmetric moderator

$h = 0.311$		$h = 0.430$		$h = 0.599$		$h = 0.954$		$h = 2.86$	
$E(\text{kev})$	R	$E(\text{kev})$	R	$E(\text{kev})$	R	$E(\text{kev})$	R	$E(\text{kev})$	R
1,162	3.69×10^{-6}	1,453	4.08×10^{-6}	1,713	4.83×10^{-6}	1,999	6.60×10^{-6}	2,382	1.66×10^{-5}
520	1.91×10^{-4}	811	1.69×10^{-4}	1,128	1.70×10^{-4}	1,540	1.98×10^{-4}	2,181	4.19×10^{-4}
104.4	1.76×10^{-3}	254	1.00×10^{-3}	489	7.25×10^{-4}	740	7.63×10^{-4}	1,833	9.23×10^{-4}
4.19	1.99×10^{-2}	24.9	4.63×10^{-3}	92.0	1.74×10^{-3}	320	7.99×10^{-4}	1,292	5.93×10^{-4}
0.167	0.186	2.43	1.76×10^{-2}	17.3	3.45×10^{-3}	112	8.48×10^{-4}	910	3.13×10^{-4}
0.00676	1.68	0.238	6.61×10^{-2}	3.28	6.68×10^{-3}	39.3	8.88×10^{-4}	642	1.63×10^{-4}
0.000268	15.56	0.0231	0.249	0.615	1.31×10^{-2}	13.8	9.28×10^{-4}	452	8.47×10^{-5}

(Fig. 17 consists of these data plotted on a 5x6 cycle log-log sheet. E runs from 0.1 to 10,000 kev, and R from 10^{-6} to 1)

Fig. 17. Spectra of 2.6 Mev neutrons scattered by a spherically symmetric moderator.

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CALCULATION OF RETENTION AND ELIMINATION OF Ra^{228} AND Ra^{224} FROM A SINGLE INJECTION OF Th^{232}

J. C. Reynolds*

A. Introduction

If a radioactive substance is injected into the body, the amounts of this substance and its successive decay products which will be present in the body and in the excreta will depend upon both the radioactive decay rates and the metabolic processes of elimination. General expressions may be obtained for the retention and elimination functions of such a radioactive decay series in terms of the decay constants and empirical functions describing the metabolic elimination.

In this report such expressions are derived for the case of injection of Th^{232} in a single dose, and they are applied to the first four decay products of the chain. The assumption is implicitly made that all daughter products enter the general circulation. In an earlier paper⁽¹⁾ the retention and elimination of the first decay product (radium) were calculated for five different isotopes of thorium. In most of these cases, the remaining decay products would be in simple radioactive equilibrium with the radium, but for Th^{232} the situation is more complicated. The third decay product, Th^{228} , comes into equilibrium only after about 15 years, and the activity of the fourth product, Ra^{224} , remains below that of the Th^{228} by a constant ratio, due to its own metabolic elimination.

B. The General Case

We first consider the general problem of determining the amounts of various members of a radioactive decay series which will be present in vivo and in excreta due to the injection of the initial member of the series. It is assumed that the nature of the metabolic elimination of each member of the series is known and can be expressed by a "chemical retention" function $R_C(t)$, which gives the proportion of an initial amount of the substance which will remain in the body after chemical elimination processes have acted on it for a time t . It must be emphasized that $R_C(t)$ describes only the chemical elimination processes; radioactive decay of the substance is considered separately, so that the actual amount of a radioactive substance remaining at a time t after its production or injection will be $R_C(t)e^{-\lambda t}$, where λ is the proper radioactive decay constant. In practice, $R_C(t)$ may vary depending on the site of the production of the substance, and it must be chosen to correspond to the actual site of the decay of the parent.

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The retention function $R_n(t)$ is defined as the amount (No. of atoms) of n th member of the radioactive decay series which will be present in vivo at a time t due to a unit dose (single atom) of the initial member at time 0. The retention function for the initial member itself is obviously

$$R_0(t) = R_{C_0}(t) e^{-\lambda_0 t} \quad (1)$$

The succeeding retention functions may be found if we can express the retention function $R_n(t)$ for each member in terms of the retention function $R_{n-1}(t)$ for its parent. Between the times τ and $\tau + d\tau$ the amount of a member produced by decay of its parent will be $\lambda_{n-1} R_{n-1}(\tau) d\tau$. At a later time t this amount will be reduced to

$$\lambda_{n-1} R_{n-1}(\tau) d\tau e^{-\lambda_n(t-\tau)} R_{Cn}(t-\tau)$$

Thus the total amount of the substance present at time t will be

$$R_n(t) = \lambda_{n-1} \int_0^t R_{n-1}(\tau) e^{-\lambda_n(t-\tau)} R_{Cn}(t-\tau) d\tau \quad (2)$$

or, by putting $u = t - \tau$

$$R_n(t) = \lambda_{n-1} \int_0^t R_{n-1}(t-u) e^{-\lambda_n u} R_{Cn}(u) du \quad (3)$$

which is the desired relationship between $R_n(t)$ and $R_{n-1}(t)$.

Special attention should be given to the case of secular equilibrium, where $\lambda_n \gg \frac{R_{n-1}'(t)}{R_{n-1}(t)}$, i.e., where the daughter is short lived in comparison with the time it takes for the amount of the parent to change appreciably. Here we can make the simplifying approximation $R_{n-1}(t-u) \approx R_{n-1}(t)$, giving

$$R_n(t) \approx \lambda_{n-1} R_{n-1}(t) \int_0^t R_{Cn}(u) e^{-\lambda_n u} du \quad (4)$$

If, in addition, $t \gg \frac{1}{\lambda_n}$, then

$$R_n(t) \approx \lambda_{n-1} R_{n-1}(t) \int_0^\infty R_{Cn}(u) e^{-\lambda_n u} du \quad (5)$$

In the particular case where $R_{Cn}(u) = 1$, this reduces to

$$R_n(t) \approx \frac{\lambda_{n-1}}{\lambda_n} R_{n-1}(t) \quad (6)$$

We next define the elimination functions $B_n(t)$, which give the rate at which each member of the series is eliminated chemically, i.e., the amount of the member present in the total excreta produced in a unit time. The time derivative of $R_n(t)$, as expressed by (2) is

$$\begin{aligned} R'_n(t) &= \lambda_{n-1} \frac{d}{dt} \int_0^t R_{n-1}(\tau) e^{-\lambda_n(t-\tau)} R_{Cn}(t-\tau) d\tau \\ &= \lambda_{n-1} R_{n-1}(t) - \lambda_n R_n(t) + \lambda_{n-1} \int_0^t R_{n-1}(\tau) e^{-\lambda_n(t-\tau)} R'_{Cn}(t-\tau) d\tau \end{aligned} \quad (7)$$

The first term in the last expression is the rate of production by radioactive decay of the parent, while the second term is the loss due to the substance's own radioactive decay. Thus the third term must be the rate of chemical elimination. Since it is a rate of decrease, we must reverse the sign to make $B_n(t)$ positive.

$$\begin{aligned} B_n(t) &= -\lambda_{n-1} \int_0^t R_{n-1}(\tau) e^{-\lambda_n(t-\tau)} R'_{Cn}(t-\tau) d\tau \\ &= -\lambda_{n-1} \int_0^t R_{n-1}(t-u) e^{-\lambda_n u} R'_{Cn}(u) du \end{aligned} \quad (8)$$

Again, for secular equilibrium we may use $R_{n-1}(t-u) \approx R_{n-1}(t)$

giving

$$B_n(t) \approx -\lambda_{n-1} R_{n-1}(t) \int_0^t e^{-\lambda_n u} R'_{Cn}(u) du \quad (9)$$

and if $t \gg \frac{1}{\lambda_n}$

$$B_n(t) \approx -\lambda_{n-1} R_{n-1}(t) \int_0^\infty e^{-\lambda_n u} R'_{Cn}(u) du \quad (10)$$

To eliminate the derivative of $R_{Cn}(u)$ from (8) and (10) we use integration by parts.

From (8) we get

$$\begin{aligned} B_n(t) &= \lambda_{n-1} R_{n-1}(t) - \lambda_{n-1} R_{n-1}(0) e^{-\lambda_n t} R_{Cn}(t) \\ &\quad - \lambda_n R_n(t) - \lambda_{n-1} \int_0^t R'_{n-1}(\tau) e^{-\lambda_n(t-\tau)} R_{Cn}(t-\tau) d\tau \end{aligned} \quad (11)$$

while from (10) we get the expression for secular equilibrium and $t \gg \frac{1}{\lambda_n}$,

$$\begin{aligned} B_n(t) &\approx \lambda_{n-1} R_{n-1}(t) - \lambda_{n-1} R_{n-1}(t) \int_0^\infty \lambda_n e^{-\lambda_n u} R_{Cn}(u) du \\ &= \lambda_{n-1} R_{n-1}(t) - \lambda_n R_n(t) \end{aligned} \quad (12)$$

It should be noted that both $R(t)$ and $B(t)$ are in terms of actual amounts (atoms) of material.

However, since from an experimental viewpoint the associated activities are of direct interest, we define two other quantities: the activity ratio $A_n(t)$, which is the ratio between the activity in vivo of the n th member of the series and the activity in vivo of the initial member at the same time; and the elimination ratio $E_n(t)$, which is the ratio between the activity of the n th member in the total excreta produced in a unit time and the activity of the initial member in vivo at the same time. In terms of $R(t)$ and $B(t)$

$$\begin{aligned} A_n(t) &= \frac{\lambda_n R_n(t)}{\lambda_0 R_0(t)} \\ E_n(t) &= \frac{\lambda_n B_n(t)}{\lambda_0 R_0(t)} \end{aligned} \quad (13)$$

C. Applications to the Decay of Th^{232}

We now apply the results of the preceding section to the specific radioactive series

The decay constants are:

$$\lambda_0 = 1.97 \times 10^{-13} \text{ days}^{-1}$$

$$\lambda_1 = 0.0002834 \text{ days}^{-1}$$

$$\lambda_2 = 2.714 \text{ days}^{-1}$$

$$\lambda_3 = 0.001000 \text{ days}^{-1}$$

$$\lambda_4 = 0.1905 \text{ days}^{-1}$$

We make the following assumptions:

- (1) Ac^{228} is in secular equilibrium with Ra^{228} .
- (2) Ra^{224} is in secular equilibrium with Th^{228} .
- (3) Thorium and actinium are not chemically eliminated to an appreciable extent.

Because of its long half-life and the fact that it is not eliminated, the Th^{232} in the body remains constant in amount, i.e. $R_0(t) = 1$. Using Eqs. (3), (6), (2), (5), (11), (12), and (13) we obtain the following expressions for the activity and elimination ratios:

$$A_1 = \lambda_1 \int_0^t e^{-\lambda_1 u} R_C(u) du \quad (\text{Ra}^{228})$$

$$A_2 = A_1 \quad (\text{Ac}^{228})$$

$$A_3 = A_1 - \lambda_1 e^{-\lambda_3 t} \int_0^t e^{(\lambda_3 - \lambda_1)u} R_C(u) du \quad (\text{Th}^{228})$$

$$A_4 = \lambda_4 A_3 \int_0^\infty e^{-\lambda_4 u} R_{C'}(u) du \quad (\text{Ra}^{224})$$

$$E_1 = \lambda_1 [1 - e^{-\lambda_1 t} R_C(t) - A_1] \quad (\text{Ra}^{228})$$

$$E_4 = \lambda_4 [A_3 - A_4] \quad (\text{Ra}^{224}) \quad (14)$$

where $R_C(t)$ and $R_{C'}(t)$ are the chemical retention functions for Ra^{228} and Ra^{224} respectively.

It is found experimentally that the chemical retention of radium may be described by the empirical formula⁽¹⁾

$$R_C(t) = (1 - b) t^{-b} \quad \text{for } t \geq 1 \text{ day.} \quad (15)$$

The simplest method of estimating $R_C(t)$ for $0 \leq t < 1$ is to fit a parabola to the conditions:

$$(1) \quad R_C(0) = 1$$

$$(2) \quad R_C(t) \text{ and } \frac{d}{dt} R_C(t) \text{ are continuous at } t = 1.$$

This gives the expression⁽¹⁾

$$R_C(t) = 1 - (b + b^2)t + b^2 t^2 \text{ for } 0 \leq t \leq 1 \quad (16)$$

It is important to recognize the possibility of different values of b for Ra^{228} and Ra^{224} ; we use b for Ra^{228} and b' for Ra^{224} . This distinction is made necessary primarily by the fact that $R_{C_1}(u)$ appears in the integral

$$\int_0^{\infty} e^{-0.1905 u} R_{C_1}(u) du$$

which is highly sensitive to the behavior of $R_{C_1}(u)$ for u on the order of a few days. In this short interval there is not sufficient experimental support for (15), and the effective value of b may be somewhat different than that obtained for larger t .

D. Calculations

Using Eqs. (14), (15), and (16), we have calculated the values of the activity ratios and elimination ratios for the Th^{232} decay series. The computations were carried out for $b = 0.5$ and 0.7 , and $b' = 0.4, 0.5, 0.6, 0.7$, and 0.8 .

The integrals of the form $\int_0^t e^{-\lambda t} t^{-b} dt$ were integrated numerically

by expanding the exponential into a power series. When the value of the exponent $-\lambda t$ was negative,* the results were checked against a table of the incomplete gamma function.

It should be noticed that the assumptions of secular equilibrium (1 and 2 in Section C) introduce an error in the calculations for small t since they depend upon the condition $t \gg \frac{1}{\lambda_n}$. The error due to these assumptions is about 5% at four days for the Ac^{228} and Th^{228} functions and about 5% at 100 days for the Ra^{224} functions. In both cases the error is inversely proportional to t . The 5% error point is indicated by a broken vertical line in the graphs.

Tables (19 and 20) and graphs (Figs. 18-21) of $A(t)$ and $E(t)$ follow. They are grouped according to the value of b .

*As in the case for A_1

Table 19

 $b = 0.5$

t (in days)	$A_1(t) (\text{Ra}^{228}) \times 10^2$	$A_3(t) (\text{Th}^{228}) \times 10^2$	$E_1(t) (\text{Ra}^{228}) \times 10^4$
1	0.020	0.000	1.417
2	0.032	0.000	1.832
5	0.055	0.000	2.200
10	0.081	0.001	2.385
20	0.118	0.002	2.516
50	0.191	0.006	2.631
100	0.272	0.017	2.689
200	0.385	0.047	2.728
500	0.597	0.166	2.762
1000	0.810	0.384	2.777
2000	1.055	0.752	2.786
5000	1.346	1.263	2.791
10,000	1.458	1.449	2.792
30,000	1.484	1.484	2.792

$$A_2(t) (\text{Ac}^{228}) = A_1(t)$$

$$A_4(t) (\text{Ra}^{224}) = A_3(t) \times \begin{array}{l} 0.4073 \text{ for } b' = 0.4 \\ 0.3320 \text{ for } b' = 0.5 \\ 0.2600 \text{ for } b' = 0.6 \\ 0.1991 \text{ for } b' = 0.7 \\ 0.1467 \text{ for } b' = 0.8 \end{array}$$

$$E_4(t) (\text{Ra}^{224}) = A_3(t) \times \begin{array}{l} 0.1129 \text{ for } b' = 0.4 \\ 0.1273 \text{ for } b' = 0.5 \\ 0.1410 \text{ for } b' = 0.6 \\ 0.1526 \text{ for } b' = 0.7 \\ 0.1625 \text{ for } b' = 0.8 \end{array}$$

Table 20

 $b = 0.7$

t (in days)	$A_1(t) (\text{Ra}^{228}) \times 10^3$	$A_3(t) (\text{Th}^{228}) \times 10^3$	$E_1(t) (\text{Ra}^{228}) \times 10^4$
1	0.161	0.000	1.984
2	0.226	0.000	2.310
5	0.337	0.001	2.558
10	0.443	0.003	2.664
20	0.573	0.008	2.729
50	0.791	0.028	2.778
100	0.998	0.071	2.798
200	1.249	0.173	2.811
500	1.649	0.512	2.820
1000	1.993	1.040	2.823
2000	2.338	1.782	2.825
5000	2.688	2.582	2.826
10,000	2.803	2.790	2.826
30,000	2.826	2.826	2.826

$$A_2(t) (\text{Ac}^{228}) = A_1(t)$$

$$A_4(t) (\text{Ra}^{224}) = A_3(t) \times \begin{array}{l} 0.4073 \text{ for } b' = 0.4 \\ 0.3320 \text{ for } b' = 0.5 \\ 0.2600 \text{ for } b' = 0.6 \\ 0.1991 \text{ for } b' = 0.7 \\ 0.1467 \text{ for } b' = 0.8 \end{array}$$

$$E_4(t) (\text{Ra}^{224}) = A_3(t) \times \begin{array}{l} 0.1129 \text{ for } b' = 0.4 \\ 0.1273 \text{ for } b' = 0.5 \\ 0.1410 \text{ for } b' = 0.6 \\ 0.1526 \text{ for } b' = 0.7 \\ 0.1626 \text{ for } b' = 0.8 \end{array}$$

Fig. 19

Fig. 21

Fig. 18

Fig. 20

Reference

1. Reynolds, J., P. F. Gustafson, and L. D. Marinelli, Retention and Elimination of Radium Produced by the Decay of Thorium Within the Body. Argonne National Laboratory Report, ANL-5689, 1957.

ANALYSIS OF GAMMA-RAY SPECTRA BY THE METHOD OF LEAST SQUARES

J. C. Reynolds*

The method of least squares may be applied to the spectra of mixtures of radioactive materials in order to determine the relative abundance of different isotopes in the mixture. The method is particularly well suited for this analysis when the error in the spectra is largely a random statistical deviation.

Suppose that a material containing a mixture of n radioactive substances produces a spectrum $f(x)$, where x is the channel number (proportional to energy) and f is the number of counts in the channel. If a unit amount of the i th radioactive substance has a spectrum $g_i(x)$ and is present in an amount a_i in the mixture, then

$$f(x) = \sum_j a_j g_j(x)$$

The $g_i(x)$ are called the reference spectra, $f(x)$ is called the unknown spectrum, and the a_i are called the spectral coefficients.

We now consider the problem of determining the a_i if $f(x)$ and the $g_i(x)$ are known. Using the method of least squares, we choose the values of a_i which minimize the expression

$$\epsilon = \sum_x [f(x) - \sum_j a_j g_j(x)]^2$$

If the values of a_i minimize ϵ , then $\frac{\partial \epsilon}{\partial a_i} = 0$ or

$$\sum_x [f(x) - \sum_j a_j g_j(x)] g_i(x) = 0$$

If we define

$$b_i = \sum_x f(x) g_i(x)$$

$$m_{ij} = \sum_x g_i(x) g_j(x)$$

then

$$\sum_j m_{ij} a_j = b_i$$

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The m_{ij} form a matrix whose inverse has the elements m_{ij}^{-1} , therefore

$$a_i = \sum_j m_{ij}^{-1} b_j$$

Thus the a_i are computed from the spectra by forming the vector elements b_i and the matrix elements m_{ij} , inverting the matrix, and multiplying the b_i by the m_{ij}^{-1} . A measure of the error in the computation of the a_i 's is given by the error spectrum

$$e(x) = f(x) - \sum_j a_j g_j(x)$$

A more compact measure of the error is the sum of the squares of the error spectrum

$$\epsilon = \sum_x e^2(x) = \sum_x [f(x) - \sum_j a_j g_j(x)]^2$$

A set of programs for an IBM 650 computer have been written for the purpose of analyzing gamma-ray spectra by the above method. They were specifically designed to process data from 256-channel pulse height analyzers and to handle up to eight reference spectra.

One restriction in the use of the method of least squares should be mentioned. It minimizes the errors in the a_i due to statistical fluctuations in the counting rates. It does not minimize errors due to other factors, such as channel shifting, dead time losses, or variations in the counting geometry. It is particularly sensitive to channel shifts in cases where different reference spectra have photopeaks close to one another.

THE COMPARATIVE DEPOSITION OF YTTRIUM, CERIUM AND THULIUM IN BONE TISSUE OF DOGS*

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

Abstract

A controversy exists concerning the site of deposition of the rare-earth elements in bone tissue, some authors maintaining that it is retained by bone protein, others that it is not. The present report describes experiments which were carried out to show the histological localization of three rare-earth elements, Y^{91} , Ce^{144} and Tm^{170} in the bones of dogs after intraperitoneal injection. Further work, in which bone was analysed for deposited radioelements in vivo and measurements made of the activity taken up by bone samples from a medium in vitro were carried out to confirm and explain the findings of the histological observations. The results show that these rare-earth elements are deposited in vivo on highly mineralised nongrowing surfaces of bone that are either resorbing or are inactive. In vitro, the activity is retained by bone mineral and not by bone protein samples. The results are consistent with the conclusion that the rare-earth elements are adsorbed onto bone mineral surfaces and are not taken up by the matrix of bone.

In 1947 Hamilton and his colleagues distinguished between the pattern of deposition in bone shown by the alkaline-earth and rare-earth elements. There is more than adequate evidence to show that the alkaline-earth or calcium-like elements are taken up in areas of bone formation in the process of calcification. However there exist contradictory explanations of the deposition of the rare earths in bone. Hamilton⁽¹⁾ and Durbin⁽²⁾ in their studies on the metabolism of the lanthanons describe autoradiographs of bone sections showing concentrations of the isotopes beneath the periosteum, endosteum and epiphyseal plate. They interpret them to demonstrate an affinity between these elements and the uncalcified osteoid matrix or bone protein. Copp⁽³⁾ compared the deposition of strontium and yttrium in the bone of normal and rachitic rats and found that yttrium retention, unlike strontium, is unaffected by age or rickets, and came to the same conclusion as Hamilton. Their evidence, however, was not conclusive because only two fractions of bone were considered as possible sites of deposition of the elements, the mineral and the organic, and the argument was rather that the rare earths did not behave like calcium and therefore were not taken up by the inorganic fraction during mineralization, but must be in the organic fraction.

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Work carried out at Oxford⁽⁴⁾ was designed to illustrate the deposition of yttrium, the product of Sr^{90} decay, in the skeleton, and the autoradiographic results showed yttrium to be deposited in areas of bone resorption and not in the osteoid matrix.

The present report will show how this work was repeated and the observations extended to two other rare earths, cerium and thulium, and it will describe the attempts to elucidate the locus and mechanism of the deposition. In other words, where are the rare earths deposited in bone and why?

Since both yttrium and a number of other rare earths are fission products which are concentrated in the skeleton, their exact site of deposition is of interest from the radiation hazard point of view. Moreover, the mechanism of their uptake in bone might possibly elucidate some aspect of metabolism even though they are not elements that are naturally found in the skeleton.

All three of the elements used may be classified as rare earths. Yttrium behaves chemically and metabolically in a way similar to cerium and thulium and may be classified with these two elements in the lanthanon rare-earth series. Cerium and thulium lie respectively at the beginning and near the end of this series. Metabolic studies on whole-body distribution⁽⁵⁾ have shown that there is increased skeletal deposition of the heavier members of the lanthanon series. For this reason the bone deposition of cerium and thulium have been carefully compared to find whether different characteristics in their localization in bone could explain these differences in body distribution. The experiments may be divided into three parts: 1) the first part is concerned with the in vivo histological localization of the three elements in bone tissue using the methods of autoradiography and microradiography; 2) the second part consists in separating bone into two components, the organic and inorganic, and in discovering to which fraction the elements are fixed in vivo; 3) the third part consists of in vitro experiments in which whole bone, bone mineral, organic matrix and other materials were incubated in media containing one of the three elements.

Methods

Six six-month old puppies were kept on a low calcium diet for one week before injection. The diet was not deficient enough in calcium to produce histological rickets but did ensure the presence of wide bands of uncalcified osteoid in areas of bone growth. It was important to be quite sure if there was or was not deposition of any of the three elements in the organic part of bone. The radioactive isotopes Y^{91} , Ce^{144} and Tm^{170} were used. The Y^{91} and Ce^{144} were obtained as carrier-free solutions from Oak Ridge, and the Tm^{170} was produced by neutron bombardment of thulium metal and

had a low specific activity (0.82 mc/mg). The isotopes were injected intraperitoneally as the chlorides (500 μ c/kg body weight) in approximately 2 N HCl neutralized with an equivalent amount of NaOH immediately before injection.

The six puppies were divided into three litter-mate pairs, each pair being injected with a different isotope. One animal from each pair injected with Ce^{144} and Tm^{170} was killed 24 hours after, the other animal 7 days after injection; both animals receiving Y^{91} were killed 24 hours after injection.

Histological Localization

The femurs and tibias were removed and fixed in absolute alcohol. The distal ends of the femurs were cleaned of connective tissue, the marrow washed out, embedded in methyl methacrylate monomer and cut into longitudinal sections for gross autoradiography and microradiography. The proximal ends of the tibias were similarly cleaned and cross sections cut and prepared for autoradiography and microradiography by methods which have already been described.⁽⁶⁾

In Vivo Deposition

The rest of the long bones were sawn into 'diaphyseal' and 'epiphyseal' bone, the 'epiphyseal' bone including the metaphysis. The pieces were cleaned of muscle and marrow; the diaphyseal and epiphyseal pieces from the different bones were pooled, extracted with acetone for 24 hours, dried and the dry, fat-free weight determined. The samples were treated separately with ethylenediamine (ED) in a Soxhlet by the technique of Williams and Irvine.⁽⁷⁾ Each sample was extracted for six hours, washed three times with water and the ED extraction and washings collected. The sample was then washed overnight in running water, dried and weighed to determine the weight lost as the result of the first ED extraction. The sample was re-extracted in ED for three days, the procedure repeated as for the first extraction and the weight lost as result of the second extraction determined. The sample was then ashed at 600°C and reweighed to make sure that the ED extractions had removed all the organic material; the difference in weight before and after ashing should be not more than can be accounted for by water and CO_2 loss. The ashed sample was dissolved in dilute nitric acid and made up to a known volume. The ED extractions were evaporated to dryness, the residue taken up in nitric acid, and the solution made up to a known amount. Aliquots of the solutions were transferred to planchets and counted with a thin end-window counter. The total activity of the samples was calculated, using a known volume of the injection solution as the standard, and the results are expressed as the per cent of the activity in the whole bone.

The rest of the carcasses from the Ce^{144} and Tm^{170} -treated puppies were kept, and the tissues of the body analyzed separately for retained activity.

In Vitro Uptake

Cortical beef bone was pulverised to a 'saw dust' with a thick circular saw cooled with water. The fat was removed with acetone and the bone powder dried and passed through a coarse copper mesh. Some of this powder was used without further treatment as a 'whole bone' sample. Some was extracted in ED for five days, washed in running water and dried to produce bone mineral. More bone mineral was made by ashing whole bone powder at 600°C overnight. Bone powder was decalcified with disodium dihydrogen ethylened nitrilotetraacetic acid (EDTA) and washed well with water to produce organic bone matrix. Other substances investigated were tricalcium phosphate, cane sugar charcoal and Pyrex wool. Two hundred and fifty-milligram samples were weighed out in small paper thimbles (10 x 50 mm) and carefully and thoroughly hydrated with water. The bone organic samples were used immediately after being washed, without being dehydrated. Reconstituted human plasma (Lyovac) was used as the incubating medium *. Part of the plasma was reconstituted with 0.1 per cent citric acid and part with distilled water, and Y^{91} , Ce^{139} or Tm^{170} was added. Each thimble containing the samples was suspended by a wire and 5 cc of one of the incubating media was added, five minutes being allowed for it to drain through. Then 5 cc of the same medium without any isotope was added as a wash and allowed to drain for five minutes and 5 cc added again and the sample washed the second time. This was repeated for each of the seven samples for the citrated and noncitrated media for each of the three isotopes. The samples in the thimbles were placed in planchets, ashed at 600°C overnight and counted with a thin end-window counter. The results are strictly comparable and are expressed in counts per minute per sample. The whole experiment was repeated to get an idea of the variations and significance of the results.

Results

Histological Localization

The autoradiographs and microradiographs show that the deposition in bone of the three isotopes Y^{91} , Ce^{139} and Tm^{170} are very similar. The following description, therefore, applies to all three, and any differences between them will be mentioned in the text.

One day after injection the gross longitudinal autoradiographs show a heavy deposition beneath the epiphyseal plate, beneath the periosteum in the metaphysis and beneath the endosteum in the diaphysis. There is also a less intense uptake above the plate in the epiphysis, beneath the epiphyseal cartilage and a spotted, uneven distribution throughout trabecular and cortical bone. When these autoradiographs are compared with their corresponding

* The rare earths are known to form colloids and nonionized complexes in plasma (5,8) and the removal of activity from such complexes must be an active retention similar to that occurring in vivo.

X-rays and with the autoradiographs and microradiographs of the cross sections, then the exact sites of deposition of the isotopes can be discovered. In the region of the plate the activity appears on the surfaces of the eroded cartilage, though not in the cartilage in the calcifying zone, and on the surfaces of trabeculae where they are resorbed as they decrease in number to form the secondary spongiosa or are being removed to form the marrow cavity. Beneath the periosteum the activity appears on the highly calcified surfaces of bone that is either being resorbed or is inactive. In the diaphysis, where bone is being laid down periosteally in the thickening of the shaft, there is no deposition of activity where there is osteoid tissue produced in bone formation. Endosteally, the marrow cavity is enlarged in the diaphysis and where resorption has produced highly calcified bone surfaces, the autoradiographs show deposition of the isotopes.

In both the metaphysis and diaphysis there are many primary and some secondary haversian systems in which growth has finished, and the calcification has progressed to the edge of the osteoid border. The surface of these small canals shows uptake, less intense than the depositions on epiphyseal and periosteal resorbing surfaces, but, since there are large numbers of them, they account for a high proportion of the total activity of a bone and are responsible for the spotty appearance as seen in the gross autoradiographs. In cortical bone remodelling is taking place, and there are resorption cavities and building sites; the deposition of activity in a building site demonstrates most clearly the characteristics of the localization of the rare-earth isotopes. On one side of a building site, where bone formation is taking place, the incompletely calcified bone has a border of osteoid tissue on its surface, and there no deposition of activity is found; on the other side, where resorption is occurring, the bone surface is highly calcified and irregular, and there heavy deposition is discovered (Fig. 22).

All the localized depositions described so far are on bone surfaces which are mineralized either because resorption has left a highly calcified face or because calcification has been completed, and the mineral is no longer covered by a layer of osteoid tissue. The depositions are irregular and vary in intensity over a single surface without there being any apparent variation in the surface as seen in the microradiograph or the bone section. However, very little is known about the mechanism of resorption, and there may be some explanation of the irregularities when more is understood of how resorption occurs. In no instance are any of the three isotopes deposited in osteoid tissue.

Throughout the bone tissue away from areas of primary concentration there appears a deposition of low intensity which may be called a diffuse reaction.* This diffuse reaction does not vary appreciably except behind

* No quantitative measurements of this diffuse deposition have been made, however, it would seem to be less intense than the corresponding deposition of the calcium-like elements when compared with the concentrated depositions.

AUTORADIOGRAPH

MICRORADIOGRAPH

HISTOLOGICAL SECTION

.5mm

Fig. 22. Cross sections from the tibial diaphysis of a dog injected with Ce^{144} and killed 24 hours later. On the left is a section showing strong activity in a resorption cavity which the microradiograph shows to have a highly calcified surface, while areas of growth show no activity, are incompletely mineralised and have deeply staining borders when stained with Azure II as shown in the histological section. On the right is another section showing a resorption cavity and haversian system respectively with and without concentrations of activity, and also showing small completed haversian systems with uptake on the surface of the canal walls.

osteoid borders where calcification is occurring. Here there is a narrow band of deposition of an intensity two to five times that of the diffuse depositions but still many times less intense than the localized depositions. In this respect the three isotopes differ; Tm^{170} produces a higher intensity in the diffused depositions in proportion to the intensity in the concentrated localizations than do Ce^{144} and Y^{91} ; and the band of reaction behind the osteoid borders is more intense after Tm^{170} injection than after Ce^{144} and Y^{91} .

Seven days after injection the depositions of Ce^{144} and Tm^{170} have been displaced by new bone growth. The deposition beneath the epiphyseal plate appears as an interrupted band in the secondary spongiosa, with an area of decreasing intensity between it and the plate; this is secondary deposition of isotope released into the blood stream by resorption of areas of primary deposition (Fig. 23). In other areas of growth the depositions become buried by new bone and appear in cement lines or lines of reversal where resorption has ceased and growth started. Areas of deposition in completed haversian systems remain unchanged, and there is still a diffuse uptake throughout mineralized tissue.

The histological localization seen by autoradiography and micro-radiography shows that these three isotopes are deposited on highly mineralized bone surfaces. The second and third parts of the experiment were designed to verify this.

Because the Ce^{144} was carrier-free and the Tm^{170} was of low specific activity, the results of the tissue distribution studies of the two isotopes could not be compared quantitatively. The data and the comparison between the 24-hour and 7-day values for tissue specific activity are of no particular interest in this report; they have been published, however,⁽⁹⁾ and they agree with previous results.⁽⁵⁾

In Vivo Deposition

Table 21 shows the distribution of the isotopes in the organic and inorganic fractions of bone. It is apparent that only a small percentage of the activity is removed by the ED treatment although the small weight loss of ashed treated bone shows that all the organic material has been removed. Seven days after injection the percentage of activity in the organic fraction is even smaller. Approximately half the organic fraction is removed from the bone in the first six-hour extraction and with it 70 to 80% of the total activity in the organic component, the percentage rising to above 80% 7 days after injection. This suggests that most of the activity is attached to the readily available connective tissue. Both the autoradiographs and the soft tissue analyses show that the connective tissue contains considerable amounts of the isotopes 24 hours after injection but loses it fairly rapidly. The higher figures for the retention of Tm^{170} in the organic fraction are a reflection of the large amount of Tm^{169} carrier in the injection solution and do not mean that thulium is not as avidly retained by bone tissue as the other rare earths.⁽⁵⁾

Fig. 23. Longitudinal section from the femur of a dog killed 7 days after an injection of Tm^{170} . There is a spotty uptake in the cortical and trabecular bone with a strong reaction in the epiphyseal region. Secondary uptake in new bone formed between injection and death results in a fading of the uptake towards the plate. Magnification 3X.

Table 21

Distribution of isotopes in the organic and inorganic fractions of bone

Expressed as % activity in a whole bone

Isotope	Time post injection	Diaphyseal bone		Epiphyseal bone	
		Organic	Inorganic	Organic	Inorganic
Y^{91}	24 hrs	2.8	20.3	4.6	72.5
Ce^{144}	24 hrs	4.9	13.3	8.3	73.8
	7 days	0.08	19.5	6.1	76.5
Tm^{170}	24 hrs	4.1	14.5	13.9	67.5
	7 days	0.33	23.5	3.8	72.5

It is evident from Table 21 that the inorganic fraction remaining after ED treatment and ashing retains the major proportion of the isotopes and that most of the activity is in the epiphyseal bone, a fact substantiated by the autoradiographs (Fig. 23).

In Vitro Uptake

The results in Figure 24 show that whole bone, ED-treated bone and $Ca_3(PO_4)_2$ all take up the isotopes from the serum to a similar extent, whereas the organic samples do not retain significantly more than the control samples of sugar charcoal and Pyrex wool. Plasma was used as the incubating medium to reproduce the situation *in vivo*, and the small amount of activity in the organic and control samples represents incubating medium incompletely washed out of the thimbles and sample.

The ashed bone samples take up considerably less activity than the ED-treated bone or whole bone, suggesting that in the process of ashing the bone crystals are changed in a way that affects the uptake of the isotope by the sample. The presence of the citrate ion also changes the retention of the isotopes. The uptake is higher in each of the inorganic samples in the citrated medium than in the noncitrated medium. This difference disappears in the organic and control samples in which the uptake from the two media is not significantly different.

Discussion

There is a considerable amount of published data on the deposition of yttrium and the rare earths in bone which is interpreted to demonstrate the deposition of these elements in osteoid tissue.^(1,2, 5) However, these experiments may also be construed to show that the rare earths are deposited in areas of bone resorption. From the gross autoradiographs the

Fig. 24. Histogram demonstrating the results of the *in vitro* uptake of Y^{91} , Ce^{144} and Tm^{170} by various samples. The results are expressed as percentage of the total activity presented to the sample in the incubating medium.

observation is made that yttrium is not deposited in a calcium-like way, and therefore is not associated with bone calcification. Since osteoid formation and calcification take place in almost identical places, then the rare earths are probably not deposited in osteoid tissue but are taken up in areas where bone is not being formed. This interpretation would explain the difference between the localization of the rare earths and alkaline earths while the interpretations presented up until now do not.

MacDonald⁽¹⁰⁾ used Ca^{45} , S^{35} and Y^{91} in a study of healing fractures, and his data demonstrate that the retentions of these three elements in the fracture site are different. Calcium-45 is taken up as the fracture callus calcifies, S^{35} is taken up in the proliferating matrix of the callus, while the Y^{91} has a different retention curve from both Ca^{45} and S^{35} , for which MacDonald could find no satisfactory explanation. However, the Y^{91} curve may be adequately explained by the deposition of the isotope on the many resorbing surfaces produced by the osteoporosis taking place in the bone adjacent to the fracture. The retention of Y^{91} in rachitic rats reported by Copp⁽³⁾ may also be explained by deposition on nongrowing surfaces since rickets only affects calcification, not resorption. The high uptake in the region of the epiphyseal plate is the result of the extensive resorption that takes place here.

All the experiments concerned with the localization of the rare earths in bone may, therefore, be satisfactorily interpreted to support the thesis presented in this report, which is that the rare-earth elements are deposited on nongrowing bone surfaces. The first part of the experiment, the autoradiographic localization of the three isotopes in bone tissue, shows where these elements are deposited in vivo, and the results are expressed as observations. In the microradiographs areas of bone resorption appear highly calcified with irregular surfaces, while bone growth is characterized by low mineralization and a smooth surface covered by osteoid tissue. The three isotopes are deposited on the highly mineralized nongrowing surfaces in areas of resorption and inactivity, and are never found in the uncalcified osteoid tissue on growing surfaces. The diffuse uptake represents isotope that diffuses through the bone and becomes attached to bone mineral less available to the circulation than the inactive and resorbing surfaces bordering on vascular spaces. The secondary uptake is high; that is bone takes up these elements from low concentration in the blood, an observation that explains the continued retention of elements which are deposited to a considerable extent on surfaces that are being actively resorbed. The localization of the rare earths is demonstrated by the first part of the experiment, but there is no information as to the property of such nongrowing surfaces that might be responsible for the deposition. The second and third parts of the experiment answer this question.

Rare earths deposited in vivo are not removed with the organic fraction during ED-treatment but remain with the inorganic fraction. This indicates that the deposition of the isotopes on highly calcified bone surfaces seen in the autoradiographs is associated with the bone mineral and not with any organic constituent. If ED containing one of the three isotopes is refluxed through a piece of nonactive bone and the extracted bone washed three times, the amount of activity remaining behind in the bone is less than 0.1% of that in the ED solution; therefore, there is no chance that the results of the in vivo extraction will be invalidated by the ED removing isotope in an organic fraction and the isotope being redeposited on the bone mineral.

The third part of the experiment suggests that the substance responsible for the deposition in vivo is responsible for uptake in vitro. If sections of bone are incubated in a suitable solution containing yttrium chloride, deposition occurs all over the face of the mineralized surface of the bone (unpublished results), suggesting that bone mineral is the substance responsible for the deposition of the rare earths, but that diffusion is responsible for the restriction of the deposition to the more available surfaces rather than the interstitial bone or the mineral covered by the osteoid tissue in areas of growth. Thulium appears to be slightly more diffusible through bone matrix than yttrium and cerium. Ashing at 600° C reduces the uptake of activity, probably as a result of reducing the available crystal surface by fusing the crystals together or by changing the outer layer of the crystals in such a way as to prevent adsorption. A comparison of the uptake of activity from the citrated and noncitrated media demonstrates the importance of the state of the isotope in the incubating medium. The citrate ion appears to hold the elements in a more easily broken complex than the plasma proteins, thus making them more available for uptake by the bone mineral. In contrast the second part of the experiment has shown that ED holds the activity in such a way as to prevent its deposition on bone mineral.

The results from the three experiments are consistent with one conclusion: Histologically the three isotopes are localized on nongrowing, highly mineralized bone surfaces. Extraction with ED of in vivo deposited isotopes shows the elements are associated with the inorganic fraction of bone, while the ability of bone salt to take up and retain the activity is demonstrated in the in vitro experiment. The rare earths must, therefore, be deposited in vivo on bone mineral, wherever this is available to the complexes in the blood. The mechanism of deposition is possibly one of surface adsorption. Growing surfaces are covered by a layer of osteoid tissue, and within the substance of the bone itself diffusion is difficult; so these elements are deposited on the available nongrowing surfaces of the skeleton where resorption is or has been taking place or where growth has stopped and calcification is relatively complete.

Summary and Conclusions

1. After intraperitoneal injection, the rare-earth isotopes Y^{91} , Ce^{144} and Tm^{170} are taken up in the skeleton and appear on nongrowing bone surfaces that microradiographs show to be highly calcified. They are not found in osteoid tissue.
2. These three isotopes remain in the inorganic fraction of bone after ethylenediamine extraction of the organic fraction from the bones of injected animals.
3. Bone mineral will take up these rare-earth elements from plasma in vitro; organic material from bone will not.
4. The three rare-earth isotopes investigated are taken up by bone mineral and are localized on the available resorbing and inactive surfaces of bone tissue.

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MASS PER UNIT VOLUME OF BONE MINERAL

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

The range of densities shown by the elements found in very small amounts in bone is essentially independent of age, at least in the dog.⁽¹⁾ This study of bone mineralization has now been extended to include other species and the human by employing microradiographic techniques already described.⁽²⁾

In Table 22 are tabulated values of bone mineralization found in two different bones in humans of different age. Both the femur and the clavicle have secondary haversian systems, but these bones vary widely in size, shape, and function. From this table it is apparent that both newly-formed haversian systems and old bone are found at practically the same density in each location. It is also apparent that, in the human, there is no obvious shift in the range of mineralization with increasing specimen age.

Table 22

Observed values of mineral density in human bone, expressed as grams of hydroxyapatite per cm³

Age	Femur		Clavicle	
	New growth HA*, g/cm ³	Old bone	New growth HA*, g/cm ³	Old bone
7 yrs.	0.95	1.29	0.90	1.21
31 yrs.	1.11	1.40	1.03	1.31
76 yrs.	0.98	1.28	1.12	1.37

*hydroxyapatite

In Table 23 are tabulated the values of mineral density found in new growth and old mineral for several different species. In this table the values for the mouse, rat and rabbit do not refer to secondary haversian systems, but to newly-formed periosteal or endosteal bone.

In contrast to the findings mentioned above for a single species, it is apparent that the density at which bone is formed and the final density it reaches do vary from species to species. These differences must be taken into consideration when extrapolating data from one species to another. In particular, these differences may be of significance in the interpretation of the mechanism of radioisotope uptake from the blood and in the subsequent loss from bone to blood.

Table 23

Observed values of mineral density in various animals, expressed as grams of hydroxyapatite per cm^3

Animal	Density, HA, * g/cm^3		Bone examined
	New growth	Old bone	
Man	0.90-1.11	1.21-1.40	femur
Dog	1.02-1.25	1.23-1.54	femur
Mouse	1.18-1.25	1.36-1.39	femur
Cow	1.11	1.50	tibia
Rabbit	1.39	1.53	tibia
Rat	1.40-1.42	1.69-1.70	femur

*hydroxyapatite

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Ca^{45} DEPOSITION AND GROWTH RATE IN CANINE OSTEONS

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

I. Introduction

When calcium-like radioactive nuclides are injected into the blood stream, intense concentrations deposit in some of the osteons in cortical bone. These concentrations of activity are interesting both because they may be a factor in the long-term radiation damage produced by Sr^{90} or Ra^{226} in bone, and because, at least for Ca^{45} , they reflect the dynamics of calcium metabolism between the blood and individual osteons in living bone. A three-dimensional autoradiographic study of Ca^{45} in osteons has been reported.⁽¹⁾

Three stages are known to occur in the formation of an osteon (secondary haversian system). First, by the process of resorption a roughly cylindrical cavity 100-150 μ * in diameter and the order of a millimeter long* is dug within previously existing bone. Secondly, new bone is deposited in layers around the walls of this cavity until the remaining canal, containing a blood capillary, has a diameter of about 20 μ .* As this new bone is deposited, it rapidly attains a mineral density of about 70% of that of primary bone. Thirdly, the mineral density of the new bone increases very slowly to that of the surrounding primary bone.⁽²⁾ If Ca^{45} is present in the blood, it will be taken up by an osteon during both the second and third stages along with stable calcium and be built into bone mineral. In addition to this accretion of calcium, an exchange of Ca^{45} for already-deposited Ca^{40} may be expected to occur. Such exchange has been demonstrated in vitro by Neuman,⁽³⁾ Amprino,⁽²⁾ and others, and has been invoked as the process of P^{32} deposition in living osteons.⁽⁴⁾ On the other hand, Arnold, Jee and Johnson⁽⁵⁾ have attributed the Ca^{45} concentrations in growing osteons of rabbits to accretion of calcium. Probably both exchange and accretion occur in osteons, the relative magnitudes of the two processes depending on the particular stage of formation of the osteon. It is our purpose to present autoradiographic measurements of the total Ca^{45} content of individual osteons in dogs following a single injection and to compare them with the Ca^{45} uptake expected from accretion of calcium during the second stage of osteon formation, appositional growth. An expression for the appositional growth rate of an average osteon, consistent with the observed Ca^{45} content of forming osteons as well as with direct measurements of osteon growth, is derived.

*These values apply to osteons of dogs

II. Method

Three adult mongrel dogs were given single, low level, intravenous injections of Ca^{45} (0.05 mc/kg body weight). Two were sacrificed 12 hours after injection, the other was sacrificed 2 weeks after injection (as shown in Table 24). The diaphyses of long bones from each dog were dehydrated in alcohol and ether and embedded in Bioplastic. Cross sections were cut to a thickness of 125 microns, a distance short compared to the length of most Ca^{45} deposits in osteons⁽¹⁾ but comparable to Ca^{45} beta-particle range so that the beta-particle emission from a section was over 95% of that of a thick source. Each cross section was sandwiched between two No Screen Autoradiographic plates. A typical autoradiogram is shown in Fig. 25. This low-resolution, thick-source technique makes possible a relatively simple method of absolute quantification.⁽⁶⁾

Table 24

Dog	Time between Ca^{45} injection and sacrifice T_s	Age	Region from which Data in Figure 27 were taken	
			Approximate number of osteons forming per cm^2 with $b > 40 \mu$	Bone location
C119	2 weeks	Adult	10	Femur midshaft
			20	Femur end of shaft
C120	12 hours	1 year	300	Humerus midshaft
C122	12 hours	6 years	10	Femur midshaft

Selection of Ca^{45} Deposits in Osteons for Longitudinal Invariance

In order to relate the observed quantities of Ca^{45} to the properties of osteons, it is necessary to eliminate the complexity introduced by the interconnecting osteon network. This has been done by using cross sections of long bones so that most osteons are perpendicular to the film and by choosing Ca^{45} deposits which are relatively invariant with depth in the given section. The two autoradiograms from each cross section were compared by projecting them alternately on a ground glass screen at a magnification of 30-100 and by tracing the spots of Ca^{45} darkening. Spots which were not nearly identical on the two autoradiograms were eliminated from further

Fig. 25 Ca^{45} autoradiogram and corresponding long-exposure microradiograph from the midshaft of the humerus of dog C 120.

analysis. Microradiographs of each bone section, taken with continuous X-rays from a tube operating at 12 kilovolts, were similarly projected and compared with the autoradiograms. Two different microradiographic exposures were used: one to reveal the small differences in high calcium density between osteons and primary bone, and another shorter exposure to reveal differences in low calcium density so as to distinguish between osteon canals which were oblique and those which continued perpendicularly through a section. By this comparison of two autoradiograms and two microradiographs, Ca^{45} deposits were selected which ran perpendicular to and continued through a section without noticeable variation in either the Ca^{45} activity distribution or in the structure of the corresponding osteon. This selection eliminated from further analysis about half the Ca^{45} spots from an average autoradiogram, as was to be expected from the three dimensional pattern of Ca^{45} deposits.⁽¹⁾ The present analysis does not include osteons with arrested growth, that is, low density osteons with canals larger than 20μ diameter but negligible Ca^{45} uptake, although several were observed. The activity per unit length and the canal diameter of each of the selected active osteons were measured as follows.

Activity per Unit Length - h

The total activity per unit length of a Ca^{45} deposit was determined from the area integral of the film darkening which it produced. Measurements from each of the pair of autograms were averaged. Film darkening was measured by a microdensitometer with an aperture 30μ in diameter and calibrated by exposures of the same autoradiographic plates to a thick plaster-of-Paris radiator containing a known Ca^{45} activity per gram. Correction for slight nonlinearity in film response was made by referring to a curve of darkening versus radiator exposure.

Provided that:

- 1) The distribution of Ca^{45} activity in the deposit is invariant with depth in the (thick) bone section, and
- 2) The osteon's canal is filled with an embedding medium of the same density, and approximately the same atomic composition as the surrounding bone, it follows (6) that

$$h = \frac{\rho}{kF} \iint D \, dx \, dy \quad (1)$$

where

h = activity per unit length ($\mu\text{c}/\text{cm}$) of Ca^{45} deposit in bone

ρ = density of bone and embedding material seen by beta particles (g/cm^3)

F = exposure time for bone (days) corrected for decay if necessary

k = calibration factor for film derived from exposure to thick uniform radiator (darkening $\text{g}/\mu\text{c day}$)

D = film darkening, a function of position, corrected for linearity and film background.

The area integral in expression (1) was evaluated by summing a series of closely-spaced net densitometer readings across each region of darkening. This procedure yields the Ca^{45} activity per unit length in an osteon irrespective of the radial distribution of activity.

The first of the above provisions, depth invariance, was satisfied by the selection of data. The second provision, homogeneity of the source, was not exactly satisfied. The bone of a young or forming osteon is generally surrounded by bone of about 30% higher density whereas the Bioplastic embedding medium within its canal had a density about 30% lower than that of the newly-formed bone. In osteons with large diameter canals, for which the inhomogeneity is greatest, the two errors work in opposite directions. That Bioplastic actually filled canal spaces was checked under an incident illumination microscope. Thus by ignoring inhomogeneity and using the density of newly-formed bone for ρ in (1), we should not introduce an uncertainty in h greater than about 30%. The value $\rho = 2 \text{ g/cm}^3$ has been used in (1) but cancels out in the results given by expressions (4) or (7).

Since the values of $D(x,y)$ in (1) were measured from the film background and integrated over the entire darkening distribution of each spot, the measured activity per unit length for an osteon corresponds to the total Ca^{45} activity in the osteon whether that activity is associated with the newly-forming lamellae around the canal or with a diffuse uptake in the recently-formed bone between canal and cement line. The probable error in the measurement of activity per unit length is 30%. All measurements of h are normalized to an injected Ca^{45} activity of 0.1 millicurie/kilogram body weight.

Canal Diameter - b

The diameter of the canal in each osteon was measured from the longer exposure microradiograph and corresponds to the inner diameter of the calcified region, or more exactly, to the sharply defined perimeter at which calcium density changes from approximately zero to 70% that of primary bone. For canals not approximately circular, the average of the maximum and minimum diameter was used. No osteons were included whose maximum exceeded their minimum diameter by a factor of two or more.

Blood Specific Activity - B

The measurements of blood specific activity B ($\mu\text{c Ca}^{45}/\text{gCa}$) as a function of time t for five Ca^{45} -injected adult dogs, reported previously,⁽⁷⁾ are shown in Fig. 26, together with the time integral of B from injection to

Fig. 26. Blood specific activity B in mc/gCa as a function of time t after a single injection of Ca^{45} taken from data for five adult dogs used in reference 7. Measured values extend from 10 minutes to 100 days and are normalized to an injection of $0.1 \text{ mc/kg body weight}$. Between 1 day and 100 days the data are proportional to $t^{-1.2}$. The initial point on the B curve is the total injected activity divided by the calcium content of the blood plotted at about one circulation time. The time integral of B is given in mc-days gCa . Only 5% of the value of the integral at 12 hours is contributed by the first 10 minutes after injection. The integral increases by a factor of 3 between 12 hours and 2 weeks and by a factor of 1.4 between 2 weeks and 1 year.

Fig. 27. The total Ca^{45} activity per unit length h in $\mu\mu\text{c/mm}$ in individual osteons of dogs listed in Table 24 plotted against the diameter of the osteon canal b in microns. The values have been normalized to a Ca^{45} injection of $0.1 \text{ mc/kg body weight}$. Curve A is given by expression (7) with $T_s = 2 \text{ weeks}$, $\beta = 0.04 \text{ day}^{-1}$ and curve B by $T_s = 12 \text{ hours}$, $\beta = 0.024 \text{ day}^{-1}$; curve C (dashed) is given by expression (4) with $T_s = 12 \text{ hours}$, $\frac{db}{dt} = -2.4 \text{ microns, day}$. Curves A and B show $h \sim b^2$. Curve C shows $h \sim b$. Curve A does not extend to the largest values of b because these values represent osteons which have started forming after injection.

time t . The three dogs used here were also injected by J. Cohen and with Ca^{45} standardized in the MIT 4 π counter, but blood samples were not taken. However, individual variation from the data of Fig. 26 is expected to be less than a factor of two. The integral is not strongly dependent upon the blood specific activity values at an hour or less, so that a time delay before interstitial fluid reaches equilibrium with the blood (probably the order of 15 min) can be neglected. Note that the value of the integral is three times as great at 2 weeks as at 12 hours.

III. Results

Experimental

The measured activity per unit length for the selected osteons in the three dogs is plotted in Fig. 27 versus canal diameter. All activity values have been normalized to an injection of 0.1 millicurie per kilogram body weight. The relative numbers of points do not indicate a frequency distribution because many sections were examined to find the larger values of h and b . The points for the two dogs sacrificed at 12 hours fall considerably lower than the points for the dog sacrificed at 2 weeks, and separate curves have been fitted to the data as described below. A displacement of the two sets of points may be expected for two reasons; increased uptake of Ca^{45} at 2 weeks as compared with 12 hours and a decrease in canal diameter due to osteon growth in the 2-week interval as compared with the 12-hour interval following injection. There may also be a displacement due to differences in the actual blood curves or in the animals used.

Theoretical

Let us calculate the Ca^{45} deposition expected in osteons for given rates of appositional growth and then compare the calculated values with the data. Let us assume that

- 1) the specific activity (Ca^{45}/Ca) seen by an osteon is equal to the specific activity of the blood measured in the large veins, and that
- 2) the Ca^{45} activity observed in osteons at 12 hours, and 2 weeks has been deposited by accretion of calcium (and does not represent exchange of Ca^{45} for Ca^{40}).

Under these two assumptions, which we will discuss in view of the results, we can calculate the rate of calcium accretion per unit length in an osteon from the observed Ca^{45} activity per unit length and blood specific activity measurements. The activity per unit length h is given by

$$h = \int_0^{T_s} g B dt \quad (2)$$

where

h = total $\mu c/cm$ in osteon

g = accretion rate per unit length of osteon ($g_{Ca}/cm \text{ day}$)

B = blood S.A. ($\mu c/g_{Ca}$)

T_s = interval between injection and sacrifice (days)

We can describe a forming osteon as a cylindrical shell with a fixed outer diameter and with an inner diameter, b , which decreases with time due to appositional growth of new bone. The rate of calcium deposition g resulting from a given rate db/dt of decrease of canal diameter is

$$g = -\frac{\pi \rho c}{2} \left(b \frac{db}{dt} \right) \quad (3)$$

where

b = inner (canal) diameter (cm)

ρ = density of newly-formed bone (g_{bone}/cm^3)

c = calcium content of newly formed bone (g_{Ca}/g_{bone})

There will also be some accretion associated with the slow increase of ρc with time, but for the purposes of this calculation we can neglect this secondary mineralization and use a constant value of ρc .

Combining (2) and (3) we can calculate the total Ca^{45} activity per unit length that would be deposited in an osteon of given canal diameter by a rate of appositional growth db/dt =

$$h = \frac{\pi \rho c}{2} \int_0^{T_s} b \left(\frac{db}{dt} \right) B dt \quad (4)$$

For short intervals between injection and sacrifice in which b and $\frac{db}{dt}$ may be considered constant, expression (4) yields rates of appositional growth $\frac{db}{dt}$

from measurements of h and b . For the 12-hour data in Fig. 27, any point on the dashed curve corresponds to a value of h and b such that $db/dt = 2.4 \mu/day$. Our assumption of constant b and db/dt for 12-hour data is therefore self-consistent because the canal diameter decrease during 12 hours at this rate is negligible. For an interval as long as 2 weeks between injection and sacrifice, however, canal diameter decrease for an individual osteon will no longer be negligible, and we must take into account changing values of the accretion rate g within the integral.

The simplest assumption for the variation of g with time would be that the osteon canal diameter has a constant rate of decrease db/dt , in which case an average of the 12-hour data would indicate that a 120μ diameter osteon takes

6 weeks to fill in to a final canal diameter of 20μ . However, the data are better fitted when we take into account the fact that an osteon fills in more rapidly at first than it does when nearing completion.⁽⁹⁾ Let us represent a decreasing rate of linear growth by an exponential:

$$b = b_0 e^{-\beta t} \quad (5)$$

$$\frac{db}{dt} = -\beta b \quad (6)$$

where b_0 refers to the initial canal diameter of an osteon; its outer or cement line diameter, β is a constant rate factor (day^{-1}), and b gives canal diameter as a function of time until, at a final diameter of 20μ , expressions (5) and (6) are no longer applied. This somewhat arbitrary mathematical description of the average forming osteon in dog provides a reasonably good fit to the data. Inserting (5) and (6) into (4) gives an expression in which activity per unit length is proportional to the square of the canal diameter at any time after injection:

$$h = b_s^2 \left[\frac{\pi \rho c}{2} \beta e^{2\beta T_s} \int_0^{T_s} e^{-2\beta t} B dt \right] \quad (7)$$

where b_s = canal diameter observed at time of sacrifice and other symbols are as defined above.

Expression (7) yields the solid curves through the data in Fig. 27, the C119 data giving $\beta = 0.04 \text{ day}^{-1}$, and the C120 and C122 data giving $\beta = 0.024 \text{ day}^{-1}$. The difference between these values is not significant, considering the possible errors of the measurements.

Summary of Results

Under our assumptions that the observed Ca^{45} was deposited by accretion at the specific activity of the blood, at either 12 hours or 2 weeks after injection the data may be described by an osteon apposition rate given by expression (6) with $\beta = 0.03 \pm 0.01 \text{ day}^{-1}$. This is equivalent to a rate of formation of an average osteon such that its canal diameter decreases to half its value in 3 ± 1 weeks. At this rate, an average osteon with outer diameter 120μ would have almost completed appositional growth by reaching a canal diameter of 30μ in 6 ± 2 weeks.

IV. Discussion

Comparison of 12-Hour and 2-Week Data

Although intercomparison between animals is here subject to an error of perhaps a factor of two, the similarity of the values of β derived from the 12-hour and 2-week data suggests that forming osteons contain no large amount

of Ca^{45} in an exchangeable fraction of bone salt in equilibrium with the blood at 12 hours or more after injection.⁽⁸⁾ The presence of such short-term exchangeable activity would make the 12-hour value of β larger than the 2-week value because during this period blood specific activity falls by a factor of 25-30. However, such a comparison by itself does not prove that the observed Ca^{45} activity is associated primarily with accretion. The possible influence of resorption is eliminated by the nature of the data: on the microradiographs resorption cavities are easily recognizable and did not encroach on any of the selected osteons. But we have not yet eliminated the possibility of long-term or nonequilibrium exchange which might arise from recrystallization or intracrystalline diffusion in bone existing at the time of injection.⁽³⁾

Comparison of Results with Direct Observations of Osteon Growth - Conclusions

How does a canal diameter half-time of 3 weeks compare with direct observations of osteon growth? High resolution osteon-labelling studies by Jee and Arnold⁽⁹⁾ in young rabbits indicated that of the osteons which were just beginning formation at the time of Ca^{45} administration, 20-30% were completed in 21 days although they did not show the distribution of growth rates. Vincent,⁽¹⁰⁾ using high resolution S^{35} labelling, concluded that an osteon in adult dog takes about 6 weeks to complete appositional growth. Thus the osteon appositional growth rate derivable from the total Ca^{45} activity in an osteon after a single injection agrees at least within a factor of two with that observed directly by the use of high resolution labels. It is conceivable that both our basic assumptions could be in error in opposite directions. If capillary specific activity were lower than average blood specific activity and Ca^{45} uptake were higher than that due to accretion alone, the effects might cancel. Also, from the present data we cannot determine the nature of the Ca^{45} uptake in almost completed osteons with small canal diameters. With these two qualifications we may conclude that (1) the specific activity of calcium being built into new bone in osteon formation is essentially the same as that measured in the large veins and that (2) the intense deposits of Ca^{45} found in osteons are due primarily to accretion of calcium.

V. Some Calculations Based on the Observed Growth Rate of an Osteon

Distributions of Forming Osteons and Ca^{45} Deposits

If in a large group of osteons the number which have just started to form per day has remained constant for 2 months or more, we would expect to find forming osteons with all values of canal diameter from b_0 to $b_{\min}$. Since according to Expression (5), an individual osteon spends more time at small diameters than at large, small diameter osteons should predominate, the number of canal diameters per unit diameter being inversely proportional to diameter. The distribution of activities per unit length h

following a single injection would also be hyperbolic, the number of osteons with activity between h and $h + dh$ being inversely proportional to h . Observed distributions of b or h in C119 and C122 agree with this hyperbolic shape fairly well. C120, however, shows a peaked distribution of both b and h , probably because a burst of new osteon formation in this 1-year-old dog has recently occurred so that equilibrium does not exist.

The bell-shaped peak darkening distribution reported by Arnold⁽⁵⁾ for forming osteons is not strictly comparable to an activity per unit length distribution because of the varying effect of beta-particle resolution for osteons of different canal diameters. Activity per unit length distributions with approximately hyperbolic shape can also be derived from the premise that deposition of calcium in osteons occurs intermittently during intervals of about a day,⁽¹¹⁾ but this calculation predicted a strong dependence of shape upon the interval between injection and sacrifice which has not been observed and fails quantitatively to yield observed growth rates.

Time for Formation of a Resorption Cavity

The number of resorption cavities in C119 was smaller than the number of forming osteons by a factor of 2-3; assuming an equilibrium between resorption cavity and osteon formation in this animal, we estimate that the tunnelling process at a given site takes roughly three weeks.

Total Accretion Rate Due to Osteon Formation

A rate of accretion and bone turnover can be related to the number of forming osteons observed in bone. If the average cross section contains 10 hotspots/cm² corresponding to forming osteons with canal diameters larger than 40μ at the time of injection, then it follows from Expression (5) that the accretion rate in the bone sample due to osteon formation is about 0.004%/day. Accretion rate is here calculated as grams bone per gram of bone/day for $b_0 = 120\mu$ and $\beta = 0.03 \text{ day}^{-1}$, and it includes secondary mineralization as well as apposition, assuming equilibrium numbers of osteons at each stage of formation. The number of forming osteons per unit area is difficult to predict for individual animals because it depends strongly upon their metabolic state. (See Table 24) However, the figure 0.004%/day derived for the femur midshaft of C119 or C122 may be typical of normal adult dogs. The corresponding mean time of bone turnover would be about 70 years. The mean turnover time by osteon formation in young C120 would be about 2 years. If the mean time of bone turnover by resorption and new osteon formation is as slow in adult humans as it is in the older adult dogs, it is difficult to explain the elimination of radium from humans by this process though it is then easy to understand the persistence of hotspots for many decades.

Calculation of Calcium Supply to an Individual Osteon

The calcium accretion rate of an individual osteon which we have derived may be compared to the quantity of calcium supplied to it by its capillary. The rate of blood flow in an $8\ \mu$ diameter capillary in dog has been calculated to be very roughly $0.7\ \text{mm/sec}$ ⁽¹²⁾ and if we take a normal calcium concentration of $55\ \mu\text{g Ca/ml}$ blood, then the calcium which flows by a particular point in an osteon is about $0.16\ \mu\text{g Ca/day}$. The calcium accretion rate per unit length in an osteon with a canal diameter of $100\ \mu$ is about $0.2\ \mu\text{g Ca/mm day}$ (using $\beta = 0.03\ \text{day}^{-1}$). The length of osteon over which the most rapid calcification occurs at any given time is the order of a millimeter,⁽¹⁾ so that the rate of calcium deposit is the same order of magnitude as the rate of calcium supply. This suggests that the length of an osteon over which rapid calcification can occur may be limited to the order of a millimeter by a limitation in the calcium deliverable by a capillary, and that a rapidly-forming osteon probably retains a large fraction of the calcium flowing by it. We would, therefore, expect to find little exchangeable calcium in rapidly-forming osteons because little calcium is left over to transfer from bone to blood at the observed rates of growth. Perhaps one function of the excess calcium concentration in the blood over that required for calcification⁽³⁾ is to pump calcium to longer regions of forming osteons than would otherwise be possible.

Specific Activity in a Capillary

A fall in calcium concentration in a capillary passing a growing osteon would not by itself reduce the specific activity (Ca^{45}/Ca) of the calcium seen by the osteon below that of the blood and extracellular fluid as a whole, our first basic assumption. If the number of resorbing cavities per unit volume were large, however, and if the resorption process immediately released calcium to the ionic form (not known) the release by resorption of relatively inactive calcium to capillaries might become significant. Cohen and Harris⁽¹³⁾ have found that longitudinal capillaries in cortical bone of dogs spiral slowly from endosteum to periosteum over a distance of about a centimeter. Thickness of the cortex in a long bone of a dog is usually a couple of millimeters. Thus, although we do not know the pattern or direction of blood flow in the capillaries, it is reasonable to assume that the average capillary path from artery to vein in cortical bone is more than a couple of millimeters but not much more than a centimeter. In an animal such as C120 which is in a burst of osteon formation, a cross section shows that about one out of every 10 canals is in a rapidly-growing osteon. If we take the length of rapid growth to be $1\ \text{mm}$ ⁽¹⁾ and the capillary blood path to be $1\ \text{cm}$, then an average blood pass through cortical bone will encounter one region of active osteon growth, and one out of two or three passes will encounter a resorbing cavity. The chance of a resorbing cavity lying upstream from a rapidly forming osteon will thus be about 20%. Therefore, in C120 there may be some reductions

of capillary specific activity below that of the large vessels due to dilution from resorbing cavities. This effect might contribute to a low β value for C120, but it should be negligible for C119 or C122 which showed less than one capillary in a hundred to be in a rapidly growing osteon.

Hotspot Migration

This same type of calculation may be applied to the hypothesis of hotspot migration⁽¹⁴⁾ in which it is postulated that an intense radioactive deposit may have moved from a previous location in bone to its present position without appreciable dilution with inactive mineral. In the light of the above calculation we conclude that migration of a calcium-like deposit by resorption and redeposition in a single capillary pass through bone is conceivable but improbable, especially in adult animals. Once the resorbed activity reached the general circulation it would be diluted rapidly in the whole blood volume and be redeposited all over the skeleton only at the existing specific activity of the blood as a whole ⁽⁷⁾

VI. Summary

The Ca^{45} content of individual osteons in three dogs given single intravenous injections has been measured autoradiographically and correlated with osteon canal diameter measured from microradiographs. An osteon was found to contain up to 10^{-6} of the injected activity, its total activity per unit length being approximately proportional to the square of its canal diameter at the time of injection. The activities observed at 12 hours or 2 weeks after injection divided by the corresponding time integral of the blood specific activity yield an accretion rate for calcium at each canal diameter such that the half-diameter time for the average forming osteon in a rather wide distribution is 3 ± 1 weeks, which is consistent with direct observations of osteon growth. It is concluded that the intense concentrations of in vivo-deposited Ca^{45} observed in osteons 12 hours or more after injection may be satisfactorily explained as accretion of calcium in appositional growth at approximately the specific activity measured in the large veins. The nature of the Ca^{45} uptake in osteons which have completed or arrested appositional growth cannot be inferred from the present data, though its magnitude is probably considerably less than that due to appositional growth. Comparison of calculated blood flow with observed osteon growth rate indicates that a rapidly-forming osteon probably accretes a large fraction of the calcium reaching it by its capillary. Rates of accretion and resorption, the shapes of osteon-hotspot distributions, the specific activity in a capillary, and hotspot migration are discussed.

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VARIATIONS IN HAVERSIAN SYSTEM SIZES

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

The haversian system is a unit of bone structure found in most animals with a bony skeleton. We wanted to find out if the size of the system varied from animal to animal depending on the age, body weight or the length of life of the animal.

Haversian systems were measured in ten different animals: one rabbit, two dogs, two monkeys, one small early Permian reptile (Diadectes sp.), two humans, one adult cow and a large Cretaceous dinosaur (Iguanodon sp.). Only secondary haversian systems in the shaft of the long bones were measured. That is, systems formed by remodeling of primary haversian or non-haversian cortical bone. Microradiographs of cross sections of bone were used, and the diameters of the cement lines surrounding the haversian systems were measured at a magnification of x 50, two dimensions being taken at right angles to each other and averaged. Only systems cut in cross section with approximately circular central canals were taken for comparison. From 25 to 200 systems were measured for each animal and the number of systems of different sizes arranged to form a histogram.

The series of histograms for the ten animals show that there is a difference of approximately a factor of two between the cement line diameters of a rabbit and a dog and another factor of two between those of a dog and of a human. Assuming that the rate of laying down of bone tissue is a property of the tissue rather than the animal, then a rabbit's haversian systems may be expected to be formed in a shorter time than a dog's and a dog's in a shorter time than a human's. Data from autoradiographic studies suggest a time of formation, not including secondary mineralization, of three weeks for a rabbit and six weeks for a dog (1, 2, and 3). Human haversian systems may then be expected to form in approximately twelve weeks

An attempt to correlate the haversian system size consistently with any other biological variant proved unsuccessful, possibly because too few animals were used, and possibly because there is no correlation. The differences in body weight and life span between the animals studied varied by factors of a thousand and more, yet the haversian system sizes varied at most by a factor of four, and did not vary at all in the larger animals.

Fig. 28

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THE PARADOX OF DIFFUSE ACTIVITY AND LONG-TERM EXCHANGE OF CALCIUM IN LIVING BONE

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

Introduction

When Ca^{45} is taken up by bone, activity deposits not only in regions of appositional growth but also in bone already existing at the time of injection. This Ca^{45} deposition in preexisting bone, which we will refer to as diffuse activity, produces a relatively uniform low level of darkening in an autoradiogram. The mechanism of this diffuse uptake of Ca^{45} is not clear. If it represents exchange of Ca^{45} atoms from the blood with Ca^{40} atoms in bone mineral, why does the Ca^{45} activity remain in bone long after the specific activity of the blood has dropped to a negligible level? If, as some suggest, this diffuse Ca^{45} deposition is irreversible, then why is there not a large increase with time in the calcium density of the bone? This paradoxical situation has been discussed in terms of recrystallization,⁽¹⁾ but most of the pertinent experiments⁽²⁾ have been performed in vitro under conditions which may differ from those in living bone. There have been many excellent experiments in vivo which support the findings in vitro, but it has been difficult to avoid ambiguity completely in the results, because accretion followed by resorption of Ca^{45} activity is macroscopically indistinguishable from exchange, and because at least part of the diffuse uptake of Ca^{45} in existing bone may be associated with secondary mineralization. (Definitions of these terms as used in this paper are given in the Appendix.)

The purpose of the present investigation is to measure autoradiographically the magnitude of the diffuse component of Ca^{45} uptake in cortical canine bone, to relate this uptake to the specific activity of the blood after a single Ca^{45} injection in order to derive the local rate of calcium uptake, and to obtain microradiographic measurements of the microscopic calcium density in the same region of bone as a function of time. Thus, we have studied the phenomenon of Ca^{45} uptake by preexisting bone in vivo, independently of the effects of appositional growth and resorption in neighboring regions of bone or elsewhere in the skeleton, and with discrimination between the irreversible Ca^{45} uptake due to secondary mineralization, and an apparently irreversible Ca^{45} uptake due to long term exchange.*

Method

Adult mongrel dogs were given single low level (0.02-0.2 mc/kg body weight) intravenous injections of $\text{Ca}^{45} \text{Cl}_2$ of high specific activity. Specific activity measurements of the blood ($\text{mc Ca}^{45}/\text{g Ca}$) were made

*See terminology in Appendix

on five of the animals⁽³⁾ and a Ca^{45} blood curve and its integral derived.⁽⁴⁾ Long bones were scraped clean, dehydrated in alcohols and ether and embedded in Bioplastic. Cross sections 150μ or more in thickness were cut through the shaft with a circular saw, smoothed on ground glass, and autoradiographed on Eastman No Screen Autoradiographic plates. Control sections of bone containing no activity produced no exposure. The diffuse darkening was found to be proportional to exposure time and to injected activity per unit body weight. The specific activity of the diffuse component was derived from densitometer measurements of film darkening calibrated with thick plaster-of-Paris radiators containing known Ca^{45} activities per gram.⁽⁵⁾ The calcium content of highly mineralized, dehydrated cortical bone was taken to be $0.25 \text{ g}_{\text{Ca}}/\text{g bone}$. All specific activity values have been normalized to an injected activity of 0.1 mc/kg body weight.

Microradiographs were made of bone sections 100μ in thickness with continuous X-rays from a 12-kv tube filtered above the calcium absorption edge by 1 mm of Be. The superficial calcium density ($\text{g}_{\text{Ca}}/\text{cm}^2$) was determined by comparison with an aluminum step wedge.⁽⁶⁾ Section thickness at the point of interest was measured by the substage probe method using incident illumination.⁽⁷⁾

Microscopic Scale of Measurement

The microscopic measurements of diffuse specific activity reported here refer to specific activity as observed on an autoradiogram by a microscope densitometer with an aperture of 30μ . The half-thickness for absorption of Ca^{45} beta particles is also about 30μ , so structures smaller than this are not resolved by the present technique. In regions of cortical bone containing no secondary haversian systems or resorption cavities, the Ca^{45} diffuse activity appears to be uniform (with the exception of the rare avascular regions mentioned below), but this observation, of course, implies nothing about uniformity of specific activity on the crystallite level (tens or hundreds of Angstroms).

The microscopic measurements of calcium density refer to the grams calcium per cubic centimeter as observed on a microradiograph through a 10-micron aperture. The volumes measured are thus $10 \times 10 \times 100$ microns and appear uniform in density, because they contain no cavities larger than canaliculae, which are not resolved on the microradiographs. The present measurements of calcium density are thus not directly comparable to those measured for bone on the cubic centimeter level. They, of course, measure only the net effect of any changes in the number, volume, or calcium density of individual crystallites on the Angstrom level.

Results

Magnitude of Ca^{45} Diffuse Activity as a Function of Time

Autoradiographic measurements of the specific activity of the diffuse component in the shafts of the long bones of six dogs are plotted in Fig. 30 as a function of time after Ca^{45} injection. The dogs were adult mongrels of unknown age which were used in a previous experiment.⁽³⁾ The estimated standard error of each measurement is indicated by the error bar. Different low-level injections were used, the plotted values being normalized to an injected activity of 0.1 mc/kg body weight. The average value of about $1 \mu\text{c/gCa}$ corresponds to about one half the average specific activity of the skeleton. The values of diffuse specific activity remain constant within a factor of two from 2 weeks to 3 months after injection. During this period the blood specific activity falls by a factor of 10, and the integral of the blood curve rises by only a factor of 1.3.⁽⁴⁾ Within the error of our measurements, therefore practically all of the Ca^{45} diffuse activity at 2 weeks or longer after injection follows the integral of the blood specific activity rather than the blood specific activity itself. Therefore the activity observed indicates a substantially one-way flow of Ca^{45} from blood to bone.

Rate of Calcium Uptake Indicated by Ca^{45} Diffuse Activity

Since Ca^{45} is retained in the diffuse component at least over the period indicated above Ca^{40} must similarly be retained. Certainly the skeleton does not distinguish between these isotopes. Therefore we can calculate the rate of uptake of calcium in bone which gives rise to the diffuse component, knowing that in times the order of a month after this calcium is deposited it is not returned to the blood. Let us define this rate of calcium uptake as a calcium augmentation rate since if unopposed by a corresponding rate of loss, it would increase the calcium density of a microscopic region of existing bone.

[It is shown below that curiously enough, there is no corresponding increase in calcium density in the areas of periosteal bone studied so that the augmentation in these regions is not the net uptake of calcium but is opposed by a comparable rate of diminution. The use of two new words is useful because they represent observable quantities which pertain to phenomena, the molecular description of which is not yet clear (variously referred to as recrystallization, intracrystalline diffusion, irreversible exchange, etc.). The rate of increase of microscopic calcium density in secondary mineralization is thus the augmentation rate minus whatever rate of diminution may exist at a particular location.]

Under the assumption that the diffuse activity is deposited at the specific activity of the blood, the increment to the Ca^{45} diffuse activity in a microscopic region of bone per gram of calcium per unit time is

$$da = fBdt \quad (1)$$

where

f = augmentation rate ($\text{g}_{\text{Ca}}/\text{g}_{\text{Ca}}$ day)

B = blood specific activity ($\mu\text{c}/\text{g}_{\text{Ca}}$)

da = diffuse activity increment ($\mu\text{c}/\text{g}_{\text{Ca}}$)

dt = time increment (days)

Since comparable results are obtained for adult dogs of different ages, the augmentation rate f may be assumed to remain constant at least for times the order of a month, and expression (1) may be integrated and solved for f :

$$f = \frac{a}{\int_0^{T_s} Bdt} \quad (2)$$

where

a = diffuse specific activity at time T_s ($\mu\text{c}/\text{g}_{\text{Ca}}$)

T_s = time of sacrifice or sampling (days)

Time zero refers to time of injection.

Diffuse values at $T_s = 12$ hours have been found as high as $1.5\mu\text{c}/\text{g}_{\text{Ca}}$ in spite of the fact that the integral of B is then only one-third of its value at 2 weeks. The diffuse component, therefore, probably contains a considerable short-term exchangeable fraction⁽⁸⁾ which probably explains the rapid initial uptake reported.⁽¹⁾ Such a short-term exchangeable fraction, which by definition has reached equilibrium with the blood with respect to labelled ions, would contain negligible Ca^{45} after B has dropped to negligible values.

The augmentation rate corresponding to the diffuse specific activity of $1\mu\text{c}/\text{g}_{\text{Ca}}$ observed at long periods after injection is then

$$f = 0.05\%/ \text{day}$$

or approximately 20%/year. This rate, which describes the data for the adult mongrels of unknown age (Fig. 30), has been plotted as a sloping arrow in Fig. 31 at a time of 3 years.

Fig. 29. Autoradiogram and microradiograph of a cross section through the femoral midshaft of an adult dog (C or C119). Note the uniformity of the diffuse component whether or not the underlying bone has been replaced by secondary haversian systems. The large cavity in the cortex is the nutrient canal close to its junction with the marrow cavity.

Fig. 30. Diffuse specific activity in $\mu\text{c Ca}^{45}/\text{gCa}$ in the femoral or tibial shafts of 6 adult mongrel dogs sacrificed at different times after a single Ca^{45} injection. Values have been normalized to an injected activity of 0.1 mc/kg body weight and plotted as a function of time after injection. Dog ages were unknown but epiphyseal and periosteal growth were complete.

Fig. 31. Microscopic calcium density of periosteal bone containing no secondary haversian systems plotted as a function of age in 4 dogs, in the oldest of which the augmentation rate in the same region of bone has also been measured. Points show standard error bars of 5%. The slope of the plotted arrows indicates values of augmentation rate derived from the diffuse Ca^{45} activity in corresponding regions of bone. The arrow plotted at 3 years refers to the average augmentation rate for all the dogs of Fig. 30. Since calcium density does not detectably increase with time, the augmentation must be primarily long-term exchange.

Histological Location of Diffuse Activity; Use of Periosteal Bone of Known Age

The values of diffuse activity used above were obtained by scanning and averaging values from autoradiographs. Some of the values were measured over secondary haversian bone, some over millimeter-wide regions of periosteal bone containing no secondary haversian systems and no resorption cavities. A systematic study of diffuse uptake as a function of location and mineral density has not yet been made. However, the diffuse activity in adult dogs with few forming osteons is remarkably uniform whether the measurements are made for periosteal bone or secondary haversian bone. (Fig. 29). In areas filled with many comparatively young but completed osteons with mineral densities 80-90% that of highly mineralized periosteal bone, the diffuse specific activity is perhaps 50% higher than that in the periosteal bone. Occasionally a small region of non-haversian bone has been found which deposits less than one-fifth the usual diffuse activity of $1 \mu\text{C/g}_{\text{Ca}}$. These regions of low diffuse uptake have a somewhat lower mineral density than ordinary periosteal bone so that an inverse correlation between mineral density and diffuse uptake does not always hold. These regions of low diffuse uptake are believed to be rare but require investigation. A few recently-formed osteons of low mineral density may possibly take up considerably more than the average diffuse activity. This point also remains to be investigated.

In spite of these known variations in diffuse activity, a value of the augmentation rate of 10-20%/year may be taken as typical of periosteal bone containing no secondary haversian systems in an adult dog a few years of age. This bone, in contrast to secondary haversian bone, may be assigned an age and hence will be taken as the experimental material for the remainder of this paper.

Microscopic Calcium Density of Periosteal Bone as a Function of Dog Age

Measurements of the microscopic calcium density ($\text{g}_{\text{Ca}}/\text{cm}^3$) of periosteal bone by quantitative microradiography in corresponding regions of femur midshaft are shown in Fig. 31 for dogs which were 2, 5, 11 and 12 years of age. Within the 5% standard error of the measurements, there is no detectable increase in the microscopic calcium density with time.

The Diffuse Paradox

There is thus a curious paradox. A microscopic volume of bone that contains no osteons and no resorption cavities is observed to be receiving and retaining calcium atoms with no corresponding increase in

the total number of calcium atoms it contains. We conclude that there is a corresponding loss of calcium atoms from the same microscopic volume so that the total number of calcium atoms, the microscopic calcium density, does not change. But since the same number of calcium atoms are leaving as are arriving, the augmentation rate measured for periosteal bone does not represent accretion. Instead, the measured augmentation rate must be the blood-to-bone transfer rate of a two-way exchange between blood and bone. But then why, over a period of weeks or months, has there been no loss of Ca^{45} atoms along with the calcium atoms transferring from bone to blood? Is there some gross error in our measurements or assumptions?

Re-examination of Assumptions

The three most crucial assumptions we have made are the following:

Assumption (1). The diffuse Ca^{45} activity is deposited at the measured specific activity of the blood. From our previous work^(3,4) this seems a good assumption. But suppose that the intracellular fluid did not reach equilibrium with blood specific activity for a long period so that the specific activity seen by bone crystallites was less than we have assumed. In that case the time integral of B in Expression (2) would be less than we have calculated so that the value of f would be greater and the paradox aggravated, not resolved.

Assumption (2). The augmentation rate f does not vary significantly over long periods of time. Most of the measured values of diffuse activity come from mongrels of unknown age. One six-year-old dog showed a diffuse uptake even at 12 hours which was less than half that of the others (E or C122). If f were decreasing sharply, the diffuse retention of Ca^{45} might be more easily explained. We might then postulate, as Tomlin has done,⁽⁹⁾ that a comparatively rapid exchange had been occurring during the time the blood was active but that the rate of exchange was decreasing sharply so that the Ca^{45} was trapped.

Assumption (3). The periosteal bone we have measured was laid down in the first year or two of life and is not recently-deposited bone.

The second and third of these assumptions were submitted to test by the following experiment.

11-Year Double Tracer Experiment

A 12-year-old mongrel dog (QA122 or A) which had been injected with Sr^{90} late in 1945 in connection with a toxicity experiment* was injected

*M. P. Finkel, et al. ANL-5696, p. 18 (March 1957).

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with Ca^{45} in 1957; blood specific activity measurements (Ca^{45}/Ca) were made; and the animal was sacrificed 11 days after Ca^{45} injection. Cross sections through the midshaft of the femur were autoradiographed on No Screen Plates, both with and without 4-mil aluminum filters, so that the Sr^{90} diffuse component and the Ca^{45} diffuse component could be measured separately.

[In December 1945, the dog was a young adult weighing 10.7 kg; it was injected with 0.30 mc Sr^{89}/kg , together with roughly 15-30 μc Sr^{90}/kg as an impurity. The Sr^{89} (half-life 54 days) has long since decayed, leaving only the $\text{Sr}^{90}-\text{Y}^{90}$. In November 1955, the Sr^{90} burden was 5.4 μc as determined by bremsstrahlung counting. The dog was failing rapidly due to the long-term effects of his radioactive burden.]

In regions of periosteal bone over a millimeter wide which contained no secondary haversian systems, and therefore, no regions of growth more recent than the original periosteal apposition, both Sr^{90} diffuse activity and Ca^{45} diffuse activity were found. The Ca^{45} diffuse activity in this old dog averaged 0.7 $\mu\text{c}/\text{g}$ Ca (with the usual normalization to 0.1 mc/kg body injected activity) a value comparable to the Ca^{45} uptake in all the younger animals. The Ca^{45} blood specific activity fell more slowly after injection than our standard Ca^{45} blood curve⁽⁴⁾ so that the corresponding calcium augmentation rate was 0.015%/day or 5%/year, only a little lower than the augmentation rate for the younger dogs. The measured microscopic calcium density in the same region of bone is plotted in Fig. 31 at 12 years of age together with an arrow indicating the rate of augmentation measured at age 12 with Ca^{45} . Thus Assumption (2) is substantially correct in that there is not a large decrease of augmentation rate with age. The precise age dependence of f requires further study.

The presence of uniform Sr^{90} diffuse activity throughout this same region of bone indicates that periosteal growth was complete at the time of Sr^{90} injection. If periosteal growth of even 1 μ/day had been proceeding in this region in December 1945, the Sr^{90} ring of blackening would be detectable above the Sr^{90} diffuse component, and the bone outside such a ring would contain negligible Sr^{90} . Assumption (3) is therefore verified for measurements on this animal.

Conclusions

Diminution and Long-Term Exchange

Since our measurements and assumptions appear to be substantially correct, how can we resolve the paradox? We have focussed our attention on a microscopic volume of bone containing neither osteons nor

resorption cavities, and we have waited 11 or more days after Ca^{45} injection before making activity measurements. Therefore the complications of appositional growth, resorption, and short term exchange have been eliminated. By tagging every millionth calcium atom in the blood, we have counted the calcium atoms going into this microscopic volume of bone, but by shooting X-rays through it we find that the total number of calcium atoms it contains has not been increasing. We must therefore conclude that calcium atoms are leaving this microscopic volume of bone at a rate comparable to the measured augmentation rate. Since these departing atoms are different from the arriving atoms -- Ca^{45} atoms are not among them -- it is convenient to define another term: the diminution rate or rate of loss of calcium from our microscopic volume. Then our result may be stated: highly mineralized periosteal canine bone has been observed to be undergoing simultaneous augmentation and diminution of calcium at a rate the order of 10%/year ($0.1 \text{ g}_{\text{Ca}}/\text{g}_{\text{Ca}} \text{ year}$). This is an enormous rate of calcium turnover to be found in what has hitherto been considered unavailable bone mineral. It has been generally believed that only recently-formed osteons provide bone mineral available for exchange with body fluids. It now appears that augmentation-diminution in highly mineralized bone probably accounts for a larger rate of calcium turnover in adult cortical bone in dogs than accretion, resorption, or exchange associated with osteons.⁽⁴⁾ The importance of this diffuse component of skeletal metabolism has also been pointed out by Arnold.⁽¹⁾ If the rate of 10%/year were extrapolated to the whole skeleton, the total rates of augmentation and diminution would have the same order of magnitude as the total adult accretion rate for dog or man, and would turn over an amount of calcium equal to that in the blood every 18 hours (taking blood calcium content to be 1/5000 that of the skeleton); however, such an extrapolation in detail is not justified by the data at this time.

Balanced rates of augmentation and diminution represent an exchange of calcium atoms. there are equal and opposite rates of transfer of calcium atoms between blood and bone occurring at the same microscopic location. But this exchange cannot have reached equilibrium with respect to Ca^{45} atoms even over the three months of our experiments because the diminution rate is not yet labeled (within the accuracy of our present measurements). Thus, although the equal and opposite rate of transfer of calcium atoms is large, the time necessary to reach equilibrium with respect to Ca^{45} atoms must be longer than the period of our measurements. We are therefore, dealing with what may be called long-term exchange. We conclude that if the augmentation rate is maintained, then a loss of Ca^{45} activity due to continuous calcium diminution should become significant if we wait long enough.

There are several molecular mechanisms which might be responsible for long-term exchange: recrystallization in its original sense of continual dissolution and redeposition of submicroscopic crystallites;⁽²⁾

deposition and long-delayed thermal reemission of calcium atoms in the first calcium layer of a crystallite coupled perhaps with diffusion of calcium within the crystallite's lattice structure;⁽²⁾ or some barrier to calcium atoms between extracellular fluid and crystallite that has a low probability of penetration. The long-term time dependence of elimination of calcium-like radioactivity by diminution would depend not only on the diminution rate as here defined but also upon the molecular mechanism responsible. Is all the mineral in a single microscopic volume of bone involved directly in the long-term exchange, or is a several-step process involved such as diffusion into crystallite interiors? Whether or not one would expect the effect of diminution on hotspots to be quantitatively the same as its effect on diffuse activity would likewise depend on the molecular mechanism and cannot be predicted from the present data. However, in view of the magnitude and persistence in the older dogs of the measured augmentation rate, a loss of Ca^{45} by diminution from the diffuse component in periosteal canine bone should be just detectable over a period of a year.

Summary

By quantitative autoradiography and quantitative microradiography the *in vivo* diffuse uptake of Ca^{45} in highly mineralized canine bone many years old has been measured and identified primarily with long-term exchange of calcium. By long-term exchange we mean an equal rate of transfer of calcium atoms in opposite directions between blood and bone in which the mean residence time of an individual calcium atom in bone is long, here the order of a month or longer. In microscopic regions of canine cortical bone with a calcium density of $0.60 \pm .03 \text{ g}_{\text{Ca}}/\text{cm}^3$, the rate of transfer of calcium atoms between blood and bone in this exchange is $0.05 - 0.20 \text{ g}_{\text{Ca}}/\text{year}$ per gram calcium in bone. This rate of calcium transfer represents a significant fraction of the total calcium turnover in the skeleton of an adult dog and may play an important part in the calcium regulation of the blood. The possible implications for the distribution of long-term radiation dose to the skeleton due to internally-deposited Sr^{90} or Ra^{226} are being examined. The observed rate of calcium transfer suggests that loss of Ca^{45} activity from highly mineralized canine bone by a process other than resorption should be just detectable over a period of a year.

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Appendix

The following terminology is used in this paper; for clarity specific reference is made to calcium though, of course, the terms need not be so restricted. We have attempted to use familiar terms in their accepted sense.

Microscopic volume: A cube the order of 10 microns on a side which may or may not be occupied by bone mineral but in which no resorption is taking place. (This arbitrary definition is used to simplify the following definitions.)

Exchange. Any process involving equal and opposite rates of transfer of calcium atoms in a single microscopic volume of bone mineral.

Time constant of an exchange process: The number of calcium atoms available for this exchange in a single microscopic volume of bone mineral divided by the rate of transfer of calcium atoms (number per unit time); approximately equal to the time necessary for labeled calcium atoms to reach equilibrium, that is, for equal and opposite rates of transfer of labeled calcium atoms to be established.

Short-term exchange: Any exchange processes with time constants much less than a week.* Labeled calcium atoms will be in equilibrium in most tracer experiments.

Long-term exchange: Any exchange processes with time constants much greater than a week.* Labeled calcium atoms will not be in equilibrium in most tracer experiments.

Accretion: Any process involving the net transfer of calcium atoms to a single microscopic volume.

Apposition. The process of accretion involving the formation of new bone and resulting in an increase in mineral volume as observed microscopically.

Secondary mineralization: The process of accretion in existing bone mineral by which the mineral density is increased as observed microscopically.

Resorption: The process by which bone is eroded resulting in a decrease in mineral volume as observed microscopically.

* The time dependence of the concepts indicated by asterisk is obvious. However, if the times of observation are specifically referred to, these concepts are valuable for analyzing data short of a more complex analogue computer analysis. The value of such a separation of time constants rests upon the apparent absence of important exchange processes with time constants the order of magnitude of a week.

Augmentation: A gross uptake in a microscopic volume of existing bone mineral of calcium atoms which remain much longer than a week.*

Diminution: A gross loss from a microscopic volume of bone mineral of calcium atoms which have resided there much longer than a week.*

In terms of these definitions, the following quantitative relations may be expressed either for small volumes of bone or for a whole bone or skeleton:

$$\begin{aligned}\text{Accretion} &= \text{Apposition} + \text{Secondary Mineralization} \\ \text{Secondary Mineralization} &= \text{Augmentation} - \text{Diminution} \\ \text{Accretion} - \text{Resorption} &= \text{Net increase of mineral}\end{aligned}$$

In terms of our definitions above, the quantity A measured and defined mathematically by Bauer, Carlsson, and Lindquist⁽⁸⁾ is:

$$A = \text{Apposition rate} + \text{Augmentation rate}$$

or

$$A = \text{Accretion rate} + \text{Diminution rate}$$

and the exchangeable fraction E refers to short-term exchange.

Evaluation of the diminution rate for the skeleton as a whole will be a difficult task, but will probably not affect the utility of the quantities A and E as presently derived from activity measurements.⁽⁸⁾

*The time dependence of the concepts indicated by asterisk is obvious. However, if the times of observation are specifically referred to, these concepts are valuable for analyzing data short of a more complex analogue computer analysis. The value of such a separation of time constants rests upon the apparent absence of important exchange processes with time constants the order of magnitude of a week.

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RADIUM TOXICITY STUDIES

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I. Quantitative Observations on Ra²²⁶ in Bone

Autoradiographic studies have shown that the pattern of deposition of radium in bone is similar to that of calcium. Studies of both human and animal bone show radium to be taken up in areas of appositional growth in the process of mineralization of the newly formed osteoid. We define these regions of high concentration of radium as "hot spots", while the more uniform deposition which appears throughout the bone tissues is called the diffuse reaction. These terms, hot spot and diffuse reaction, as we use them, are descriptive terms referring to the appearance of an autoradiograph and are not associated with any one mechanism.

Autoradiographs of Ra²²⁶-burdened bone are easily evaluated quantitatively. For this we employ an Eastman Kodak NTA plate with a 25 μ thick emulsion. The number of alpha tracks observed in this emulsion after a suitable exposure to a bone section depends upon the following:

1. The quantity of Ra²²⁶ per unit volume of bone.
2. The fraction of the radon produced which escapes from the bone.
3. The range of the alpha particles in bone. We have demonstrated that approximately 70% of the radon produced in a bone section embedded in methyl methacrylate escapes, the same fraction that escapes from the bone in vivo.⁽¹⁾

Thus, for every radium atom which emits an alpha particle, there will be four alphas emitted by its daughters, of which only 30% will be emitted in bone. The average range of these alpha particles in bone ($\rho = 2 \text{ g/cm}^3$) is 30 μ . Hence, we write the number of radium disintegrations per unit time per unit volume as

$$\frac{4 \times \text{No. of tracks}}{2.2 \alpha \text{'s per Ra disintegration}} \times \frac{1}{\text{Time}} \times \frac{1}{\text{Area} \times 30\mu}.$$

From a volume of bone 30 μ thick one quarter of the alphas produced will reach the surface and produce a track within the emulsion. Hence above we multiply the number of tracks by four. Since there are 2.2 alpha particles emitted in bone per radium disintegration ($1 + 0.3 \times 4$), we divide the number of alphas produced by 2.2 and obtain the number of radium disintegrations per unit time per unit volume.

To obtain this activity in terms of microcuries, we simply divide by 3.7×10^{10} disintegrations/sec:

$$\mu\text{c Ra }^{226}/\text{mm}^3 = 1.9 \times 10^{-8} \frac{T}{A \times t}$$

where T = the observed number of tracks in area A.

A = the area in mm^2

t = the time of the exposure in days

The quantitative study of Ra^{226} depositions in bone yields information on the following:

1. The dose in microscopic regions of bone.
2. The relationship between the diffuse and the hot spot activities in bone. Dose distributions and the correlation with damage to bone structure will be reported at a later date.

After a single injection of radium, the specific activity of the blood drops rapidly, and the mineral formed during this period will have a constantly changing specific activity. A narrow band of bone a few microns wide will be labeled with the isotope, but this band is too narrow to allow evaluation of an average specific activity of the mineral. However, when the specific activity of the blood is elevated for a considerable period of time as in the Elgin State Hospital patients,^(2,3) or as may be assumed to have occurred in the dial painters, then the activity in regions of appositional growth covers a cross-sectional area of dimensions sufficiently large to allow the evaluation of the specific activity by means of an autoradiographic measurement of activity and a microradiographic measurement of density. In these cases we can define a hotspot to diffuse ratio with some precision. The ratio must be clearly understood to refer to the ratio of the specific activities, but not to the rates of uptake of the radioactive label in the two regions.

The diffuse uptake of radioactivity we picture as taking place as long as the specific activity of the blood is elevated. The appositional growth process, instead, labels a closing osteon with the specific activity of the blood, and thus, to a first approximation, the activity found is independent of the length of time the blood level was elevated. Of course, a region labeled as a result of appositional growth can later be diffusely labeled, but this is a second order effect.

In Table 25 are shown the quantitative values of hot spot and diffuse activities obtained to date for a number of cases. Here we have used the concept of "uniform label," which we define as the body burden divided by the bone volume. The bone volume is calculated assuming a wet density of 2 g/cm^3 , and assuming the bone weight is 10% of the body weight. This

"uniform label" would be expected to describe the distribution occurring in an animal which had always a constant level of a radioactive tracer in the blood. Under hot spots we have listed the value of the highest specific activity found for a large haversian system. Under diffuse we have placed a range of values which were obtained from counting tracks over areas ten times as large as a typical hot spot. Finally, we have calculated the ratios hotspot to diffuse activity, hot spot to "uniform label," and diffuse to "uniform label,"

Table 25

Summary of the autoradiographic observations

Case	Radium acquired	Body burden at death, μc	Body weight, kg	Uniform label, $\mu c/mm^3$	Measured activity		Calculated ratios		
					Hot spot, $\mu c/mm^3$	Diffuse, $\mu c/mm^3$	Hot spot Diffuse	Hot spot Uniform label	Diffuse Uniform label
R	Elgin patient 41 weekly inj. at age 32 yrs. Died 56 yrs.	3.6	55	1.3×10^{-6}	2.9×10^{-5}	$.67-.98 \times 10^{-6}$	30-43	22	0.5-0.8
I	Elgin patient 18 weekly inj. at age 27 yrs. Died at 53 yrs.	0.7	55 assumed	0.25×10^{-6}	1.3×10^{-5}	0.2×10^{-6}	65	52	0.8
103	Medically, I-V inj. at age ~35 yrs. Died ~64 yrs.	6.8	57.5	2.4×10^{-6}	2.6×10^{-5}	0.7×10^{-6}	37	11	0.3
118	Drank Ra water at age 48 yrs. Died at 84 yrs.	10.5	52.	4.0×10^{-6}	6.7×10^{-5}	$2.2-3.4 \times 10^{-6}$	20-30	17	0.55-0.85
302	Dial Painter Painted at age 15 for 2 mo. Died at 47 yrs.	3.8	68	1.1×10^{-6}	2.5×10^{-5}	$1.0-2.0 \times 10^{-6}$	12.5-25	23	0.9-1.8
	Dial Painter, Painted at age 21 yrs for 1 yr. Died at age 54 yrs.	2.7	55 assumed	0.98×10^{-6}	3.4×10^{-5}	$0.14-1.6 \times 10^{-6}$	21-240	35	0.14-1.6
Dog	Age ~1-2 yrs. R Single I.V. Inj. Sacrificed at 1 mo.	16	9.1	35×10^{-6}		$22-32 \times 10^{-6}$			0.6-0.9

These values were all obtained from cortical bone sections, usually from the femur, but in some instances from the tibia. Note that in these long bones the activity associated with the diffuse label is of the order of magnitude of one-half the "uniform label" defined above as shown by the last column in this table.

In Table 26 we list the activities found in various bones of Elgin patient I. In this patient the radium burden had been carried for 26 years. While no great importance can be placed on samples ranging from 100 to 300 g, it is still apparent that most of the activity remaining is in the long bones. These gross samples were evaluated by gamma-ray spectroscopy by Dr. C. E. Miller.

Table 26

Bone radium content, Elgin patient I

Bone	Sample, g	Average Activity $\mu\text{c/g}$	Activity Uniform label
Femur	285	3.5×10^{-4}	2.4
Vertebrae and rib	74	0.91×10^{-4}	0.62

II. The Loss of Ra^{226} from Human Bone

The Elgin State Hospital patients form an extremely important and valuable group for study, for they were given known quantities of Ra^{226} at weekly intervals for varying lengths of time. Thus we know the total input and can assume, without great error, the specific activity of the blood during the period over which the I.V. injections were given.

In Fig. 32 is shown a blood specific activity curve for a human given weekly injections of $10\mu\text{c}$ of Ra^{226} . This was drawn by transforming the blood curve for Sr^{85} in humans⁽⁴⁾ into a curve for Ra^{226} , the transformation used being that required to transform an observed Sr^{90} curve for beagles⁽⁵⁾ into an observed Ra^{226} curve for beagles.⁽⁶⁾ Admittedly, this is not the most satisfactory manner of obtaining a blood curve, but we feel that at least it gives the proper order of magnitude to be expected.

Fig. 32 Postulated blood specific activity in a human receiving weekly injections of $10 \mu\text{c}$ Ra^{226} . The highest value plotted for each week is the value occurring at one hour.

Also indicated in Fig. 32 is the average value of the blood specific activity over each weekly interval. Note that this curve seems to approach a constant value after several weeks. This average value is of considerable importance, for if there is no discrimination against the radium from blood to bone, then regions of appositional growth should be built at this specific activity during the period over which the injections were given.

In Table 25, given in part I of this report, are tabulated bone specific activities for Elgin cases R and I. The values for R are quite good, having been obtained from studies of many different bone sections, while those from I must be considered tentative at present as they have been made from only a very few observations. From Fig. 32 we would expect hot spots to show a specific activity of the order of $0.7 \mu\text{c Ra}^{226}/\text{g Ca}$.

Transforming this value from units of $\mu\text{c Ra}^{226}/\text{g Ca}$ to units of $\mu\text{c Ra}^{226}/\text{mm}^3$ of bone, using a microradiographically determined value of 1.3 g of hydroxyapatite per cm^3 of bone (for human bone)⁽⁷⁾ where hydroxyapatite is 40% calcium by weight, we obtain $36 \times 10^{-5} \mu\text{c}/\text{mm}^3$. This is the specific activity we would expect a forming osteon to incorporate if it were built during the 41 or 18 week interval in patients R or I. However, from Table 25, we find that this value is 12 and 27 times greater than the highest activities found in these two cases. If these systems are thought to have been formed after the injection period, then examination of the blood activity curve indicates they were formed within a few weeks after the injections stopped, and one must postulate that all original systems formed during the period of the injections have been resorbed. This consideration convinces us that these are osteons formed during the injection period, not afterward.

Thus it is apparent that either radium is not built into forming osteons at the specific activity of the blood, or that a loss of activity from these regions has occurred without resorption taking place. To describe this removal of a label without removal of the total calcium within a system we use the term diminution. We are tacitly assuming that the results we obtain have not been greatly modified by radiation damage.

The magnitude of the diffuse label in these two Elgin cases is quite informative. While the period over which the specific activity of the blood was elevated differs by a factor of two, the actual retained radium levels after 26 years differ by a factor of five, and the observed diffuse levels also differ by a factor of five. The dependence of the magnitude of the diffuse label upon the body burden is quite evident in the last column of Table 25, where the ratio of the diffuse level to the uniform label is tabulated.

Since roughly the same diffuse to uniform label relationship exists one month after injection in the dog (see Table 25) and 26 or more years later in the human, it is tempting and not unreasonable to assume that the

magnitude of the diffuse radium label in the bone has decreased with time, apparently at the same rate as the activity in the regions of appositional growth.

While we must emphasize that these results are tentative, it is apparent that they indicate resorption per se may play a relatively minor role in the long-term removal of radium from cortical bone. It is obvious that the mechanisms herein discussed are of decisive importance in the description of radiation dosimetry in bone following intake of bone-seeking radioelements. Before entering into this aspect of our investigation, however, we feel that it is necessary first to extend our observations to suitable human material in order to assess the extent to which our preliminary impressions apply.

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Th^{228} , Ra^{228} , AND Ra^{226} IN NORMAL BONE AND IN BONE SPECIMENS FROM RADIUM PATIENTS

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I. Normal Bone

Table 27 summarizes data obtained in the course of testing the di-butyl phosphate (DBP) method for measurement of Th^{228} in bone.⁽¹⁾ Groups of bone samples from the first cattle were analyzed at intervals of about 6 months. The Th^{228} content increased with time, indicating that appreciable amounts of Ra^{228} were present and that Th^{228} was growing into the sample according to the genetic relationship known to exist for these nuclides:

Since the history of intake of Ra^{228} and Th^{228} was not known for these samples, it was not possible to calculate the Ra^{228} content by application of equations of radioactive growth. However, extraction of Th^{228} by DBP leaves Ra^{228} in the aqueous solution, and a subsequent extraction of Th^{228} which has been allowed to grow in can now be used to calculate the amount of Ra^{228} present. This was done for the group of samples from which Th^{228} was extracted in July and August, 1956; a second extraction in June, 1957 provided a value of $0.57 \mu\mu\text{c Ra}^{228}/\text{g}$ of ashed bone calculated to Aug. 1, 1956. Comparison of this value with the series of Th^{228} values indicates that most of the Th^{228} was present because of growth from Ra^{228} instead of independent deposition in the bone.

Table 27

Th^{228} , Ra^{228} , and Ra^{226} in normal bone

Sample	Bone specimen	Date of analysis	No. of samples	$\mu\mu\text{c Th}^{228}/\text{g}^{(a)}$	$\mu\mu\text{c Ra}^{228}/\text{g}^{(a)}$	$\mu\mu\text{g Ra}^{226}/\text{g}^{(a)(b)}$
Cattle - 1	Tibia	July-Aug., 1956	4	$0.25 \pm 0.01^{(c)}$	0.57 ± 0.02	0.66 ± 0.03
		Nov.-Dec., 1956	3	$0.30 \pm 0.01^{(c)}$		
		Apr.-May, 1957	5	$0.36 \pm 0.01^{(c)}$		
Cattle - 2	Tibia	Oct., 1957	4	$0.11 \pm 0.01^{(c)}$		0.22 ± 0.02
Normal Human - 1	Cortical Tibia	Dec. 4, 1956	1	0.016 ± 0.004		$0.07^{+0.01}_{-0.06}$
Normal Human - 2	Cortical Tibia or Femur	Apr. 3, 1957	1	0.007 ± 0.004		0.016 ± 0.006

(a) Radioactivity per g of bone ashed at 620°C

(b) Analyses for Ra^{226} were performed by radon emanation techniques (Reference 9). The large uncertainty indicated for low values of Ra^{226} was caused by an uncertainty of $0.02 \mu\mu\text{g}$ in the blank correction.

(c) The mean deviation from the mean value for replicate samples was well within the estimated error of 5 to 10% for a single measurement.

The concentration of Ra^{226} in these samples was only slightly greater than that of Ra^{228} . It is likely that only 3 or 4 years elapsed from birth of the cattle to time of analysis, so decay of 6.7-yr Ra^{228} should not greatly distort the implied intake of approximately equal amounts of these two isotopes by the animal from its environment. The uniformity of values for different samples implies that intake of radium was continuous and, therefore, from natural sources, but the concentrations of radioactivity are considerably higher than those found for normal human bone (see below). An average value of $0.03 \mu\text{g Ra}^{226}/\text{g}$ ashed bone would be expected from the natural body burden of about $10^{-10} \text{ g Ra}^{226}$ for humans.

Another sample of bone was obtained from Wisconsin cattle whose history was known. Ra^{228} values have not yet been obtained, but comparison of the first measurements of Th^{228} with Ra^{226} values indicates again that approximately equal amounts of the isotopes of radium are present (Table 27). On the other hand, the concentrations of radioactivity are only one-third of those found for the first cattle although much higher than those for humans. It appears that a ruminant diet may be considerably higher in radium than that of humans. The first of the cattle may have come from a region having an unusually high Ra to Ca ratio in the environment; the sample was purchased locally but the origin of the animal is unknown.

The bone samples from the two normal humans listed in Table 27 were analyzed primarily to provide an estimate of Th^{228} normally present in human bone for comparison with figures from radium patients. The results given are tentative because of the low concentrations and the small amount of sample available. However, no detectable radioactivity ($<0.005 \mu\mu\text{c Th}^{228}$) was found in reagent blanks carried through the analysis. Because the samples represent accumulation of radioactivity over several decades, one may assume that the Ra^{228} is in equilibrium with Th^{228} . Thus it appears from these analyses that Ra^{228} may be present in amounts equal to or greater than Ra^{226} if one includes corrections for decay of Ra^{228} .

One conclusion which may be drawn from the data of Table 27 is that estimates of the natural radioactivity of humans must include Ra^{228} which has been largely neglected in the past. One reason for this neglect is that very little is known about the rate of intake of Ra^{228} . However, Ra^{228} in amounts comparable to or greater than those of Ra^{226} has been reported for numerous radioactive hot spring waters.⁽²⁾ In addition, preliminary work on the water supplies of certain communities near Chicago has shown that Ra^{228} is present in concentrations approximately equal to those of Ra^{226} ($3\text{--}8 \mu\text{g Ra}^{226}/\text{l}$). Previously, water from these sources was found to contain Ra^{224} and Ra^{226} in equal amounts, but no Th^{228} was found.⁽³⁾

II. Bone Specimens from Radium Patients

It is now widely recognized that, in addition to Ra^{226} , sufficient Ra^{228} may have been present in the radium preparations taken by some patients to contribute appreciably to the radiation dosage to the bone.⁽⁴⁾ Thus, measurement of the Ra^{228} content of specimens of bone from radium patients is of importance in determining whether correlation of bone damage with the Ra^{226} burden alone is valid. Since no means of directly measuring Ra^{228} in bone with sufficient sensitivity is available as yet, the Th^{228} content is being used as a measure of Ra^{228} in the present work. Assuming radioactive equilibrium to exist, the amount of Ra^{228} at the time of radium intake can then be calculated and compared to the Ra^{226} content of the same sample of bone.

Table 28 summarizes data on bone specimens from six radium patients. The code numbers in Column 1 are those used in the report by Looney, et al.,⁽⁵⁾ which describes the case histories of these radium patients. Under the heading of Th^{228} , values are given for the amount of Th^{228} per g of bone ashed at 620°C as of the date given. In the last column, the ratio of Ra^{228} to Ra^{226} at the time of intake is expressed in per cent values for each bone specimen. Ra^{228} values were obtained by extrapolating to the approximate time of dosage, using calculations of radioactive growth of Th^{228} and the assumption that direct deposition of Th^{228} need not be considered. These figures were not corrected for natural amounts of Ra^{228} , deposited during the entire life of the subject, so the extrapolation may provide values that are much too high for those cases in which the concentration of Th^{228} was very small. It should be noted that considerable variation in the concentration of Ra^{226} was found for bone specimens from each patient (as previously reported by Norris and co-workers⁽⁶⁾ for Case No. 112) and that the concentration of Ra^{223} varied similarly so as to provide ratios for each patient which were constant within reasonable limits of error.

The amount of Th^{228} present (Col. 8, under Th^{228}) was sufficiently high in only three cases to be considered positive identification of intake of Ra^{228} with the Ra^{226} dose. The highest ratio of Ra^{228} to Ra^{226} was 0.02 at the time of intake for a patient (designated by * in Table 28 because no literature reference for this patient is available as yet) who painted radium dials in the 1920's. Patient 112, who had the next highest ratio, took radium orally for therapeutic purposes in 1928. A bone specimen from this patient was reported by R. D. Evans to contain a Th^{228} to Ra^{226} ratio of 0.0024 ± 0.0005 several years ago;⁽⁷⁾ allowing for radioactive decay, this value is in reasonable agreement with our Th^{228} to Ra^{226} ratio of 0.001 in 1957. The calculated ratio of Ra^{228} to Ra^{226} at the time of ingestion is 0.01. The third patient, number 313, was a dial painter who seems to have ingested very little Ra^{228} .

Table 28
 Th^{228} , Ra^{228} , and Ra^{226} in specimens of bone from radium patients

Patient	Body burden, $\mu\text{g Ra}^{226}$	Intake route ^a	Approx. yr. of intake, t_1	Bone specimen	Weight of ashed bone, g	Th^{228}		$\mu\mu\text{g Ra}^{226}/\text{g}^b$	% Ra^{228} at t_1 relative to Ra^{226}
						Date	$\mu\mu\text{c/g}$		
112	1.3	Oral-Med.	1928	Humerus	2.99	7-12-56	0.16 ± 0.01	180	1.2 ± 0.1
				Humerus	1.88	8-15-56	0.12 ± 0.01	160	1.0 ± 0.1
				Humerus	3.13	7-10-57	0.53 ± 0.03	550	1.4 ± 0.1
313	1.3	Oral-Dial	1924	Femur	1.36	4-26-56	0.02 ± 0.01	520	0.08 ± 0.04
				Femur	1.36	5- 4-56	0.06 ± 0.01	730	0.15 ± 0.04
				Femur	1.44	10-26-56	0.02 ± 0.01	280	0.14 ± 0.04
				Vertebra	1.22	7- 9-57	0.07 ± 0.01	1420	0.11 ± 0.02
*	1.3(?)	Oral-Dial	1924	Femur	2.42	7- 9-57	0.54 ± 0.03	560	2.1 ± 0.1
R	3.1	Intravenous	1931	Tibia	1.67	5- 7-56	< 0.006	290	< 0.02
				Tibia	2.1	5- 7-56	< 0.005	190	< 0.03
				Tibia	4.70	10-12-56	0.005 ± 0.002	430	(0.01)
				Femur	5.95	7-10-57	0.013 ± 0.002	1220	(0.01)
				Femur	4.94	7-10-57	0.004 ± 0.002	330	(0.01)
103	5.3	Intravenous	1925	Femur	4.68	10-12-56	0.021 ± 0.004	1240	0.03 ± 0.01
				Femur	5.93	7-10-57	0.020 ± 0.005	1090	0.04 ± 0.01
118	2 to 10	Oral-Med.	1918	Vertebra	1.04	4-26-56	< 0.01	3220	< 0.013
				Vertebra	1.36	5- 4-56	0.04 ± 0.01	5430	0.03 ± 0.01

^a"Oral-Med." denotes radium water taken as a medicine; "Oral-Dial" refers to radium dial painter.

^bErrors in measurement of Ra^{226} are estimated to be less than 5 per cent.

* No literature reference available for this patient.

Patient R is one of the series of patients at Elgin State Hospital who received intravenous injections of radium. Solutions of radium believed to be of the same composition as given to those patients have been reported to contain no more than 0.05 per cent Ra^{228} relative to Ra^{226} , corresponding to a maximum of 0.66 per cent at time of injection.⁽⁸⁾ As seen in Table 28, our values for Th^{228} in bone specimen from Patient R do not exceed the values found in "normal" bone, and an extrapolation to time of injection yields a Ra^{228} to Ra^{226} ratio of only 10^{-4} , even without attempting to correct for the natural Th^{228} content of human bone.

Patients 103 and 118 received radium as a therapeutic measure, Number 103 by intravenous injection and the other orally. In both cases, the Ra^{228} content seems to have been negligible.

It is evident that the total radiation dosage from Ra^{228} was minor compared to that of Ra^{226} for each case listed in Table 28. However, the presence of Ra^{228} may be relatively more hazardous if its 1.9-yr Th^{228} descendant migrates to a different portion of the bone--such as the organic matrix.

Specimens from patients 115, 313, R and 118 were extracted with hot ethylenediamine (EDA) as a means of separating the organic constituents. No detectable Th^{228} was found in the organic portion. However, these results are inconclusive in that the absolute amount of Th^{228} in the whole bone was small and several years had elapsed since the bone material had died.

On the other hand, positive results might be expected for samples from the first cattle listed in Table 27 from which a large supply of fresh bone was available. The comparison of inorganic and organic portions given in Table 29 shows that less than 5 per cent of the Th^{228} in each sample was recovered in the organic fraction. Probably an even smaller percentage was actually extracted by the EDA, because contamination with inorganic matter is indicated by the large weight of residue in the organic fraction for the three samples in which Th^{228} was found. Such contamination was also demonstrated by the presence of Ra^{226} in the organic residues.

The results in Table 29 do not fully resolve the question as to migration of Th^{228} in bone because there was no assurance that all the organic matter was extracted by EDA. In particular, the tissue closest to the bone crystals may have been inaccessible to the EDA. That the amount of organic material left behind may be small is shown by the fact that bone transplants are made successfully after removal of proteins with EDA.

We wish to thank Dr. Robert J. Hasterlik of the Argonne Cancer Research Hospital and Drs. Asher Finkel and William P. Norris for bone specimens from radium patients. We are grateful to Commander G. W. Hyatt of the National Naval Medical Center, Bethesda, Maryland, for providing a sample of normal bone from the Tissue Bank Department, and to Captain F. Losee, also of Bethesda, for valuable advice on treatment of bone with ethylenediamine.

Table 29

Comparison of Th^{228} in inorganic and organic portions
of bone from Cattle-1

Sample	Fraction ^a	Weight of ashed residue, ^b g	Th^{228}	
			Date	Total Amount, $\mu\mu\text{c}$
1	Inorganic	4.13	8-15-56	1.02 ± 0.06
1	Organic	0.055	8-17-56	< 0.01
2	Inorganic	4.73	11-23-56	1.54 ± 0.09
2	Organic	0.38	11-27-56	0.03 ± 0.02
3	Inorganic	4.46	11-23-56	1.33 ± 0.08
3	Organic	0.56	11-27-56	0.03 ± 0.01
4	Inorganic	5.26	5-23-57	1.92 ± 0.12
4	Organic	0.31	5-23-57	0.10 ± 0.02

^aAnalyses were also made on the filter paper thimble which contained the bone sample during extraction with EDA; no Th^{228} was found.

^bWeights of residues from the organic fraction include material leached from the glassware as well as portions of the inorganic fraction of bone carried over mechanically.

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IDENTIFICATION OF I^{138} AS A DELAYED NEUTRON PRECURSOR*

G. J. Perlow** and A. F. Stehney

Following previous work⁽¹⁾ in which 15.5-sec Br^{88} was found to be a delayed neutron precursor, a search was made for other halogen precursors which may have escaped detection in experiments without chemical separation. Sources were prepared from solutions of fissionable material irradiated in the thermal column of CP-5. A gas flow system was used in which suitable chemistry separated iodine or bromine fission products, which were subsequently absorbed by bubbling the gas through CCl_4 . After the gas flow was stopped, the decay of the neutron activity in the CCl_4 was observed by means of a BF_3 long counter. The experimental technique differed from that used previously by us chiefly in having smaller solution volumes and less decay of activity in transit between the reactor and the counter. The iodine sample obtained showed the expected I^{137} activity for which a half-life of 24 seconds was obtained. In addition a prominent 6-second group was observed. This is attributed to I^{138} , an odd-odd precursor analogous to Br^{88} . Among the bromine fission products, a neutron activity of 4-second half-life was found as expected by previous work.⁽²⁾

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POSITIVE ION EMISSION PROGRESS REPORT: EFFECT OF VARIOUS GASES

H. A. Schultz

The halogen-sensitive positive ion emitter is a promising device for the study of gaseous diffusion in the atmosphere.⁽¹⁾ For maximum usefulness it must be insensitive to changes in concentrations of the normal constituents of the atmosphere. To achieve this and to learn more about the mechanism of operation, a study of the effects of various gases, singly and in combination, has been undertaken.

The present equipment has been described in previous reports.⁽²⁻⁵⁾ The electrodes include a heated filament, a separately heated boat as source of ionizable material, ion collectors and guard rings.^(2,3) With conditions (temperature and collecting potential) which gave stable operation in air, a trace of chlorine added to the air used in filling the vessel gave an increase in ion current, as expected. The increase was transient, however, as if the chlorine were being trapped out of the reaction.⁽⁴⁾ To study effectively this important reaction, steps were taken to eliminate other types of transient phenomena. To reduce thermal transients, the design of the boat and of the temperature control equipment was improved.^(4,5)

To study the effects, either transient or continual, of the normal constituents of the atmosphere, an inert gas, argon, was chosen as control, and other gases were added, or substituted. So far, most of the surveys of this type have been made under one set of operating conditions (temperatures and collecting potential) chosen to avoid time-dependent operation in air.⁽³⁾

A difference in ion current depending upon the gas enclosed in the reaction vessel may be due partly to the reactive properties of the gas (affinity for other gases, for electrons, for solids) and partly to the transport properties of the gas (viscosity, thermal conductivity, coefficient of interdiffusion with the ionizable material in the gas).

The transport effect may be large under certain conditions, such as in the comparison of helium with argon discussed in the most recent report.⁽⁵⁾ However, this is an extreme case. The transport effect may be of minor importance in the case of additives in small concentration, particularly if the additives do not differ greatly from the background gas in molecular weight and external molecular force field.

Some changes of gas produce ion current changes which are essentially immediate, continual, and reversible. This is the case for the inert gases argon, helium, and mixtures of the two, and also for nitrogen.

In other cases, there is a large immediate effect, followed by a decay process as in the case of chlorine in air⁽⁴⁾ or water vapor that has been allowed to remain in the vessel while the unit is not in operation.⁽⁶⁾

In still other cases, such as a few per cent of oxygen, carbon dioxide, or water vapor added to argon, the immediate effect is not very large, but there are delayed effects, even after the trace-containing gas has been replaced with argon several times. In overnight operation with argon, there is sometimes a maximum in the ion current about four to six hours after the last filling of the vessel. There has been some evidence that this is a delayed effect of oxygen, but it cannot always be produced at will; apparently something else is involved. To reduce the possibility of unknown components entering the vessel, steps have been taken to improve both the argon purification train and the exhaust system.

Butane, as an example of possible hydrocarbon impurities from the exhaust system or from the atmosphere, was added to argon in a concentration of about 70 ppm. The changes in gross ion current were not particularly large, but the entire operation of the instrument was gradually changed. The filament required increasing amounts of power to maintain the same resistance. The background ion current (boat turned off) became abnormally high, and when the boat was turned on or off, the ion current change took place very slowly. The effects of butane were removed, gradually, by continually flushing the vessel with argon, while operating the unit. In order to examine this hydrocarbon effect a little further, the effect of filament temperature and of collecting potential were studied first with argon, then after 24 hours' operation, with argon plus 70 ppm butane. Large differences were found in both effects. The filament burned out near one end before the study was completed. Examination of the filament indicated that most of the central portion was covered with a nonuniform, dark deposit which was insoluble in HCl, therefore not PtO. A portion of the dark material was heated in a crucible in air with a torch, and the material disappeared readily, like carbon. The balance was retained for further analysis.

The tests with butane indicated clearly that the effects of added gases should be studied ultimately under several sets of conditions of filament temperature, boat temperature and collecting potential. After a new filament was installed, a new survey of the effect of these variables upon the ion current in argon was started. Each set of conditions has been maintained for a much longer time than usual in order to observe any very slow transients. It has been discovered that the boat passes slowly through some sort of transition at temperatures slightly higher than used in most of the surveys of gas effects. The effect of filament temperature shows a lack of complete saturation as noted previously for argon.⁽⁶⁾ The effect of collecting potential follows the usual pattern.⁽³⁾

Incomplete data suggest that, if adequate time is allowed to reach equilibrium, the boat operates by one mechanism below about 400 °C, by another above about 600 °C, and by a combination of both mechanisms inside this interval.

Mr. R. G. Hussey* last summer set up equipment for the study by an independent method of the release of alkali from a boat of the type now being used. The amount of alkali released was determined by the flame photometer technique. The method is promising, but the data obtained involved boat temperatures only in the region of 600 °C and up. The presence of a transition between 400 and 600 °C had not been suspected at that time.

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THE NATURAL Ra^{226} CONTENT OF ILLINOIS MUNICIPAL WATERS

H. F. Lucas, Jr. and F. H. Ilcewicz

In previous work, water from deep wells near Chicago was found to contain unusually high (up to $8 \mu\text{g/liter}$) concentrations of Ra^{226} . This is in contrast with a value of $0.03 \mu\text{g/l}$ reported for tap water in Chicago which is obtained from Lake Michigan.^(1,2) This work has been extended to much of the State of Illinois with several related questions in mind: (1) Are similarly large populations drinking water of even higher natural radium content? (2) From what geological formations is water of high radium concentration being obtained? (3) Is there a correlation between the radium concentration of potable water and the incidence of bone cancer in the populations drinking it? Regarding the first two questions, it may be said that concentrations as high as $25 \mu\text{g Ra}^{226}/\text{liter}$ have been found in well water obtained from the St. Peters sandstone formation and that this formation appears to be the source from which the unusually high concentrations of radium are derived. Answering the third question must await the completion of statistical studies on the geographical incidence of bone tumors now being carried out by Dr. Harry Auerbach of the Division of Biological and Medical Research.

Sampling Program

Samples were collected from one-half of the 102 counties of the state, 28 selected at random by drawing of names and 23 specifically selected to obtain data pertinent to the tumor survey. The location of these counties is shown in Fig. 33.

Within each of these 51 counties, at least one sample was obtained from a surface supply and, where available, samples were obtained from ground water supplies penetrating each of four general geological classifications. The geological description of each well was obtained from Bulletin 40 of the Illinois State Water Survey Division and from the records of the Illinois Water Division. All water samples were classified in accordance with Table 30 as surface, unconsolidated, shallow sandstone, limestone, or deep sandstone. For purposes of sample collection, the deep sandstone samples were not further classified. The formations listed in this table are found at various depths and thicknesses, and some are not found in all parts of the state. Furthermore, the quantity and quality of the water from any single formation varies from area to area. In general then, not all classifications of water are used for municipal supplies in a given county.

The water samples were further classified as raw (R), treated (T) or unknown as to treatment (RT). Raw water samples were collected directly from the well head. Those classified as treated had been passed through a cation exchanger or had been softened by lime or lime-alum

Fig. 33. Municipal water supplies

Table 30

Well classification guide

Geological description	General description	Group name
Lake, river or impounding reservoir	Surface water	Surface
Quaternary system Pleistocene formation	Unconsolidated sand and gravel	Unconsolidated
Pennsylvanian system	Shale, coal, shallow sandstone	Shallow sandstone
Mississippian system Devonian-Mississippian system Devonian system Silurian system	Limestone Dolomite, limestone Limestone Dolomite	Limestone
Ordovician system Maquoketa formation Galena-Platteville Glenwood St. Peters sandstone Shakopee Oneota Cambrian system Trempealeau Franconia Galesville Eau Claire Mt. Simon Pre-Cambrian Fond du Lac	Dolomite, shale Dolomite Sandstone, dolomite, shale Deep sandstone Dolomite Dolomite Dolomite Dolomite Sandstone Dolomite Sandstone Sandstone	Deep sandstone

flocculation techniques. The addition of chlorine, hypochlorite, or phosphate was not considered treating of the water in this classification.

Procedures for Collection and Analysis

For this present survey 200 water samples, each two gallons in volume, were collected. Additional one-quart-samples, obtained from 59 of the 159 sites, were sent to the Illinois State Water Survey Division for partial chemical analysis. In Tables 31 to 33, which list the concentration of Ra^{226} in the various water samples, comments taken from the reports furnished by the Illinois Water Division are included; in particular, those samples typical of water from each well were identified.

Table 31
Concentration of Ra^{226} in surface water

Location Fig. 33	Municipality	Ra conc. (a) $\mu\mu\text{g}/1$	Location Fig. 33	Municipality	Ra conc. (a) $\mu\mu\text{g}/1$
G-6	Bloomington	0.12	G-3	Macomb	0.16
L-6	Breese	0.18	I-8	Mattoon	0.12
F-4	Canton	0.11	D-3	Moline	0.08
N-6	Carbondale	0.06	L-7	Mt. Vernon	0.08
J-5	Carlinville	0.05	L-6	Nashville	0.09
I-9	Charleston	0.11	K-9	Newton	0.10
G-10	Danville	0.14	K-9	Olney	0.11
H-7	Decatur	0.12	J-7	Pana	0.06
K-4	East St. Louis	0.06	F-8	Pontiac	0.15
J-8	Effingham	0.07	H-6	Springfield	0.03
O-8	Golconda	0.15	E-7	Streator	0.12
J-3	Grafton	0.21	H-4	Virginia	0.10
I-4	Jacksonville	0.21	M-7	West Frankfort	0.06
E-9	Kankakee	0.10	I-3	White Hall	0.12

(a) Estimated error is 0.02 or 0.03 $\mu\mu\text{g}/1$ in each case.

Table 32
Concentration of Ra^{226} in unconsolidated well water

Location Fig. 32	Municipality	Well No.	Sample ^(a) Type	Ra conc., ^(b) $\mu\text{mCi/l}$	Comments ^(c)
H-4	Ashland		RT	0.03	
H-5	Athens	1	R	0.47	Typical
H-6	Athens		T	0.18	
H-3	Peardstown	New	R	0.13	Typical
F-8	Bement	1	R	0.52	
I-4	Carrollton	Spring	R	0.06	Typical
H-6,9	Champaign-Urbana	41, 50, 49, 43, 42, 41	R	0.46	
H-8,9	Champaign-Urbana		T	0.55	
I-7	Chinoia	2	R	1.60	Looks like deep sandstone water leaking into formation
C-7	Compton	West	R	0.41	
I-8	Dwight	4	R	0.52	Typical
F-7	El Paso		RT	0.11	
F-6	Eureka	1	R	1.22	Typical
F-7	Eureka		T	0.01	
I-4	Fairbury	1	R	0.28	Typical
I-4	Fairbury		T	0.06	
P-6	Freeport	5	R	0.65	
D-4	Geneseo		R	0.25	
I-6	Germantown	2	R	0.19	Typical, iron low
I-9	Gilman	12 ^a	R	0.38	
I-9	Gilman	12 ^a	T	0.07	
J-4	Greenfield	4	R	0.35	
J-9	Greenup	1	R	0.11	
H-7	Greenview	1-50	R	0.01	Typical
G-10	Hopkinstown	4	R	0.26	Typical
I-10	Hutsonville		RT	0.09	
J-4	Jerseyville	Spring	R	0.10	Typical
F-2	Keithsburg		RT	0.28	
D-7	La Salle		R	0.55	
K-10	Lawrenceville	6	R	0.16	
K-10	Lawrenceville		RT	0.1	
K-10	Lawrenceville		I	0.16	
K-5	Leland	3	R	0.29	Typical
K-5	Leland	3, 4	T	0.04	
G-4	Lewistown	6, 7, 8	R	0.34	Typical
H-6	Lincoln	1-6	R	0.13	
G-7	Mc Lean	1	R	0.9	Typical
I-7	Macon	2	R	1.12	
I-7	Macon	2	I	0.27	
I-6	Metamora	1, 2, 3, 4	R	0.43	Typical, iron low
H-8	Monticello	1, 2	R	0.32	
H-8	Monticello	1, 2	T	0.19	Typical
H-6	Mt. Pulaski	2	R	0.07	Typical
G-7	Normal	3	R	0.58	Typical
G-7	Normal	4, 7, 8	I	0.10	
I-6	Okawville	1	R	0.37	
I-6	Okawville	1	T	0.15	Typical
J-10	Palestine		RT	0.18	
J-10	Palestine		RT	0.1	
I-5	Peoria	7	R	0.47	Typical
H-5	Petersburg	3	R	0.24	Typical
H-5	Petersburg	3	T	0.11	
G-9	Rantoul	H2	R	0.17	Typical
G-9	Rantoul	H2	T	0.02	
B-6	Rockford	7A	R	0.09	
A-7	Rockton	3	R	0.17	
G-10	Rossville		R	0.24	Typical
H-3	Rushville	1	R	0.28	Typical
I-7	Shelbyville	1, 2, 3	R	0.20	Typical
F-6	Spartanburg		RT	0.09	
C-7	Steward	1	R	0.23	
D-5	Tampico		R	0.14	
D-5	Tampico		I	0.11	
C-4	Thomson	3	R	0.14	
C-4	Thomson		I	0.21	
F-9	Watseka	New	R	0.35	Typical
I-8	West Dundee		R	0.11	

(a) Estimated error is 5 per cent or 0.02 $\mu\text{mCi/l}$, whichever is larger.

(b) Raw (R), type uncertain (RT) may be either raw or treated.

(c) Comments of Illinois State Water Survey about samples submitted for chemical analysis.

Table 33

Concentration of Ra^{226} in shallow sandstone well waters

Location Fig. 33	Municipality	Well No.	Sample(a) type	Ra conc.,(b) $\mu\mu\text{g}/\text{l.}$	Comments(c)
M-6	Ava	1	R	1.65	Typical
M-6	Ava	1,2	T	0.42	
M-6	Campbell Hill	1	R	1.84	
H-10	Catlin	--	RT	0.25	Typical
N-6	Cobden	1	R	0.22	Typical
L-4	Millstadt	--	RT	3.72	Typical
J-8	Neoga	2	R	0.35	Typical, mixed shallow sandstone and unconsolidated
J-8	Neoga	--	T	0.17	
L-5	New Baden	--	RT	0.09	
K-9	Noble	--	RT	2.05	
F-8	Saunemin	1	R	0.32	
F-8	Saunemin	--	RT	0.42	Typical, iron low
K-10	St. Francisville	--	RT	0.88	
L-5	Trenton	6	R	1.08	Typical

(a) Raw (R) treated (T) type uncertain (RT) may be either raw or treated.

(b) Estimated error 5 per cent or 0.02, whichever is larger.

(c) Comments of Illinois State Water Survey about samples submitted for chemical analysis.

The water samples were analyzed for their Ra^{226} content by the radon emanation technique. For each sample, 350 ml of water were placed in a flask and flushed with radon free air. The flask was sealed and allowed to stand at least 10 days. Then the radon which had grown in was transferred to an alpha scintillation radon counter.⁽³⁾ Duplicate radon analyses were made of all samples having concentrations greater than 2 $\mu\mu\text{g Ra}^{226}/\text{l.}$ and of all non-sandstone samples having greater than 0.5 $\mu\mu\text{g Ra}^{226}/\text{l.}$ Blank values of 0.02 $\mu\mu\text{g Ra}^{226}$ were obtained with 350-ml portions of distilled water, implying that a correction of 0.06 $\mu\mu\text{g Ra}^{226}$ per liter should be applied to the water sample values. However, even smaller values were obtained for some samples collected for this survey. Thus, some contamination in the blank samples is suspected and no blank correction has been applied to the values listed in the tables below. If it were necessary to obtain more accurate values for the low level samples, larger volumes of sample would be used.

Surface Water

Surface waters are obtained from rivers, lakes, or impounding reservoirs. In general the water is filtered, chlorinated, softened and rechlorinated. Samples of such treated water were analyzed from 28 municipalities and, as shown in Table 31, the concentration of radium is uniformly low. The average for all samples is $0.10 \mu\mu\text{g Ra}^{226}$ per liter and the maximum value was $0.21 \mu\mu\text{g/l}$. Since these values are not corrected for a blank of possibly $0.06 \mu\mu\text{g/l}$, it is apparent that very little radium is to be found in this source.

Unconsolidated Well Water

Unconsolidated well waters are obtained from sand and gravel deposits of glacial origin. These deposits are usually shallow in depth and only limited amounts of water are available. This source supplies water for the majority of the farms. In addition, most of the small municipalities in the state obtain water from this source, except in sixteen counties which have only surface water supplies. In the central third of the state, from about Bloomington (G-6) on the north to Shelbyville (I-7) on the south, larger supplies are available and constitute the primary source for all except the largest cities. Large supplies have been developed at Peoria, Rockford and Beloit.

As shown in Table 32, the concentration of radium in water from unconsolidated wells is low and remarkably consistent. There are five exceptions: Chenoa, Eureka, Freeport, Macon, and McLean. Partial chemical analysis of unconsolidated well water at Chenoa indicates that deep sandstone water was leaking into the unconsolidated deposits. A concentration of $7.5 \mu\mu\text{g Ra}^{226}$ per liter was found in the deep sandstone well nearby. Thus, leakage from other formations probably provides the unusually high concentration of radium in shallow wells for all five locations.

Shallow Sandstone Well Water

The limits of the area in which wells penetrating this formation may be found have not been determined. In general, however, this formation is found in the southern part of the state. The concentration of radium in water from the wells at Catlin and Saunemin in the northern half of the state is low and does not differ appreciably from that found in unconsolidated or limestone well water, while higher radium concentrations are usually found for shallow sandstone wells in the southern third of the state.

The concentration of radium in water from shallow sandstone wells is summarized in Table 33. The average Ra^{226} content of all except Catlin and Saunemin is $1.3 \mu\mu\text{g/l}$. A maximum value of $3.7 \mu\mu\text{g/l}$ was found in water from Millstadt.

Limestone Well Water

The water from limestone wells varies in quality more than any other type. In an area near Lake Michigan, water of low hardness and residue is obtained, whereas, at Peoria water from such wells is not potable and may have 8 to 10 grams of salt residue per liter. In general, limestone well water is more heavily mineralized than waters obtained from unconsolidated wells.

The concentration of radium in waters from limestone wells is summarized in Table 34. It is believed that water from the limestone formation is rather low in radium content, but high values are obtained in some cases because of infiltration of high radioactivity water from the underlying St. Peters sandstone formation, either by artesian flow through crevice systems or through nearby deep wells. This latter effect probably causes the high value noted for Toulon since a deep well located 60 feet from the limestone well has no casing to prevent entrance of deep sandstone water into the limestone formation. Similarly, the limestone wells at Coal Valley, New Windsor, Orion, Clifton, and Hampshire probably obtain water from the underlying deep sandstone formation. The high value for Mounds is suspected to result from infiltration of water from the shallow sandstone formation. The average concentration of radium in all raw water samples excluding the above is 0.43 $\mu\mu$ g Ra²²⁶ per liter.

Deep Sandstone Well Waters

Deep sandstone wells are found in the northern third of the State and are an important source of municipal and industrial water. The major water-producing deep sandstones, namely the St. Peters, Galesville, and Mt. Simon, underlie practically the entire area of the state but vary greatly from place to place in thickness, depth and water yield. These sandstone formations outcrop in southern and central Wisconsin where snow and rain water is absorbed. This water then moves at about 100 feet per year down to Illinois.

Deep sandstone wells are drilled through rock and metal casings are generally used only to exclude sand or undesirable water. Thus many of the deep sandstone wells are constructed so that water is obtained from several formations; therefore, it is not surprising to find large variations in the concentration of radium in this group. As summarized in Table 35, concentrations as high as 25 $\mu\mu$ g/l. and as low as 0.1 $\mu\mu$ g/l. have been observed.

The highest concentration of radium was found in well waters obtained only from the St. Peters sandstone formation. These well waters at Cuba and Ipava had 23.2 and 25.4 $\mu\mu$ g Ra²²⁶/l respectively. The highest concentration of radium in the water from a well which penetrates the Galesville formation was 8.5 $\mu\mu$ g/l. at Elgin. This well obtains water from both the St. Peters and Galesville formations. The highest concentration of radium in the water from a well which penetrates the Mt. Simon formation was 4.8 $\mu\mu$ g/l. at Byron.

Table 34
Concentration of Ra²²⁶ in limestone well water

Location Fig. 33	Municipality	Well No.	Sample ^(a) type	Ra conc. ^(b) $\mu\mu$ g/l.	Comments ^(c)
O-6	Anna	1A	R	0.36	Typical
O-6	Anna		T	0.11	
C-9	Argonne		RT	0.08	
F-9	Ashkum	1	RT	0.40	Typical, iron removed
E-9	Bourbonnais	1	R	0.41	
E-9	Clifton	1	T	1.07	Typical, iron low
D-3	Coal Valley	1	R	1.84	
O-6	Dongola	1	R	0.13	Typical
B-8	Hampshire	2	RT	0.89	Drilled to St. Peters then backfilled.
E-6	Hennepin		RT	0.19	
O-6	Jonesboro	1	R	0.15	Typical
O-6	Jonesboro	1,2,3	T	0.19	
E-9	Manteno		RT	0.48	
F-6	Mound City	1	R	0.24	Typical, iron low
P-6	Mounds	North	R	2.20	Typical, iron high
P-6	Mounds	North	T	0.70	
E-4	New Windsor	2	R	1.21	
D-4	Orion	1	R	1.49	
D-10	Park Forest		R	0.73	
D-9	Peotone	1	R	0.57	
D-9	Plainfield	3	R	0.79	Typical, unconsolidated
D-5	Prophetstown		RT	0.59	
A-7	Rockton	1	R	0.34	
I-4	Roodhouse	1	R	0.16	
E-5	Toulon	2	RT	5.91	

(a) Raw (R), treated (T) tap (RT) type uncertain, may be either raw or treated.

(b) Estimated errors are 5 per cent or 0.02 $\mu\mu$ g/l, whichever is larger.

(c) Comments of Illinois State Water Survey about samples submitted for chemical analysis.

Table 35
Concentration of Ra²²⁶ in deep sandstone well water

Location Fig. 33	Municipality	Well No.	Sample (a) type	Ra conc., (b) μg/l.	Comments (c)
E-3	Aledo	3	R	3.59	St. Peters
C-6	Amboy	2	R	2.12	Galesville
C-6	Amboy		T	0.16	
B-7	Belvidere	4	R	0.64	Mt. Simon
B-6	Byron	2	R	4.18	Galesville
B-6	Byron	1	R	4.80	Mt. Simon
A-7	Capron	1	R	0.30	St. Peters
A-7	Capron	1	T	0.11	St. Peters
D-4	Cambridge	1	R	1.78	St. Peters
F-8	Chatsworth	East	R	0.69	St. Peters
F-8	Chatsworth		T	0.50	
F-7	Chenoa	1	R	7.50	St. Peters
F-7	Chenoa	1	T	4.26	St. Peters
G-4	Cuba	1	R	23.2	St. Peters only typical, iron high
G-4	Cuba	1	T	11.0	St. Peters
F-8	Cullom	1	R	0.36	
C-6	Dixon	3	R	2.94	Mt. Simon
A-6	Durand	1	R	2.38	St. Peters
D-4	East Moline	3	R	2.55	Mt. Simon
C-8	Elgin	1	R	8.5	Galesville
C-8	Elgin	1, 2, 3, 5	T	0.66	1, 2, 3 Galesville 5 limestone
F-4	Farlington	2	R	7.46	St. Peters typical
B-5	Freeport	4	R	3.62	St. Peters
E-6	Granville	2	R	10.2	St. Peters
G-4	Ipava	1	R	25.4	St. Peters only typical
G-4	Ipava	1	RT	25.4	St. Peters only
B-5	Lanark	3	R	1.35	Galesville
B-5	Lanark	2	R	0.41	St. Peters
E-7	Lostant		RT	10.1	St. Peters
F-7	Minonk	2	R	7.97	St. Peters, typical
D-8	Morris	3	R	8.20	St. Peters only typical
D-8	Morris	4	R	2.37	Galesville only typical
D-8	Morris	3, 4, 5	T	6.35	
B-5	Mt. Carroll	2	R	2.41	St. Peters
B-5	Mt. Carroll	3	R	2.76	
B-6	Mt. Morris	4	R	1.97	Mt. Simon
B-6	Mt. Morris	3	R	0.69	Mt. Simon
C-6	Oregon	2	R	3.16	Mt. Simon
C-6	Oregon	1	R	3.52	Mt. Simon
D-7	Ottawa	8	R	6.60	Galesville typical
B-5	Pearl City	1	RT	1.43	St. Peters
B-6	Pecatonica	2	R	2.63	St. Peters
B-6	Pecatonica	1	R	0.78	St. Peters
D-6	Peru	4, 5	R	4.84	4, St. Peters 5, Mt. Simon
D-6	Peru	4, 5	T	1.23	4, St. Peters 5, Mt. Simon
D-9	Plainfield	5 group	R	8.24	Galesville typical
B-6	Rockford	2	R	3.65	Mt. Simon
A-7	Rockton	2	R	2.07	St. Peters
B-4	Savanna	2	R	3.09	Galesville
B-5	Shannon	2	R	17.3	St. Peters
A-7	So. Beloit	3	R	1.80	Mt. Simon
C-5	Sterling	1	R	4.02	Mt. Simon
C-5	Sterling	1, 2, 3, 4	T	4.83	
D-6	Sublette	1	R	0.73	St. Peters
E-6	Toluca	2	R	7.4	St. Peters typical
E-5	Wyoming	2	R	11.1	St. Peters

(a) Raw (R) treated (T) type uncertain (RT) may be either raw or treated.

(b) Estimated error 3 per cent or 0.02, whichever is larger.

(c) Comments of Illinois State Water Survey about samples submitted for chemical analysis and deepest formation penetrated.

This well obtains water from the St. Peters, Galesville and Mt. Simon formations. Thus it seems most likely that the radium found is associated with water from the St. Peters sandstone formation. Analysis of core samples from a deep well are under investigation.

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MEASUREMENT OF GAMMA-RAY EMITTING ELEMENTS PRESENT IN SOIL

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The gamma-ray spectrum of fallout and natural radioactivity as obtained from both extended and finite (200 g) samples of soil has been described in a previous report.⁽¹⁾ Additional work has shown that somewhat larger samples may be used to advantage without an appreciable increase in scattered radiation. One-kg soil samples placed in 1-liter polyethylene bottles have been used as the standard sample size. Such a sample results in an increase in counting rate of a factor of 4.6 over that found with a 200 g sample having the same concentration of activity. Fig. 34 shows a comparison of gamma-ray spectra of two top soil samples, one collected in August 1950 and the other in October 1957. Both were collected on the Argonne site and were counted in October 1957. The fresh sample clearly indicates the presence of three lines which are absent in the spectrum from the older sample. These lines are due to the fission products indicated in the figure.

The intensity of the gamma rays from these fission products is most misleading if any attempt to derive the dose rate due to fallout is made from such data. Due account must be taken of the concentration and distribution of both natural and fission radioactivity before any reasonable estimate of dose may be made.

The fact that fission product activity is present in soil can thus be readily ascertained by gamma-ray spectroscopy; however, of more interest and importance is the actual concentration of such activity, and the dose arising therefrom. In order to arrive at the concentration it is necessary to determine the portion of the net counts from a given soil sample which are due to each radioisotope or radioactive series present in the sample. The method whereby this is done has been discussed briefly in the previous report.⁽¹⁾ Some modifications have been introduced in the methodology which increase the validity of the results and will bear a more detailed description.

The 5 in. x 4 in. NaI crystal with which the spectra were taken is canned in electrolytic copper, there being a thin copper window over the face of the crystal through which the radiation enters. This thin window may also allow beta rays to penetrate into the crystal, especially since

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Fig. 34

Fig. 35

the maximum beta energy coming from the fallout isotopes present in soil is 3.57 Mev (Rh^{106}). To avoid counting any beta rays, thereby reducing any uncertainty which might be due to differences in absorption either in the sample itself or in the polyethylene bottle, sufficient absorber was placed between sample and crystal to absorb 3.5 Mev betas completely. Empirically, 1/2 in. of Lucite was found to be satisfactory for this purpose. A Lucite holder with this thickness between bottle and crystal was designed to position the bottle so that reproducible sample geometry is maintained. The crystal and phototube assembly is mounted vertically, with the sample bottle also in a vertical position. One kg of soil placed in a 1-liter bottle constitutes a right cylinder 9 cm in diameter and 14 cm in height.

Na_3PO_4 is the mock soil which is used for background purposes as well as serving as a soil-like medium with which the individual radioisotopes are mixed to make up the standard sources. The fact that Na_3PO_4 represents a good approximation to true soil as far as its scattering and absorption properties for gamma rays are concerned is illustrated in Fig. 35. In the one case Cs^{137} was mixed with 1 kg of soil and in the other a like amount of Cs^{137} was added to 1 kg of Na_3PO_4 . Appropriate background subtractions were made, and the net spectra are almost identical. This validates the use of mock soil in the analytic procedures herein employed.

It is not possible to examine both high- and low- energy gamma rays with the same gain setting. Thus if the 2.62-Mev line from ThC'' is visible, the 0.134-Mev line from Ce^{144} is below the low energy threshold of the instrument. Analysis for all components of the complex gamma-ray spectrum from soil thus becomes a two step process. This is illustrated in Figs. 36 and 37. Figure 36 shows the higher energy portion of the spectrum running from about 0.2 to 3.0 Mev. The net spectrum is that obtained from a 1-kg soil sample counted for 16 hours after subtracting a 16-hour background run using a 1 kg of Na_3PO_4 . The most energetic line is that at 2.62 Mev due to ThC'' . By using a mock soil consisting of Na_3PO_4 mixed with monazite sand of known thorium content made up to 1-kg weight and similarly placed with respect to the crystal, the thorium spectrum may be normalized at the 2.62 Mev peak and peeled off throughout the entire spectrum. Since the thorium content of the standard sample is known and the counting time is also known, normalization expresses the thorium concentration in the unknown soil sample in terms of that in the standard. By this means the concentration of thorium in the soil sample was found to be 4.0×10^{-6} g/g soil. The spectrum remaining, also shown in Fig. 36, has lines at 1.76 and 2.17 Mev, both of which pertain to the uranium series; by a procedure identical to that used for the thorium series but utilizing mock soil and carnotite, the spectrum due to uranium is normalized at these energies and peeled off. The uranium concentration was found to be 5.3×10^{-6} g/g soil. This left the K^{40} peak at 1.46 Mev to be compared to that obtained from 1 kg of KCl , serving as a K^{40} source; normalization

Fig. 36

Fig. 37

showed 2.27×10^{-2} g K/g soil. The result of peeling this activity off is a spectrum due to fission products alone which, in turn, may be broken down into the contribution due to each fission product as indicated in the figure.

By increasing the gain, the spectrum shown in Fig. 37 is obtained from the same soil sample. Expanding the spectrum in this manner also provides greater separation between photopeaks as regards the number of channels between adjacent peaks. The peeling procedure starts with the removal of the natural activities. Since the unknown soil sample was counted for equal times at both gains, namely 16 hours, and since the standard reference sources were also run for equal lengths of time at both gains, the normalization factors will be the same. Thus the natural activities may be peeled off without resorting to further normalization. Successive normalization and removal of the fission isotopes in order of decreasing energy is also illustrated in Fig. 37. Such a procedure enables one to determine the concentration of the various radioisotopes present in a given soil sample.

On the basis of knowledge concerning the distribution in depth of fallout, samples were made 12 in. x 12 in. in area and down to a depth of $4\frac{1}{2}$ in. Natural and fission activities were determined, and the activity per square mile due to the fission products was calculated as indicated in Table 36.

Table 36

Fission product activity on the ground at
Argonne National Laboratory

Isotope	mc/mi ²		
	May	July	October
Zr ⁹⁵ -Nb ⁹⁵	180	195	230
Cs ¹³⁷	35	34	37
Ru ¹⁰⁶ -Rh ¹⁰⁶	175	180	180
Ru ¹⁰³	175	200	240
Ce ¹⁴⁴ -Pr ¹⁴⁴	240	260	275

Sampling was carried out in May, July, and October 1957 at the Argonne site. For the purpose of determining whether or not additional fission activity might be present in the Argonne samples due to the local facilities, soil was collected in July some 45 miles southwest of the laboratory. The fission activities found in this set of samples are shown in Table 37.

Table 37

Fission product activity on the ground
at Braidwood, Illinois as of July, 1957

Isotope	mc/mi ²
Zr ⁹⁵ -Nb ⁹⁵	160
Cs ¹³⁷	32
Ru ¹⁰⁶ -Rh ¹⁰⁶	200
Ru ¹⁰³	150
Ce ¹⁴⁴ -Pr ¹⁴⁴	285

In the case of all soil samples, sufficient time had elapsed between the time of collection and the time of counting to allow for the decay of the shorter lived isotopes. Correction for decay back to the time of collection was then made for the isotopes which were present and for which analysis was carried out.

As an additional step, the dose rates pertaining to the natural and fission activities found in local soil as of May 1957 were calculated. That due to uranium, thorium, and K⁴⁰ was determined, using a method developed by Hultqvist.⁽²⁾ The dose due to fission activity was calculated by a method proposed by Dunning,⁽³⁾ making the assumption that fallout comprises a uniform, non-self-absorbing surface layer. The dose values so obtained are therefore maximum values since, in fact, there is some distribution in depth of the fallout material. To compare the fallout dose with that arising from natural radioactivity and cosmic rays the value in mr/year was also computed, making allowance for the decay of the fission isotopes in the integration. These dose rates are shown in Table 38. In calculating the dose in mr/year a further assumption was made, namely, that the fallout dose arose solely from the activity on the ground in May 1957, taking no account of subsequent fallout which occurred. Ba¹⁴⁰-La¹⁴⁰ were detected in addition to the other fission isotopes in the October samples. Based on approximate activity values, the dose in mr/year from these radionuclides is from 1.0 to 1.5 mr.

Confidence in this entire method of reducing complex gamma-ray spectra into their individual parts was attained in several ways. Dummy samples were made up of Na₃PO₄ with known amounts of natural and/or fission radioisotopes mixed in. These dummy samples were then counted and analyzed as if they were unknown soil samples. Comparison between concentration values arrived at by gamma-ray analysis and the known amounts incorporated in the samples showed the results to be within

Table 38

Gamma-ray dose from the radioactivity in the ground
at Argonne National Laboratory as of May 1957
(Sample Series S_{II})

Isotope	$\mu r/\text{hour}$	mr/year*
U daughters	4.02	35.21
Th daughters	1.45	12.70
K ⁴⁰	3.32	29.08
Zr ⁹⁵ -Nb ⁹⁵	1.57* *	3.45
Cs ¹³⁷	0.12	1.08
Rh ¹⁰⁶	0.23	1.47
Ru ¹⁰³	0.50	0.70
Ce ¹⁴⁴ -Pr ¹⁴⁴	0.09	0.37
Total		84.06 mr/year***

*May 1957 to May 1958

**Assuming Zr⁹⁵-Nb⁹⁵ in equilibrium.

***7 mr from fallout, 77 mr from natural radioactivity,
and ≈ 35 mr due to cosmic rays.

10 per cent over a fairly wide range of values. Furthermore, the concentration values found for the natural radioelements in local soil are in good agreement with values obtained by other methods for the same type of soil.^(2,4) The Cs¹³⁷ activity expressed in mc/mi² for May 1957 was 35 mc, which value is in accord with the Sr⁹⁰ findings for this area based on chemical extraction and assay techniques.⁽⁵⁾ Since these two radionuclides are produced in like quantity during the fission process, are both daughters of radioactive noble gases, and have almost identical half-lives, their concentrations should likewise be very nearly equal. Since this was found to be the case, the usefulness of this nondestructive assay method in determining indirectly the Sr⁹⁰ concentration in soil is open to investigation. A more detailed study of this aspect of the soil work is contemplated.

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THE HUMAN SPECTROMETER

C. E. Miller

I. Introduction

In the period since the last report⁽¹⁾ a second iron-shielded facility has been constructed. The reduction in the gamma-ray background of an 8 by 4 inch NaI(Tl) crystal as the detector was surrounded with various thicknesses of Fe is reported. Extensive data is given for 0, 3/4, 1½, 3, 5½ and 8 inches of Fe. After the background with the steel present was obtained, the detector was surrounded with 1/8 inch of lead to determine the effects of lining the room with sheet lead. Summation of the data clearly indicates that little is gained by building Fe shields thicker than 5 inches until radioactively cleaner photomultipliers become available.

The radioactive body content of numerous subjects has been measured during the past 6 months. A 2-year follow-up study was made of an Am²⁴¹ subject. Four subjects who had inhaled radioactive material were brought to Argonne National Laboratory from other AEC installations. The isotopes were identified, the amount determined and their location ascertained.

A search for former dial painters in the vicinity of Chicago has yielded the identity and location of many subjects containing various amounts of Ra²²⁶, with prospects of eventually locating several hundred. Thus far 42 individuals have been measured prior to clinical studies now under way. Because of the low quantity in some of them, it has been necessary to evaluate the uncertainty in a radium measurement due to fluctuations in the subject's Cs¹³⁷ and I¹³¹ arising from fallout and in K⁴⁰ due to the subject's musculature. A lung-equivalent phantom which may be used to calibrate a detector for inhalation accidents is discussed.

II. Gamma Ray Background of Iron Room II

The complete success of this Division's human spectrometer has led to emulation of the facilities by scientific groups all over the world. Judging from requests for engineering drawings and visits, at least 25 projects, modeled directly on the Argonne type, are contemplated, and other groups are modifying the design for special purposes. A few groups are investigating other shielding materials with the hope of economizing on initial construction costs. Although the effect of steel shielding on the counting rate of NaI(Tl) crystals has been reported several times by our group,⁽²⁾ it seems advisable to continue to report pertinent information as it accumulates.

The construction of this Division's second Iron Room provided the opportunity to re-examine the attenuation of the background scintillation spectrum with a large 8 by 4 inch NaI(Tl) crystal as the walls of the room were made progressively thicker. The data tabulated in this report should prove valuable in the construction and design of other low-level facilities since background attenuation of bulky shieldings can be checked during the course of their construction and the data compared to the curves published in this report. Excessive deviation will warrant investigation and may lead to the recognition of contamination problems as they occur. The data in this report will also allow the calculation of the sensitivity and of the accuracy possible with a low-level room of given wall thickness. Others interested in particular types of measurement can determine beforehand the feasibility of an Iron Room for their particular purpose.

Comparison with Other Types of Shielding

All data were taken with the same NaI(Tl) crystal (8 inches in diameter, 4 inches thick) used in Iron Room I. The walls, floor and ceiling of the second room were built up uniformly to each scheduled interval so that all surfaces were of equal thickness while the background attenuation was being determined. Moreover, assessment was made of the shielding effect of 1/8-inch sheet lead placed immediately around the crystal at various wall thicknesses.

The scintillation spectrum of this large crystal in Iron Room II is plotted in Fig. 38 and recorded in Table 39 by thirty-one 50-kev wide bands. A wider segment of the spectrum was also obtained using bands 100 kev wide covering the energy spectrum from 50 kev to 3.150 Mev; during this series of tests the sheet lead around the crystal was not used since 1/8 inch of lead would not substantially affect it at higher energies. These data are given in Table 40.

After the walls had been built up to their full 8-inch thickness, the inside surfaces of the room were covered with 1/8-inch thick sheet lead. The low-energy background pulses were reduced but not to the degree expected from the results obtained with Iron Room I (see below). The background with the crystal surrounded with sheet lead was higher than without the additional lead on the walls, thus demonstrating that this particular lead was contaminated.

To demonstrate the pattern of attenuation, the total background c/m in the neighborhood of the K^{40} peak (1.325 - 1.575 Mev band) were determined for each thickness and plotted as the solid line in Fig. 39. This curve is apparently composed of two components, one of which may be considered as due to natural activity originating entirely outside of the walls, and this fraction should decrease as the thickness of the wall is increased. The second component is due to the activity in the detector,

Table 39
Gamma-ray background reduction

Energy band, kev	0" Fe	3/4" Fe	1 1/2" Fe	1 1/2" Fe +Pb	3" Fe	3" Fe +Pb	5 1/2" Fe	5 1/2" Fe +Pb	8" Fe	8" Fe +Pb
Counts per minute										
25-75	3707	507	256	152	142	85.6	88	61.7	76.1	56.5
75-125	5693	1231	566	197	261	99.2	142	60.9	120	54.6
125-175	4768	1622	816	216	315	106	168	62.0	141	56.0
175-225	3665	1445	768	226	280	107	151	63.7	125	56.5
225-275	2674	1141	624	226	224	105	119	61.4	96.2	53.6
275-325	2018	905	498	227	185	102	96.9	56.5	80.3	49.6
325-375	1543	728	407	216	154	97.1	81.8	54.2	66.2	46.9
375-425	1168	589	345	203	138	92.9	69.1	51.7	60.1	45.9
425-475	1033	526	313	205	136	94.7	70.6	53.2	59.9	47.2
475-525	936	476	290	192	125	88.5	69.1	52.5	59.7	43.9
525-575	894	413	243	167	94.4	68.1	49.9	39.9	42.0	36.7
575-625	814	362	210	151	83.3	65.5	43.7	36.2	36.5	32.3
625-675	602	293	179	131	72.2	58.5	37.2	32.9	31.0	28.6
675-725	526	263	157	121	64.6	53.6	34.5	30.0	27.1	26.3
725-775	485	243	145	112	60.6	49.7	31.5	29.0	26.4	24.0
	(30,527)	(10,745)	(5,816)	(2,743)	(2,334)	(1,273)	(1,251)	(746)	(1,017)	(659)
775-825	467	217	132	105	55.6	45.3	27.5	25.8	24.7	23.4
825-875	436	205	122	95.6	49.8	41.7	25.7	23.2	20.3	19.3
875-925	416	194	112	86.8	44.5	37.9	21.9	20.6	19.0	18.3
925-975	387	177	103	80.0	40.7	34.0	20.4	18.1	16.3	14.7
975-1025	326	157	94.3	72.6	36.4	30.9	17.6	16.0	14.7	13.3
1025-1075	313	146	84.5	68.0	32.9	27.9	15.8	14.5	13.3	11.8
1075-1125	306	136	80.5	65.0	30.2	25.3	13.0	11.6	11.0	10.2
1125-1175	287	130	74.2	58.6	26.9	22.4	11.8	9.6	8.8	8.5
1175-1225	262	118	66.0	54.1	24.4	20.5	10.4	8.9	7.3	6.5
1225-1275	253	108	62.7	52.0	22.8	19.0	9.4	8.5	7.4	6.7
1275-1325	268	112	60.7	48.6	21.5	18.7	9.3	8.3	6.6	7.3
1325-1375	303	122	64.5	51.5	22.4	19.4	9.5	8.6	7.5	6.7
1375-1425	405	146	75.8	57.4	24.5	21.1	9.7	8.7	8.8	7.3
1425-1475	426	159	79.5	60.9	25.3	20.4	9.4	8.8	7.5	7.4
1475-1525	331	122	58.0	46.7	18.9	15.0	8.4	7.5	6.1	6.3
1525-1575	180	77.2	38.9	32.6	13.7	11.0	6.4	5.4	4.9	5.2
	(5,363)	(2,326)	(1,310)	(1,035)	(491)	(411)	(226)	(204)	(184)	(173)

Table 40

Gamma-ray background reduction

Energy band, kev	0" Fe	3/4" Fe	1½" Fe	3" Fe	5½" Fe	8" Fe
Counts per minute						
50-150	10,192	2,874	1,437	582	315	263
150-250	6,174	2,575	1,384	500	272	224
250-350	3,504	1,622	891	330	178	152
350-450	2,236	1,124	662	269	147	122
450-550	1,895	887	521	213	120	103
550-650	1,346	660	381	148	82.6	70.1
650-750	1,040	510	300	122	64.9	56.3
750-850	906	425	248	98.4	52.5	44.6
850-950	792	368	211	81.7	41.1	33.9
950-1050	658	296	172	66.8	30.6	25.5
1050-1150	585	266	147	54.3	24.1	19.6
1150-1250	528	224	131	46.7	18.8	15.4
1250-1350	688	254	137	47.2	18.9	15.1
1350-1450	818	307	154	48.1	19	16.4
1450-1550	322	148	81	27.2	12.2	9.8
	(31,683)	(12,540)	(6,855)	(2,633)	(1,397)	(1,170)
1550-1650	185	82	49.2	17.9	9	7.2
1650-1750	160	72.7	41.8	16.3	7.6	5.6
1750-1850	101	48.3	30	12.0	6.1	4.5
1850-1950	68.6	35.9	22.2	10.9	5.5	4.4
1950-2050	73.3	35.8	22.1	9.6	4.8	3.7
2050-2150	78.1	39.6	22.4	9.8	4.8	4.0
2150-2250	64.7	31.2	20.0	9.4	4.3	3.6
2250-2350	55.9	27	17.1	7.5	3.8	3.5
2350-2450	60.9	29.4	17.2	7.3	3.7	3.3
2450-2550	80.4	35.6	18.9	8.4	3.8	3.1
2550-2650	60.7	29.4	18.2	6.7	3.0	2.9
2650-2750	22.9	13.4	9.2	4.4	2.8	2.6
2750-2850	10.8	6.4	5.2	3.6	2.5	2.5
2850-2950	7.2	5.5	4.1	3.3	2.7	2.2
2950-3050	4.6	4.5	3.6	2.6	2.7	2.5
3050-3150	4.9	4.7	3.7	3.0	2.7	2.4
	(1,040)	(501)	(305)	(133)	(70)	(58)

detector mount, inner plates of the room and cosmic rays. This value should not change appreciably as steel is added to the outside of the room. Subtracting 34 c/m from the background at each wall thickness yields the straight dashed line (Fig. 39), a result consistent with the hypothesis that within this band, 34 c/m originate from cosmic rays and internal activity within the confines of the room and less than 1 c/m originates outside the room.

Fig. 38 Scintillation spectra of gamma-ray background of Steel Room II as a function of wall thickness.

Fig. 39. Gamma-ray background of Steel Room II between 1.325 and 1.575 Mev as a function of wall thickness.

A similar analysis performed across the spectrum indicates that with 8-inch Fe walls about 1 c/m per 50-keV band at the low energies and 0.2 c/m per 50-keV band at the higher energies were originating outside the room. The empirically determined straight lines determined from the calculations at all these other energy spans had practically identical slopes. The mass absorption coefficient calculated from these slopes is 0.045; namely, the mass absorption coefficient of iron from gamma rays in the 1.5 - 2 MeV region. The reason for the fact that the same mass absorption coefficient is found for low energies of the spectrum is not immediately apparent, but this finding could conceivably be due to the fact that most of the low energy pulses observed are due to the degraded high-energy rays from the Ra and Th present in the surrounding concrete.

From the above studies, it seems evident that 95% of the background inside the 8-inch Iron Room is indigenous to the room and cosmic rays; therefore, any practical amount of additional shielding will not significantly reduce the background.

The data of Table 39 demonstrate also that the low-energy background can be reduced by adding 1/8 inch lead on the interior walls. These low-energy rays which are absorbed by the lead apparently originate within the steel. It has not been demonstrated whether these counts are due to the interaction of cosmic rays or due to contamination of the steel.

III. Iron Room I

The reduction in background realized by covering the floor and one wall of Iron Room I with 1/8 inch sheet lead was reported in the last semiannual report.⁽¹⁾ The remaining interior surfaces have since been covered with lead and the reduction in background noted. An additional shielding of 1/8-inch sheet lead was positioned immediately around the crystal and a further reduction in background noted. This further reduction cannot be easily interpreted, but it may in part be due to the result of shielding the detector from natural airborne activity such as radon and other gases within the enclosure, like A^{41} emitted by the research reactor and N^{13} from the cyclotron. Additional studies will be made and reported at a later date.

The data obtained with Iron Room I and the comparable data from Iron Room II are assembled in Table 41. A comparison of the data of the two rooms seems to indicate that some material used in Room II is contaminated.

The radium and K^{40} present in the photomultiplier tube have been found to be responsible for 275 c/m of the background of the 8 by 4-inch crystal between 25 kev and 1.575 Mev. It has been calculated that the K^{40} content of the crystal would account for an additional 170 counts. These two spectra were subtracted from the actual background. The residual spectrum, suffering from considerable statistical uncertainties, suggests the presence of gamma rays at 510 kev, 860 kev and 1.3 Mev, and the presence of a small amount of Ra^{226} .

It should be realized that although it is desirable to reduce the background as much as possible when counting low activity samples, the background itself, in the region below 800 kev, is not of primary importance when measuring the activity of the human body. As will be shown in a later section, the variations of I^{131} and Cs^{137} in the body seem to be the factors limiting the ultimate sensitivity of the present experimental setup.

IV. A Follow-up Study of an Americium Accident

A chemist who had received a puncture type wound of the hand while working with Am^{241} was studied on September 8, 1955 and the results reported.⁽³⁾ This individual was brought back on July 25, 1957 and the

Table 41

Comparison of backgrounds in rooms I and II

Energy band, kev	8" Fe walls		8" Fe + 1/8" Pb on walls		8" Fe + 1/8" Pb on walls + 1/8" Pb around crystal	
	Room I	Room II	Room I	Room II	Room I	Room II
	Counts per minute					
25-75	79.4	76.1	76.2	118	60.9	63.1
75-125	110.5	119.8	66.7	96.4	50.4	59.3
125-175	113	140.8	71.4	98.5	50.5	59.4
175-225	98.4	124.5	69	97.9	52.5	60.2
225-275	80.1	96.2	63.7	85.4	48.7	54.1
275-325	64.8	80.3	58.1	74.2	45.1	50
325-375	55.4	66.2	51.9	64	41.4	49.4
375-425	50.8	60.1	46.1	57.1	41.4	46.1
425-475	50	59.9	48	53.8	40.6	44.1
475-525	44	59.7	46.1	52.7	40.1	43.3
525-575	36.8	42	38.3	41	34.6	36.4
575-625	33.6	36.5	34.4	35.6	30	33.5
625-675	28.5	31	30.5	30.4	24.7	28.6
675-725	26.6	27.1	26.7	28.4	24.7	27.4
725-775	26.5	26.4	24.7	25	25.2	25.7
	(898)	(1047)	(752)	(958)	(611)	(680)
775-825	24.2	24.7	23.4	23.8	22.7	23.6
825-875	21.4	20.3	20	20.9	20.4	20.0
875-925	20.5	19.0	19.9	18.9	17.6	18.7
925-975	17.1	16.3	17.5	15.9	16.7	17.1
975-1025	14.7	14.7	15	14.3	13.3	14
1025-1075	12.5	13.3	12.6	12.9	12.3	12.8
1075-1125	10.9	11	11	11.5	10.5	11.1
1125-1175	9.2	8.8	10.2	9.9	9.4	9.2
1175-1225	8.1	7.3	8.2	7.9	8.5	8.7
1225-1275	7.3	7.4	7.8	7.4	7.2	7.7
1275-1325	7.6	6.6	8.2	7.3	7.7	7.9
1325-1375	8.2	7.5	8	7.3	8.7	7.8
1375-1425	8.4	8.8	9.4	7.5	8.7	8.2
1425-1475	8.8	7.5	9.1	7.6	8.3	7.4
1475-1525	6.8	6.1	6.9	6.4	6.7	6.5
1525-1575	5.4	4.9	5.7	4.9	4.8	4.6
	(191)	(184)	(193)	(184)	(184)	(186)

study of his gamma activity was repeated with the same collimated crystal. The body burden and body distribution was identical with that found almost 2 years ago, indicating no appreciable migration within the interval.

The hand was scanned using a 1/4 by 1/4-inch NaI(Tl) medical scintillation probe, previously described.⁽⁴⁾ The bulk (ca. 90%) of the remaining material was found to be concentrated in one area, less than 1/4 inch in diameter, just below the surface about 1/8 inch to the side of the old incision line. Figure 40 shows a 1/2-inch diameter circle drawn around the outside of the crystal holder. The activity was located in the darker erythematous area. Utilization of this type of scintillation probe in the operating room should permit ready excision of the contaminated tissue.

Fig. 40. Photograph of subject's hand contaminated with Am^{241} from a puncture type wound. See text.

V. The Presence of I^{131} Fallout in Humans

The net in vivo spectrum of CEM was obtained 8-1-57 in the course of the long-range evaluation of Cs^{137} in humans. The low-energy end of the spectrum was observed to be higher than was to be expected from previous

observations (Fig. 41). The run was repeated the following day with identical results. In order to pursue the matter further, the subject's net *in vivo* spectrum of 3-22-57 was subtracted from the anomalous spectrum, and the spectrum of Fig. 42 resulted. The main radioactive isotope responsible for the result was found to be I^{131} with some Ru^{106} - Rd^{106} and Zn^{95} - Nb^{95} present. The Cs^{137} content was also slightly higher than as of 3-22-57. A collimated crystal was placed over the subject's thyroid and a distinct I^{131} spectrum was evident.

Fig. 41. Net gamma-ray scintillation spectra of control subject C.E.M.

Fig. 42. Increase in net spectrum of control subject C.E.M. from 3-22 to 8-2-57.

All the other control subjects were then checked near that date, and amounts varying from 10^{-10} to 10^{-9} c of I^{131} were found.

Additional subjects from other parts of the U.S. were also checked, and the I^{131} photopeaks observed were of the same order as those found in local residents.

VI. Ra^{226} and Th^{232} Activity in Humans

Interest in the long-term effects of radioactivity in humans has focused our attention on individuals who, by one means or another, have accumulated radium or thorium in their bodies for long periods of time. Contemporary literature indicates that thousands of people in all parts of the world have accumulated varying amounts of radioactive materials

from industrial employment or medical treatment. However, the number of individuals adequately studied,^(5,6) is not sufficient to establish, within satisfactorily narrow limits, the incidence of pathological manifestations as a function of radioactive burden. Moreover, this limitation in number is aggravated by the fact that many of the subjects were selected for study by the appearance of clinical symptoms. It is obvious that an extension of this type of study to unselected large groups is most desirable. In this laboratory, in addition to the inquiry on the relationship between incidence of bone sarcoma with radium content of potable water of Illinois by A. F. Stehney, H. F. Lucas, and H. Auerbach, effort has not been spared to locate groups of individuals suited to the purpose of this task.

Radium Subjects

In the last few months, many individuals formerly connected with the radium dial paint industry have been traced by virtue of persistent efforts requiring a great deal of time mostly devoted to personal contacts with the subjects involved and to word-of-mouth education as to the strictly scientific intent of our labors. Response has ranged from the most enthusiastic cooperation to downright suspicious hostility, but the over-all effect has been gratifying. By means of group pictures at the time of employment, autograph and address books, Christmas card lists, etc., a nonindifferent group of about 300 people has been identified. Of these 42 have already been measured and found to have from 10^{-9} to 3×10^{-6} c of Ra^{226} . It is hoped to continue the study in the immediate future and to establish, if possible, the entire numerical potential in the vicinity.

Thorium²³² in Humans

Two groups of thorium-burdened people seem suitable for study: industrial workers of thorium refineries and patients injected with Thorotrast.

Thorium refineries have been handling thorium compounds for many years, and undoubtedly many workers in these plants must carry detectable amounts. Two hundred people are located in the vicinity of the Laboratory and hence potentially available for study. Since the main route of contamination is the lung, the physical studies in prospect are not expected to be simple. One subject - connected with the firm for several decades - was measured as a test some time ago with the $4 \times 1\frac{1}{2}$ inch NaI crystal and showed Ra^{228} and Ra^{224} photopeaks corresponding to about 50 mg of Th^{232} in equilibrium with its daughter products.

Thorotrast (ThO_2) was injected into humans during the early thirties for various diagnostic purposes. The potential usefulness of this group, however, remains to be elucidated because of the unsettled question of life

expectancy. It is to be noted, however, that the injected group is undoubtedly large (a single incomplete survey⁽⁷⁾ reports nearly 5,000 cases in the United States alone), and that since the work was performed mostly by radiologists in hospitals, and records are available, there exists a good opportunity of follow-up. Moreover since the volumes injected do not vary greatly and - judging from work reported to date - the injection of ThO_2 is followed by a quantitatively fairly well defined pattern of deposition and excretion - it might be possible to evaluate the series by relatively modest effort without a great number of physical measurements beyond those required to establish without doubt the dosimetric features of the element. To date one Thorotrast patient has been measured and it is reported below:

Study of Thorotrast in Humans

The in vivo gamma ray spectrum of a patient who received 10 cc of Thorotrast intravenously in 1939 was obtained utilizing the "one-meter-arc" and the "standard-chair" techniques. In addition, a rough estimate of the body distribution was determined by positioning the crystal 1 cm above the umbilicus, abdomen and head. Both the absolute quantity and the ratio of the photopeaks to backscatter troughs were much higher at the head indicating that a considerable amount of the material was still present at the locus of the injection in the neck.

The 1-meter-arc spectrum and the standard-chair spectrum are identical if the value of the ordinates of the latter are reduced by a factor of 5.5. This ratio is higher than the 4.1 ratio found in the past for subjects containing Ra^{226} , Cs^{137} and K^{40} . This may be due to the unusual anatomical features of this subject, only 5' 2" tall but weighing 218 pounds; it may also be due to the fact that the material was not uniformly distributed throughout the body volume.

The spectra were analyzed in the following manner: The gamma-ray spectrum of a normal unexposed subject of the same height and weight was subtracted from the spectrum of this Thorotrast patient, yielding a net thorium spectrum as shown in Figs. 43 and 44. The spectra obtained from both a RdTh source and an old Thorotrast source (1937) buried in a 28-cm phantom were normalized so that the area under the 2.62 Mev ThC' photopeak matched the area under the patient's ThC'' 2.62 Mev photopeak. The normalized RdTh spectrum was then subtracted from both the patient's net spectrum and the normalized Thorotrast spectrum. The difference spectra, thus obtained, which are due to MsThII , should have been identical if the MsThII in the patient had been in equilibrium with its daughters and the scattering from the phantom was representative of the patient as a whole. From the portion of the patient's and phantom's spectra above 1 Mev, there is no reason to question the latter assumption.

Fig. 43. Net in vivo scintillation gamma-ray spectrum from 25 keV to 1.6 MeV obtained from a Thorotrast patient.

Fig. 44. Net in vivo gamma-ray scintillation spectrum from 50 keV to 2.8 MeV obtained from a Thorotrast patient.

Since the subject was found to contain 1.22×10^{-7} c of MsThII and 1.1×10^{-7} c of ThC'' or 12% more MsThII than the source, it seems likely that the patient was eliminating about 12% of the daughter products between MsThII and ThC'' . The value compares very well with the value found by Hurst et al. for the elimination of Ra^{224} from Thorotrast patients.⁽⁸⁾

VII. Observations and Calculations Concerning the Error of Ra^{226} Measurements in vivo

The 1-meter-arc technique as described in another report⁽⁹⁾ is the absolute method for determining the amount of radioactivity in a human body. In practice, measurements are made with the subject in the standard chair.⁽¹⁰⁾ The reasons for using the chair are that the subject is more comfortable, the subject's body can be more reliably positioned, and the crystal can be moved closer to the subject's body with a resultant higher counting rate and greater sensitivity.

As discussed in an earlier report⁽¹¹⁾ the chair position cannot be calibrated using a phantom since all parts of the body are not equally distant from the crystal. For certain purposes, such as measurement of K^{40} content, the chair can be "calibrated" directly by the administration of tracer doses of K^{42} to a standard subject. However, for obvious reasons, this technique is not applicable in the study of long-lived bone-seekers.

A radium and thorium calibration factor for the chair has been determined indirectly. A number of subjects with relatively large burdens of these elements have been counted both in the chair and on the arc. The

absolute amount in microcuries has been determined from the arc data. The net in vivo radium or thorium spectrum in c/m per 50-kev energy band obtained with the chair has been divided by the amount in microcuries found by the arc technique. The resultant parameter (counts per minute per μc per band, in the chair) can be used to calculate other subjects' body content directly from the chair measurements.

The gamma-ray spectrum of a radium patient is analyzed as follows: The net gamma-ray spectrum of a normal unexposed control subject of the same height, weight, and general build is subtracted from the net gamma-ray spectrum of the radium subject yielding the in vivo radium spectrum. The total counts per minute in each of three energy bands (75 kev to 375 kev, 525 kev to 675 kev, and 775 kev to 1.175 Mev) are divided by the factor evolved above, giving three independent evaluations for the subject's body content. The three values should agree if the net spectrum is due entirely to radium. The values will not agree if the patient's Cs^{137} or K^{40} level differed from that of the control's, or if there are any additional isotopes present. This may be the case when the Ra^{226} burden is low.

The theoretical uncertainty in a radium measurement is determined by the statistical variations in background in the spectrum of a normal unexposed control subject and in the in vivo radium spectrum. The actual measured values for background, CEM's natural and the calibration factors for radium are given in Table 42. The statistical variations to be expected for a 50-minute counting period have been computed for background and subject CEM.

Table 42

Uncertainty in in vivo radium measurements

Standard chair technique, counting time of 50 minutes

Energy band, kev	Background, c/m	CEM		<u>In vivo</u> c/m per μc of Ra	Theoretical uncertainty, μc of Ra	Realistic uncertainty, μc of Ra
		Gross, c/m	Net c/m			
75-375	408.5 ± 2.9	714.6 ± 3.8	306.1 ± 4.7	11,000	6.1×10^{-4}	3.10^{-3}
525-675	103.7 ± 1.4	168.2 ± 1.8	64.5 ± 2.3	1,650	2.1×10^{-3}	1.2×10^{-2}
775-1,175	105.5 ± 1.75	215.3 ± 2.1	64.8 ± 2.8	1,500	2.8×10^{-3}	2×10^{-3}

The statistical variation in the patient's net data due to his natural K^{40} and Cs^{137} and due to the room background will be the same as that given for the "Net CEM" column. Subtracting the net CEM control data

from the patient's net data yields a net difference with a statistical uncertainty of $\sqrt{2}$ times the net CEM uncertainty. The minimum theoretical uncertainty in a calculated body content (expressed in μc of radium) is found by the following expression:

$$\text{Minimum uncertainty} = \frac{\text{variation of net CEM} \times \sqrt{2}}{\text{c/m per } \mu\text{c of radium in vivo}} .$$

This value could be realized if the control subject were measured, given a tracer and remeasured within a few days.

In actual practice, the uncertainty is much greater since the amount of Cs^{137} depends upon an individual's diet. Control subject CEM's net counting rate was observed to increase by 164 c/m in the low energy band, 23.5 c/m in the middle band and 13.5 c/m in the high energy band between 3-22-57 and 8-2-57 because of I^{131} fallout intake. The errors resulting from using this individual's March spectrum as a control spectrum in August would result in errors of 1.5, 1.4 and 1×10^{-8} c, respectively.

The variation resulting by the substitution, for purposes of control, of people of the same height and weight at the same location on the same date would not be quite as extreme. Data from the control subjects suggest that a variation of 30 c/m in the lower band, 20 c/m in the middle band and 3 c/m in the upper band are representative of actual fluctuations leading to realistic uncertainties of 3×10^{-9} , 1.2×10^{-8} , and 2×10^{-9} c. See Table 42. The measured values for the radium dial painters support these as realistic uncertainty values.

VIII. Standard Deviations in the Measurement of Potassium in Humans

The uncertainty in measurement of K^{40} in a human being depends upon several factors such as the person's build, the relative position of the crystal with respect to the body, length of counting time, and energy band. The first two factors were examined and the results reported in a previous report from this Laboratory.⁽¹¹⁾ The remaining two factors have been examined and shown in Fig. 45 where a typical scintillation spectrum from an "uncontaminated" individual is shown.

Three energy bands were chosen as follows: Band I: 0.775 Mev to 1.275 Mev, Band II: 1.325 Mev to 1.575 Mev, and Band III: 0.775 Mev to 1.575 Mev, which includes Bands I and II. A counting time of 50 minutes was used for the background run and 10 minutes for the subject's run. The counting rates of background, subject J. P. (gross and net counts) and the attendant standard deviations are shown for each energy band.

Fig. 45. Gamma-ray scintillation spectrum of background, gross and net, of typical uncontaminated subject. The standard deviations of the net spectrum, assuming a 10-minute run, have been calculated for 3 bands.

It seems apparent that it is possible to determine a subject's K^{40} level with a standard error of 4%, using a 10 minute run.

In the past we have calculated a human's K level by using Bands I and II independently. Agreement between the two resulting values (by calibration with K^{42}) is a proof that the K^{40} value is correct and that the subject did not contain any appreciable amount of any other element emitting gamma rays in these bands.

IX. The Measurement of Natural Gamma Activity in Humans above 1.5 Mev

It has been observed for several years that with an 8-inch by 4-inch crystal a net human *in vivo* gamma-ray spectrum yielded about 6 counts per minute between 1.7 and 2.8 Mev over a background of 50 counts per minute. This section of the energy spectrum was examined in detail (1) to explain the source of these counts, and (2) to determine the possibility of measuring the natural radium and thorium content of humans by detecting the high energy gamma rays from these isotopes. In theory, greater accuracy might be achieved using this energy band since no other isotope occurring in humans emits gamma rays above the 1.46 Mev of K^{40} .

On the basis of data obtained from subjects with relatively large Ra^{226} burdens, the 6 c/m would suggest a body content of about 10 times the natural value, since it would require a burden of 6×10^{-9} c of radium or 1.2×10^{-9} c of thorium to yield 6 c/m. A. F. Stehney finds a maximum

of 4×10^{-10} c of radium⁽¹²⁾ and predicts about 10^{-10} c of thorium⁽¹³⁾ in individuals from the Joliet area. These amounts would account for a total of approximately 1 c/m only.

It has been suspected that the body must be scattering secondary rays from cosmic rays into the detector. The background was obtained with the chair alone, with cans filled with distilled H_2O in place of the humans and with empty cans. The background with the distilled water present was about 5 c/m higher than the other two, demonstrating that the body would increase the apparent background between 1.7 and 2.8 Mev. The increase in background at low energies, while detectable, is not significant when compared to the actual counting rates.

These additional counts are probably the result of cosmic rays interacting in the human or the H_2O . Comparison of the background from 2.85 to 3.25 Mev, which is above the most energetic naturally occurring gamma ray, with the background from 2 to 2.85 Mev, leads to the conclusion that most of the background counts in the latter energy band are due to cosmic rays. The Ra and Th present in unexposed humans might be measured utilizing these energy bands if the cosmic rays' photons produced could be eliminated by anticoincidence counting. The counting rates will be rather low, about 1 c/m, and long counting periods will be required. A 16-hour count, the time usually required for an accurate radon breath test on a normal subject,⁽¹²⁾ would give 960 counts due to the same source. This could be measured with about 10% uncertainty of the background. Some advantages might accrue: the subjects may be more comfortable during these long counting periods than when being measured on the radon breath apparatus. They would not have to wear a mask, with the attendant discomforts, but would recline in the contour chair. The run could be interrupted and the subjects allowed to walk around for periods of time with no adverse effects. This probability will be kept under consideration.

X. Inhalation Accidents

In the period since the last report, three people who inhaled radioactive dust have been brought to our laboratory from out of town for an estimate of content and biological half-life. One individual, a machinist who inhaled a uranium compound containing U^{235} during a grinding operation, has been measured on three occasions. The other two subjects, who inhaled air-borne Pa^{233} , have been measured twice. All will return at future times for further studies.

The total body content, the amount in the lungs, and the quantity in selected volumes of the body have been measured during each visit. The subject's total body content has been measured using the standard-chair and the 1-meter-arc techniques. The total amounts in the lungs, head, shins, and abdomen have been measured by placing the 8-by-4-inch NaI(Tl) crystal about 1 cm over the ventral thoracic, dorsal thoracic, abdominal

and lumbar regions and the shins and the head. It is hoped that by repeated measurements the lung clearance rates and the fraction deposited in the bones for each of the two isotopes can be determined with some accuracy and compared to data from excretion studies.

The chest area of each subject was scanned with a collimated detector on his first visit, to determine whether the material was concentrated in any particular area or uniformly distributed throughout the lung volume. Nine points on a grid over the chest were measured. The distances from the top of the subject's head to the horizontal grid lines were 34, 42, and 50 cm, respectively, and the vertical grid lines were the subject's mid-line (over the sternum) and lines 8 cm to the right and to the left of the mid-line. The highest readings were found 8 cm to the right and left of the mid-line, 34 cm from the top of the head. The values on either side of the mid-line, 42 cm down were about 20% less. The points 50 cm down and 8 cm to the right and left were about one-fourth of the top values. The values along the mid-line were somewhat lower than the values on either side, demonstrating that the material was distributed throughout the lungs. Essentially the same distribution was found for all three individuals. These positions will be correlated to lung volumes by superimposing the grid on chest X-rays of each subject to define the chest volume which was actually scanned.

The reading obtained with the crystal 1 cm from the back was identical to the reading obtained with the crystal 1 cm from the chest proving that the material was either located on a plane midway from the front to the back of the individual, or that it was distributed throughout the lung volume.

U²³⁵ Case Data

The total amount of uranium in the thorax was calculated using the techniques described in the following section. The dates of the measurements and the amounts were:

<u>Date</u>	<u>Total amount in chest area, mg</u>	<u>Decrease, mg</u>
11-2-57	2.75	
11-19-57	2.05	0.7
12-10-57	1.47	0.58

These three values were plotted on a semilog graph, fall on essentially a straight line and indicate a biological half-life in the lungs of 41.5 days. This value is considerably shorter than the value of 125 days usually assumed.

All of the subject's urine has been collected and analyzed by the employer's bio-assay group. They report that the subject eliminated 0.75 mg during the first period and 0.45 mg during the second period. These values are in substantial agreement with the amount leaving the lungs as estimated by the above measurements. These data demonstrate that most, if not all, of the material is being eliminated from the body after it leaves the lungs and that apparently it is not being deposited in other organs in comparable amounts.

To date, no detectable amount has been found in any organs other than the lungs. The measurement made over the kidney region is a few per cent of the lung value. Measurements made with appropriate phantoms in place of the human demonstrate that the gamma rays detected over the kidney are probably scattered rays originating in the chest volume. More exact measurements of the more sluggish U deposits will be possible after the relatively short-lived lung content has been eliminated.

The half-time of Pa^{233} in the lungs cannot be stated with certainty on the basis of only two measurements. A biological effective half-time, i.e., $[T\ 1/2\ (\text{eff}) = T\ 1/2\ (\text{phys}) \cdot T\ 1/2\ (\text{biol}) / T\ 1/2\ (\text{phys}) + T\ 1/2\ (\text{biol})]$ of 22.6 days was found for both subjects. Pa^{233} thus has a biological half-time in the lungs of 129 days which is in agreement with the 112-day half-life found for RaSO_4 by Marinelli, et al.⁽¹⁶⁾ but at variance with the half-life in the lung of 66 days for T^{129} and 41.5 days for U^{235} as observed by Miller.⁽¹⁴⁾

The lung volumes were scanned on the second visit and about the same distribution noted as on the first visit. It was felt that the original distribution might reflect different particle sizes and that the lung clearance rates might also reflect these effects. However, the scans were not exact enough for this detailed study.

XI. Phantom Calibrations for Inhalation Cases

The net counts per minute as obtained from measurements on humans must be converted into an absolute quantity of radioactivity if the measurement (and the facility) is to serve any useful purpose. A constant, such as the net in vivo counts per minute per microcurie, must be determined for the particular isotope involved, the particular geometry used and the specific location of the isotope in the body.

The inhalation case is the most difficult one to calibrate since the material is distributed throughout a low-density volume which is surrounded by a denser layer of unknown thickness. The U^{235} and Pa^{233} inhalation cases have enabled us to reappraise and extend some previous observations. The data obtained with U^{235} will be given in detail.

The expression, counts per minute per microcurie, must be obtained from an appropriate phantom since it is impractical to use tracers in humans for inhalation studies in these cases. The ratio of the area under the backscatter peak to the area under the photopeak may be used to test the "equivalence" of the phantom to the human. In the case of U^{235} , the ratio was established as the area under the curve from 71 keV to 126 keV (the uranium X-ray plus backscatter from the 188-keV gamma ray) divided by the area under the curve from 154 keV to 209 keV (the 188-keV photopeak). The in vivo ratio was found to be 1.14. The measurement and ratio from the back of the subject was identical to the reading taken from the front.

The following tests were made to arrive at an equivalent phantom. A 100-mg uranium source was placed at various depths in a 20-cm thick phantom built up of 1/4-inch sheets of Presdwood. The crystal was positioned 1 cm above the top plate of the stack as it was for the measurement in vivo over the thorax. The ratio was found to vary from 0.8 when the source was placed on top of the phantom to 1.8 when the source was buried the full 20 cm. The phantom ratio as measured at the top surface matched the in vivo ratio when the source was buried under the 1.4 cm of Presdwood. This, of course, cannot be the location of the U^{235} source in vivo. Moreover, it was not possible to distribute sources through a solid phantom which would yield this ratio for both front and back measurements. The contribution from the source buried at the far side increased the ratio considerably above the desired value. This indicated the total mass through the phantom was too great.

A phantom was constructed as shown by Fig. 46 to approximate the mass distribution of the human. The mass at positions X and Y correspond to the denser masses of the chest walls and the spaced area approximates the spongy tissue of the lung cavity. The over-all thickness of the phantom was made equal to the subject's thickness to provide the same geometry.

The source was located at the center of the phantom and the thicknesses used at X and Y varied. Varying the back thickness (Y) varies the area under the backscatter peak only. Increasing the front thickness (X) reduces both the photopeak and the backscatter peak. The phantom uranium spectrum matched the in vivo spectrum as shown by Fig. 47 when 6 cm of Presdwood was used in positions X and Y.

The chest span indicated that the material was distributed through the lung volume rather than concentrated at a point. The calibration of the phantom must therefore be corrected for this effect. The source was placed at the five positions (A,B,C,D,E) shown by Fig. 46 and the spectra obtained. Due to the inverse square effect, the average reading was 7% higher than the reading obtained with the source at the center, point C. The ratio varied from 1.04 when the source was at position A to 1.3 with the source at position E, with an average value of 1.12.

Fig. 46. Cross-sectional view of Presdwood phantom used to reproduce a gamma-ray spectrum obtained from radio-activity in a human's lung.

Fig. 47. A comparison of U^{235} gamma ray spectra obtained from the lung-equivalent phantom with an actual lung burden.

The source was then placed at the center of the phantom and at 3, 6, 9, 12, and 15 cm to the left on the mid-plane as shown by positions I through VI. The ratio did not change although the average value for the simulated distributed source was 33% lower than the value at the center.

The counts per minute per milligram of U^{235} as found with the source located at the center was multiplied by 1.07 to account for the inverse square effect and by 0.67 to account for the distribution effect. The net counts per minute as measured from the subject divided by this factor yields the lung content in milligrams.

Essentially the same approach was used during November, 1956 to build up an equivalent lung phantom for two Te^{129} inhalation accident subjects. The same phantom thicknesses and effects were noted at that time.

The spacing between the front thickness (X) and the back thickness (Y) has a critical effect on the ratio. The crystal subtends a particular solid angle to the unscattered gamma rays from the source itself. The solid angle subtended to forward scattered rays in volume X is higher than the solid angle to the direct rays. The effective solid angle for the back scattered rays from volume Y is considerably lower than either of the other two. Bringing the volume Y closer to the source increases the back-scatter solid angle and increases the amount of backscatter detected by the crystal. Therefore the spacing between X and Y must be equal to the lung cavity.

The actual spacing used between the masses X and Y is not as critical as when the detector is placed 1 meter from the subject because the solid angles subtended to the front and back surfaces are almost equal. However, the thickness of volume X is still very important since increasing X increases the absorption and thus affects the counting rate per microcurie.

The physical construction of the phantom which is used to calibrate the 1-meter-arc thus depends upon the location of the radioactive element in the body, and this can be important when dealing with relatively soft gamma radiation. The spectrometric trial and error technique⁽¹⁴⁾ will probably result in more accurate values than the empirical approach.⁽¹⁵⁾ This was also suggested by measurements performed. In the case of one radium dial patient, where a phantom 18 cm thick was suggested by empirical methods based on the subject's dimensions it was found that an equivalent phantom 14 cm thick yielded the correct spectral shape.

In the case of a subject who had just drunk a K^{42} tracer, it was necessary to bury a K^{42} source 6 cm deep in a 22-cm thick solid phantom to match the spectral curve obtained from the front and back of the subject.

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RADIOSTRONTIUM IN RAIN WATER

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General Considerations

The primary purpose of the present study has been to investigate the possibility of estimating the rate of stratospheric Sr^{90} fallout, and thus the mean storage time of Sr^{90} , without resort to data from upper-air sampling or knowledge of the total amount of Sr^{90} so far introduced into the stratosphere.

An attempt is made herein to estimate the fraction of the stratospheric or "delayed" Sr^{90} in the total Sr^{90} transported to the ground by rain, by observing the variations of the relative amounts of Sr^{89} , Sr^{90} and Ba^{140} in a series of rainfalls. The principle of the method is as follows:

Suppose that (a) no material transfer to or from the stratosphere took place during a relatively short time interval between t_1 and t_2 , and (b) the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the stratosphere at time t_1 was $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_1}$. Then, the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the stratosphere at time t_2 will be

$$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2} = [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_1} \cdot \frac{e^{-\lambda_{89}(t_2 - t_1)}}{e^{-\lambda_{90}(t_2 - t_1)}} \quad (1)$$

where λ_{89} and λ_{90} are the decay constants of Sr^{89} and Sr^{90} , respectively. Similarly, if no material transfer to or from the troposphere took place during a time interval between t_1 and t_2 , the following relationship should hold between the $\text{Sr}^{89}/\text{Sr}^{90}$ ratios in the troposphere at t_1 and t_2 ;

$$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_2} = [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_1} \cdot \frac{e^{-\lambda_{89}(t_2 - t_1)}}{e^{-\lambda_{90}(t_2 - t_1)}} \quad (2)$$

Under the natural conditions, however, part of the strontium isotopes in the stratosphere will be transported to the troposphere during the time interval between t_1 and t_2 , and the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the troposphere at time t_2 will become different from that given by Eq. (2).

If the stratospheric fallout took place in such a manner that α curies of the stratospheric Sr^{90} were mixed with $(1 - \alpha)$ curies of the tropospheric Sr^{90} , the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the resulting troposphere at time t_2 would have become

$$\begin{aligned}
[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_2}' &= (1 - \alpha) [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_2} + \alpha [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2} \\
&= (1 - \alpha) [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_1} \cdot \frac{e^{-\lambda_{89}(t_2 - t_1)}}{e^{-\lambda_{90}(t_2 - t_1)}} \\
&\quad + \alpha [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2}
\end{aligned} \tag{3}$$

Assume that a rainfall occurred at time t_1 and the next rainfall occurred at time t_2 , and the $\text{Sr}^{89}/\text{Sr}^{90}$ ratios in the two rainfalls have been measured. We may consider that the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in rain water is the same as the ratio in the troposphere at the time of rainfall;

$$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1} = [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_1} \tag{4}$$

and

$$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2} = [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{T},t_2}' \tag{5}$$

Since $t_2 - t_1$ is usually of the order of magnitude of days or weeks,

$$e^{-\lambda_{90}(t_2 - t_1)} \approx 1 \tag{6}$$

Introducing (4), (5) and (6) into Eq. (3), one obtains

$$\begin{aligned}
[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2} &= (1 - \alpha) [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{89}(t_2 - t_1)} \\
&\quad + \alpha [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2}
\end{aligned} \tag{7}$$

A similar relationship should hold in the case of the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio:

$$\begin{aligned}
[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_2} &= (1 - \alpha) [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{140}(t_2 - t_1)} \\
&\quad + \alpha [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2}
\end{aligned} \tag{8}$$

The values of $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1}$, $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2}$, $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_1}$

and $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_2}$ can be measured experimentally, and α ,

$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2}$ and $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2}$ are the unknowns. If it is assumed that

$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2} = 0$ and $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2} = 0$, the following relationship is obtained from Eq. (7) and (8):

$$\begin{aligned} & [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2} / [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{89}(t_2 - t_1)} \\ & = [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_2} / [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{140}(t_2 - t_1)} \quad (9) \end{aligned}$$

Experimental data obtained so far, however, suggest that, in general, the following relationship holds:

$$\begin{aligned} & [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2} / [\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{89}(t_2 - t_1)} \\ & > [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_2} / [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_1} \cdot e^{-\lambda_{140}(t_2 - t_1)} \quad (10) \end{aligned}$$

indicating that

$$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2} > [\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2} \geq 0 \quad (11)$$

Because of the short half-life of Ba^{140} (12.8 days), the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio in the stratosphere will rapidly decrease after the introduction of the fresh fission product mixtures, while it will take considerably more time until the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the stratosphere becomes negligibly small.

If the time interval between the last stratospheric injection and t_1 is of the order of magnitude of a few months, one may consider $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2}$ to be practically zero, and it will be possible to calculate the value of α from the observed values $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_1}$ and $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t_2}$, according to Eq. (8). Introducing the value of α thus obtained and the observed values of $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_1}$ and $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t_2}$ into Eq. (7), the value of $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2}$ can also be calculated.

If, on the other hand, the time interval between the last stratospheric injection and t_1 is unknown, one may be able to calculate a minimum value of α by introducing $[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{S},t_2} = 0$ into Eq. (8). Introducing the minimum value of α thus obtained into Eq. (7), one may also be able to obtain a minimum value of $[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{S},t_2}$.

If the values of α for every rainfall that occurred during a certain period of time at a certain locality are known, it is a simple matter to calculate the rate of stratospheric Sr^{90} fallout in the area. It might also be possible to estimate the mean storage time of Sr^{90} in the stratosphere from the values of $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the stratosphere obtained by the method described above since the $\text{Sr}^{89}/\text{Sr}^{90}$ ratio in the stratosphere is related to the rate of removal of Sr^{90} from the stratosphere, as well as to the decay constants of Sr^{89} and Sr^{90} .

Experimental

Chemical Procedure

Sampling. The rain water samples were collected in six enamelled trays (surface area 1,500 sq cm each) placed on the roof of the Physics Building of Argonne National Laboratory.

Separation and purification of strontium and barium. A 2-ml portion of concentrated nitric acid and 5-ml portions of strontium and barium carrier solutions (10.66 mg Sr and 10.06 mg Ba per ml, respectively) were added to one liter of rain water, and the solution was evaporated to approximately 50 ml. The solution was neutralized with ammonia, and the strontium and barium were precipitated as the carbonates by the addition of a few grams of ammonium carbonate. The precipitate was dissolved in dilute nitric acid, and the insoluble residue was discarded. The solution was evaporated almost to dryness and treated with 20 ml of fuming nitric acid. The precipitate was filtered and washed with a few ml of fuming nitric acid. The precipitate was then dissolved in water, and the solution was neutralized with ammonia. A 2-ml portion of 6M acetic acid, 4 ml of 6M ammonium acetate, and 2 ml of potassium chromate solution were added; the barium chromate precipitate was filtered after the solution was warmed for about 10 minutes. One ml of barium carrier solution (10 mg Ba) was added to the filtrate and the precipitate was filtered. A few grams of ammonium carbonate were added to the filtrate and the strontium carrier was precipitated as the carbonate. The precipitate was filtered, washed with water, and dissolved in dilute nitric acid. One ml of ferric chloride solution (5 mg Fe per ml) was added to the solution and the ferric hydroxide was precipitated by the addition of ammonia. The precipitate was filtered and the scavenging procedure was repeated once more. The strontium was finally precipitated as the carbonate, filtered, washed, dried, weighed and mounted on a planchet.

The first barium chromate precipitate was dissolved in dilute nitric acid and one ml of strontium carrier solution (10 mg Sr per ml) was added to the solution. The solution was neutralized, and the barium carrier was again precipitated by the addition of acetic acid-ammonium acetate-potassium chromate mixture. The precipitate of barium was filtered and redissolved in dilute nitric acid. The solution was evaporated almost to dryness and treated with 20 ml of fuming nitric acid. The precipitate of barium nitrate was filtered, washed with fuming nitric acid, and dissolved in water. One ml of ferric chloride solution (5 mg Fe per ml) was added to the solution and the ferric hydroxide was precipitated by the addition of ammonia. The precipitate was filtered and the barium carrier was precipitated from the filtrate by the addition of ammonium carbonate. The precipitate was filtered, washed, dried, and mounted on a planchet.

Separation of Y^{90} from Sr^{90} . The strontium carbonate, after being counted for about two weeks, was dissolved in a few ml of dilute nitric acid, and one ml of ferric chloride solution (1.81 mg Fe per ml) was added to the solution. The solution was heated for a few minutes, and the ferric hydroxide was precipitated by the addition of ammonia. The precipitate was filtered, washed, redissolved in nitric acid, and the procedure was repeated three times. The final precipitate was collected on a glass fibre filter paper (2.4 cm, No. X-934-AH, Hurbur Paper Co). The precipitate was washed with hot water and acetone, dried and mounted on a planchet.

Radioactivity measurements. Two flow counters were used for the radioactivity measurements. The backgrounds of the two counters were 1.1 and 2.9 cpm in anti-coincidence with the surrounding cosmic ray counters.

Typical growth-decay curves are shown in Fig. 48.

Control experiments. The glass fibre filter paper used was found to be slightly radioactive, as shown in Table 43. The strontium and barium salts used were practically free of radioactive contamination, but the iron salt showed the presence of a slight contamination.

Table 43

Control experiments

	Background cpm	Observed activity, cpm	Difference, cpm
(1)Glass fibre filter paper	1.10 ± 0.03	1.25 ± 0.03	0.15 ± 0.06
(2)90 mg Sr carbonate on filter paper	1.10 ± 0.03	1.26 ± 0.03	0.16 ± 0.06
(3)72 mg Ba carbonate on filter paper	1.10 ± 0.03	1.14 ± 0.03	0.04 ± 0.06
(4)Fe carrier, 1.8 mg Fe on filter paper	2.90 ± 0.04	3.24 ± 0.04	0.34 ± 0.08

Fig. 48. Counting data on Oct. 22/23 rainfall.

Reproducibility of the results. The precision of the method is estimated to correspond to $< \pm 1\%$ average deviation for 10 - 100 cpm Ba^{140} and Sr^{89} , and $< \pm 10\%$ average deviation for about 1 cpm Sr^{90} .

The Sr^{89} determination of a sample of rain water was repeated twice and the results shown in Table 44 were obtained.

Table 44

Reproducibility of the results

Sample: Rain water collected at Argonne National Laboratory on August 28, 1957.

Exp. No.	Amount of sample, ml	Time of Sr separation, t_0	Activity at t_0 , cpm	Chemical yield, %	Self-absorption correction, %	Sr^{89} content as of Aug. 28, $\mu\mu\text{c}/1$
(1)	400	1430, Oct. 10	53.5 ± 0.5	86.9	21.2	341 ± 3
(2)	400	1508, Oct. 14	49.0 ± 0.5	83.5	20.0	339 ± 3

Experimental results. A set of experimental data obtained during the month of October 1957 is shown in Table 45. A series of weapon tests was held prior to the beginning of October and there was a recess period until the British hydrogen bomb test was held at Christmas Island on November 9, 1957.

Table 45

Relative amounts of Ba^{140} , Sr^{89} and Sr^{90} in rainfalls which occurred during the month of October 1957

Time of rainfall	Amount, inches	$[\text{Ba}^{140}/\text{Sr}^{90}]_{\text{R},t}$	$[\text{Sr}^{89}/\text{Sr}^{90}]_{\text{R},t}$
Oct. 15 (t_1)	0.56	301 ± 30	104 ± 10
Oct. 22/23 (t_2)	2.35	15.8 ± 1.6	17.5 ± 1.8

An additional set of data was made available to the author through the courtesy of Dr. J. Sedlet of the Industrial Hygiene and Safety Division of Argonne National Laboratory⁽¹⁾ and is shown in Table 46.

Table 46
Relative amounts of Ba^{140} , Sr^{89} and Sr^{90} in rainfalls
occurred in March and in April 1956

Time of rainfall	Amount, inches	$[Ba^{140}/Sr^{90}]_{R,t}$	$[Sr^{89}/Sr^{90}]_{R,t}$
March 27/28 (t_1)	0.28	7.05	14.1
April 2	0.02	not determined	not determined
April 25 (t_2)	0.42	0.79	9.3

Introducing the experimentally obtained values of $[Ba^{140}/Sr^{90}]_{R,t}$ shown in Table 45 into Equation 8, and assuming that

$$[Ba^{140}/Sr^{90}]_{S,t_2} = 0 ,$$

one obtains a minimum value of $\alpha (= 0.92 \pm 0.02)$ for the rainfall occurred on October 22 - 23, 1957.

Introducing the value of $\alpha = 0.92 \pm 0.02$ into Equation 7, one obtains a minimum value of the Sr^{89}/Sr^{90} ratio in the stratosphere at time t_2 (October 22-23, 1957)

$$[Sr^{89}/Sr^{90}]_{S,t_2} \text{ (minimum value)} = 11 \pm 5 \quad (12)$$

Introducing the experimentally obtained values of $[Ba^{140}/Sr^{90}]_{R,t}$ shown in Table 46 into Equation 8, and assuming that

$$[Ba^{140}/Sr^{90}]_{S,t_2} = 0 ,$$

one obtains a minimum value of $\alpha (= 0.46)$ for the rainfall occurred on April 25, 1956.

Introducing the value of $\alpha = 0.46$ into Equation 7, one obtains a minimum value of the Sr^{89}/Sr^{90} ratio in the stratosphere in time t_2 (April 25, 1956):

$$[Sr^{89}/Sr^{90}]_{S,t_2} \text{ (minimum value)} = 8.6 \quad (13)$$

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ION CHAMBER MONITORING OF FALLOUT AT THE ARGONNE SITE JULY 1 TO OCTOBER 10, 1957

H. A. May

The monitoring of atmospheric ionization has been continued throughout most of the reporting period. Instrumentation and preliminary results have been discussed previously.⁽¹⁾ Use of an unsealed ion chamber was at first considered desirable for rapid detection of gaseous or air-borne activity, such as reactor effluents; however, the large chart excursions resulting from the counting of alpha particles originating in the decay of radon and thoron daughters, together with the diurnal variation in radon level, seriously limits the sensitivity to fallout

The ion chamber first used was cylindrical in shape, having an inside diameter of 17.2 cm and inner length of 20.3 cm. The walls and the base away from the collecting electrode support were 1/8-inch bakelite, having a close-packed array of 1½-inch holes cut out, so that 38% of the wall area was air equivalent. A single layer of aluminum foil (less than 5 mg/square cm) covered the wall and end. The chamber was located inside the aluminum calibration garage, except for short intervals when readings were taken outside.

The effects noted above are well illustrated by an incident recorded on July 12 and 13, during and immediately after a very heavy rainfall. A section of the Esterline-Angus recording chart is reproduced as Fig. 49, showing an extremely large increase in chamber current. Extrapolation yields a peak value of at least 9 times normal background. The observed current is plotted on a semilog scale versus time in Fig. 50, the lower portion of the figure showing approximate rate of rainfall. Two maxima are observed, occurring 1½ to 2 hours after rainfall. Decrease in activity following the larger peak can be separated into two concurrent decays with half-lives of 58 minutes and 11.5 hours. Figure 49 shows the large bursts which are typical of heavily ionizing alpha particles within the unsealed chamber. Because of another experiment in progress in the calibration garage, which required use of the 250 Kvp X-ray generator, the chamber input was shorted until about 8:00 p.m., thus explaining the sudden rise at that time.

These findings suggest the sudden release to the local atmosphere of thoron in high concentration, resulting in decay of its daughters, Pb^{212} (thorium B--10.6 hours) and Bi^{212} (thorium C--60 minutes), within the chamber. It is well established⁽²⁾ that the background of beta-and gamma-activity may show a sharp rise following rainfall, so the incident reported may not be entirely surprising. Unfortunately, the filter-paper air sampler operated by the background monitoring group of the Industrial Hygiene and

Fig. 49

Fig. 50

Safety Division failed to collect useful samples during this period because of excessive moisture being blown into the air intake where the equipment was located. The time of occurrence--early in a weekend--also prevented prompt followup. Gamma analysis of surface soil samples collected shortly after the storm indicated an increase in fission product activity of not more than 20%.⁽³⁾ Considering the geometry involved, it would seem that this could not have contributed to the large current excursion.

A sealed chamber of identical dimensions was next obtained. The chamber walls and one end are 1/8-inch bakelite; the other end is 1/2-inch lucite. The greater thickness is required by the guard ring and high impedance connector assembly, which threads into the end plate, with O-ring seals. All fixed joints in the chamber are sealed with Glyptal varnish. Figure 51 illustrates typical records obtained with each chamber under comparable conditions.

Ion chamber current vs ionization in air may be calibrated by observation of the current produced by a standard radium source at known distance, or by absolute methods requiring only a knowledge of chamber volume and input resistance. In the first case, backscatter effects must be eliminated if the inverse-square law is to be strictly valid; in the second, the effective chamber volume may be difficult to determine, or alternately, elaborate field-defining electrodes are required. The chamber must be truly air equivalent in either case to assure electronic equilibrium.

By comparison of data obtained with the two chambers, it is calculated that the effective chamber volume of the second, sealed chamber is 4.25 liters, or 9.6% less than its physical dimensions indicate. The usual precautions to insure chamber saturation and freedom from collection polarity effects were performed. A potential of 90 volts, with electron collection at the center electrode, was employed for all measurements reported.

Typical calibration readings, using a radium in 0.5-mm Pt source, certified to contain 1.11 mg of radium, are summarized in Table 47. Current collected in the sealed chamber is consistently low by about 8%. Deviations from the inverse-square law appear to be within experimental error.

100 millivolts full scale
One horizontal division = 1 hour

UNSEALED CHAMBER
Showing diurnal variation in radon level, and "noise"
from alpha particles within chamber.

Load resistance = 9.82×10^8 ohms
Chamber volume = 4.7 liters

SEALED CHAMBER
Showing reduction in radon "drift" and "noise"
Automatic zero check at six hour intervals.

Fig. 51

Table 47

Distance, cm	Chamber 1--unsealed		Chamber 2--sealed	
	I, amperes	Id^2	I, amperes	Id^2
50	1.52×10^{-12}	3.80	1.32×10^{-12}	3.30
100	3.76×10^{-13}	3.76	3.48×10^{-13}	3.48
150	1.74×10^{-13}	3.92	1.525×10^{-13}	3.43
200	0.917×10^{-13}	3.67	0.838×10^{-13}	3.35
300	4.21×10^{-14}	3.79	3.74×10^{-14}	3.36
		mean $3.79 \pm .080$		$3.38 \pm .063$

Data taken with chamber and source 100 cm above concrete floor.
Separate experiments show no evidence of backscatter effects.

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TRANSPORTABLE BACKGROUND GAMMA-RAY SPECTROMETER

H. A. May

Following the demonstration of the feasibility of direct measurement by gamma-ray spectroscopy of fission products in the top soil, together with a quantitative determination of the principal natural radioelements present,⁽¹⁾ a request was received from the Division of Biology and Medicine, Atomic Energy Commission, Washington, to construct a transportable device capable of obtaining spectra, if necessary, in the field with a less expensive and less bulky apparatus.

Considerations of power consumption, ease of maintenance and initial cost led to the purchase of a 30-channel pulse-height analyzer of the multiple biased-discriminator type.* Consideration of cost and availability resulted in the selection of a 5-inch diameter by 4-inch sodium iodide (Tl) crystal, factory canned in electrolytic copper. A shield designed by J. E. Rose and R. F. Selman consists of four sheet steel (1/4-inch) welded tanks with appropriate offsets at all joints including the vertically hinged door to provide radiation baffles. A tank thickness of 8 inches provides, when filled with lead shot of 6.8 grams per cc, a minimum in any direction of ~ 5 inches of lead. The resulting gamma-ray attenuation (minima at 1.6 Mev) is equivalent to > 7.7 inches of iron to which the 1/2-inch tank walls may be added. The shield is thus of sufficient mass to produce the lowest possible backgrounds when operated with anti-coincidence rigs. The central body is U-shaped in horizontal cross section and to its top and bottom are bolted the D-shaped offset tanks which carry the ball bearing hinges for the offset door. All are equipped with suitable pipe plugs for draining the lead shot into canvas bags. In addition, the inside surfaces are lined with 1/4-inch lead to reduce the low energy component. The inner dimensions of the shield are $29\frac{3}{4}$ inches high, 14 inches wide, and 14 inches deep. Over-all height, exclusive of angle iron base is $47\frac{1}{2}$ inches. The empty steel shell weighs approximately 620 pounds. Total capacity is slightly more than 7,500 pounds of lead shot. Two men may readily assemble or disassemble the shield.

The feasibility of "peeling apart" a complex spectrum such as that from a soil sample containing numerous fission products in addition to the naturally-occurring radioisotopes depends almost entirely upon the stability of the amplifier and pulse-height analyzer employed, the latter being the limiting factor. The multiple discriminator type of analyzer is inherently subject to random drifts of adjacent channel boundaries, which can only be

*Designed by Larsh and Ghiorso at University of California Radiation Laboratory. Commercially available as model PA-400, Eldorado Electronics, Oakland, California.

detected by frequent calibration. When the activity level of the samples is such as to require long counting periods, little can be done about these residual random drifts, except to hope that they are sufficiently random and of appropriate period as to average out.

Numerous spectra have been taken of soil samples collected on the laboratory site in an attempt to evaluate the stability and reliability of the 30-channel analyzer. It was found convenient to adjust the low-energy intercept and over-all gain so that each channel covered a 50-kev energy band, and the total spread was from 50 kev to 1.55 Mev. Realignment of channel widths with a precision pulser was done about once a week, while energy calibrations at two points near the extremes were performed before and after each run. All determinations were made using 1-kg samples in polyethylene bottles, held in an inverted position in contact with the end surface of a 5 x 4-inch crystal, after the manner of Gustafson et al.⁽¹⁾ Thus any high-energy beta particles--as, for example, the 2.75-Mev beta from Pr^{144} --will also be counted. The following figures are presented as pertinent.

Figure 52 compares the net spectra of two counts made of the same surface soil sample, N-1, one of which was counted immediately upon collection and the other after 68 days. The presence of $\text{Ru-Rh}^{103,106}$ and Zr-Nb^{95} at around 500 and 750 kev, respectively, are clearly seen in the fresh sample, as well as a suggestion of a peak in between them, at around 620 kev. A decrease of 6 cpm in channel 12 would make this peak disappear. The standard deviation in the difference count for this channel was ± 0.93 c/m, so the peak is statistically significant. A drift of almost 10% in channel width during the sample run, or of +7% during the sample run and -7% during the Na_3PO_4 blank run would be required to make this much of a change. This amount of drift is greater than is believed to be probable. Hence, we conclude that the data strongly suggests a true peak. Furthermore, in the 68-day-old sample, the 620-kev peak persists and is much more pronounced, because of the decay of the activities formerly found on either side of it.

Figure 53 is the spectrum of sample S_{11} -1A 136 days after collection. The energy of the central, most prominent peak (neglecting for the moment the strong low energy peak due to Ce^{144}) is read from the original plot at 630 kev--in good agreement with the energy assigned from the former figure. In view of fission yields and half-lives, the peak seems to be that of Cs^{137} .

Stability and resolution of the analyzer is further illustrated by Fig. 54, in which the net difference between Sample N-1 at 68 days and Sample N-III, collected at a later date from 6 inches below grade level, is plotted. The $\text{Ru-Rh}^{103,106}$, Cs^{137} , and Zr-Nb^{95} peaks are clearly separated. The slight hump visible in Fig. 53 on the high energy side of the Ce^{144} peak appears well resolved also; this peak has not been identified as yet.

Fig. 52

Fig. 53

Fig. 54

Comparison of the Background Counting Rates of Two NaI Crystals of Different Size in Lead Shielded Facility

In the operation and development of a low-level counting facility such as our whole-body gamma spectrometer, a great many determinations must be made on relatively small volume samples, where the available space and flexibility of the Iron Room is superfluous. Examples of such analysis, selected from recent experience, include silicone "O" rings, magnesium oxide powder, steel and plastic parts (all parts of crystal canning), filter papers, and bone and tissue samples. Such determinations may be made in a smaller shield, such as described above,

Fig. 55

if the workload requires it, but for greatest convenience in comparing results, the background counting rate and energy spectrum should be the same as those of the larger facility which it supplements. Figure 55 compares the background spectrum of our 5-inch diameter by 4-inch crystal, in electrolytic copper, as observed in the Iron Room and in the lead-filled shield. There is excellent agreement in the two curves above 300 kilovolts, with a difference in total counts of just two

counts per minute. The backscatter peak, at approximately 200 kilovolts, is seen to be more prominent in the lead shield. Counting rate over the whole energy span from 30 kilovolts to 1.54 Mev is 569 cpm in the shield versus 513 cpm in the Iron Room. This 10% difference is too great to be attributable to the slight difference in wall thickness. It has been conclusively shown by Miller⁽²⁾ that, provided the shield thickness exceeds 6 inches of iron, or its equivalent of proven nonradioactive material, any further major reduction in background must be accomplished within the shield by choice of "radioactivity-free" crystal, phototube and associated "hardware." In view of the fact that the same tube and crystal were used throughout this experiment, also that the counting rate and energy spectrum are so nearly identical above 330 kilovolts, the observed difference in counting rate at low energies is most likely caused by

contamination of the "virgin" lead lining the shield It should not be difficult to verify this experimentally *

Intercomparison of data would also be made easier if identical crystals were used, but this may not be economically feasible. The dependence of background energy spectrum on crystal shape and size and our understanding of the relative contributions from activity within the crystal, phototube and housing, shield materials, and from the penetrating components of cosmic origin are all so inexact that theoretical analysis appears futile. The background spectra of two crystals of different size have been compared, all other factors remaining constant, and an attempt made to establish an empirical relationship between counting rate per energy band and crystal size. Although the crystals employed were too similar to permit extensive extrapolation, certain trends are evident, and it is hoped that this may be of some value to others engaged in low level determinations. The data permit an estimate to be made of the lower limit to the counting rate expected within a thick radiation shield.

The crystals compared, size 5 inch diameter by 4-inch and 4-inch by 4-inch[†] were grown and canned by the Harshaw Chemical Company. Both crystals were from ingots containing between 1.5 and 3 parts per million of potassium (3). Thus the background contribution from the K^{40} beta rays should not exceed 23 cpm and 15 cpm, respectively, or in the order of 1 count per minute per 100 kev band (2). This was considered small enough to be neglected. The counting rate per 100 kev energy band, as observed in the lead filled shield is tabulated in Table 48.

At low energies, the efficiency of the crystal is very nearly unity, and it would seem to be a reasonable approximation that the number of gamma-ray pulses observed should vary with exposed surface area of the crystal. The phototube effectively shields one end from scattered radiation from the shield, but introduces a fairly significant source of radiation, namely, the potassium in the tube envelope. Hence, whether one considers one or both of the flat surfaces as contributing to the interception of gamma-rays depends upon the relative importance of the phototube contribution. If only one end face is considered, the surface area is $\pi r(2l + r)$ (r = radius, l = length); for both surfaces it is $2\pi r(1 + r)$. For the two crystals being compared, the ratios in each

*Note in proof. Further counts have verified that the lead is indeed at fault. An extra 1/16-inch of lead, known to be extremely "clean," lowered the total count over the stated energy band to 480 cpm.

[†]We are indebted to Dr. Robley Evans and Mr. J. L. Bear, Dept. of Physics, Massachusetts Institute of Technology, for use of the 4-inch by 4-inch crystal.

Table 48

Background counting rates in lead-filled shield

Energy	Crystal size		Ratio
	5" x 4"	4" x 4"	
	Counts per minute		
50 - 150 kev	119.1	87.0	1.37
- 250	93.6	73.4	1.28
- 350	73.6	59.6	1.23
- 450	52.2	42.6	1.22
- 550	50.6	37.6	1.34
- 650	33.2	25.5	1.30
- 750	25.2	19.4	1.30
- 850	21.8	16.3	1.34
- 950	17.0	13.1	1.30
- 1.05 Mev	14.3	11.0	1.30
- 1.15	11.6	9.4	1.23
- 1.25	9.6	7.3	1.31
- 1.35	8.6	7.1	1.21
- 1.45	10.2	9.4	1.08
- 1.55	9.0	6.5	1.39
	546.6	425.2	1.26 (mean)

case are 1.31 and 1.35; hence agreement with experiment is satisfactory, regardless of the assumption. We note also that the ratio of radii is 1.25; and of the volumes, 1.56. The ratio is seen to be surprisingly constant, and very nearly the ratio of the radii.

Attempts have been made to justify this constancy on the basis of known efficiency of NaI vs thickness⁽⁴⁾ for the case of a uniform parallel beam, fitting some empirical geometric function of radius and length to the data. Similar Monte Carlo calculations for the case of uniform flux per unit solid angle would be useful. For the present, we must consider the observed data due perhaps to a particularly happy choice of crystal sizes without satisfactory theoretical basis. Further attempts to find a more general relationship are planned.

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METEOROLOGY

I. SOIL TEMPERATURE STUDIES

J. E. Carson* and H. Moses

A. Introduction

The space and time distribution of temperature and heat flux in the upper soil layers are subjects of considerable importance to many types of activities. The meteorologist, ecologist, farmer, soil scientist, civil engineer, heating engineer, military commander, and many others are interested in the thermal conditions of the soil.

Our atmosphere is capable of absorbing a relatively small amount of the short-wave radiation from the sun but is almost opaque to the long-wave radiation emitted by the earth's surface. This opacity to the long-wave radiation results in higher earth and air temperatures. The atmospheric property of being relatively transparent for short-wave radiation and opaque for long-wave radiation is known as the greenhouse effect.

Although the ultimate source of energy received by the atmosphere is the sun, most of the energy enters the atmosphere by way of radiation and conduction of heat from the earth's surface plus the release of latent heat of water evaporated. Since radiation from the soil surface has such a marked influence on the energy transmitted to the atmosphere, information on the soil temperatures and heat flux is of utmost importance.

The uppermost layer of the earth acts as a large heat reservoir, absorbing some of the excess heat of midday and summer and returning this heat to the atmosphere during the night and in winter. The heat stored acts as a climatic equalizer that helps to lessen temperature extremes. The thermal properties of the soil such as conductivity, specific heat, density, moisture content, water and water vapor movement within the soil will determine the quantity of heat that can be stored and the rate at which it can be absorbed or released.

It has been found that simple heat flow relations such as Fourier's equations do not apply to the soil. These equations assume that heat is transported by conduction only and that the medium is homogeneous in time and space; none of these conditions is found in the ground.

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The changes of soil heat flux and temperature at various depths are a function of both the thermal properties of the soil and the complicated interaction of a number of processes. The processes affecting the thermal structure are: absorption of short wave solar radiation and long wave radiation from clouds and the atmosphere, heat conduction between soil and air, evaporation of soil moisture, deposition of dew and frost, precipitation and the heat of fusion of soil moisture.

Some of these variables are easy to measure; others offer considerable difficulty. Among the difficult ones to measure accurately is soil moisture, a variable which strongly affects the thermal properties of the soil. The amount of moisture in the upper layers of the soil shows a daily cycle (evaporation during the day and condensation at night), an annual cycle (greater soil moisture in winter and spring), and a random variation due to alternating periods of precipitation and drying.

B. Instrumentation

The program of taking soil temperature observations (currently in progress) was begun by the Meteorology Group in November 1952. Soil temperatures are measured by means of 100-ohm copper resistance thermometers (Thermohms, manufactured by the Leeds and Northrup Company) at the following depths below the surface: 1 cm, 10 cm, 20 cm, 50 cm, 100 cm, 10 feet and 29 feet. These depths include all levels recommended by the World Meteorological Organization. The sensing elements are located about 240 feet east southeast of the Meteorology Building. They are under a grass surface cover which is completely open to the sun and sky. Throughout the growing season the grass is kept mowed. Air temperatures at Argonne are observed continuously at 5.5 feet above the ground by means of a ventilated thermopile. Details of the observational procedures for measuring the meteorological variables may be found elsewhere.⁽¹⁾

The soil temperature data are recorded both on a strip chart and on a five-channel paper tape in a programmed sequence (see Section III). Readings are made hourly from the strip chart for the top four levels, every fourth hour for the 100-cm level, and once per day for the lowest two levels. Since the diurnal temperature wave is damped out before reaching the 100-cm level no error is introduced by this procedure.

An extensive search of literature shows that the soil-temperature information collected at Argonne is unique since hourly values combined with a rather large number of other weather elements are recorded simultaneously. Most previous observations of soil temperature were undertaken as part of other research activities, such as agricultural experiments and were usually made once or twice per day at only a few levels. In order to study the diurnal temperature cycle in soil, it is necessary to observe the soil temperature at hourly intervals at several depths. Hourly values are

expensive to observe and cumbersome to reduce. The work has been considerably facilitated by the use of IBM punch card techniques with the help of Mr. Roy A. Kucera and Miss Dorothy Maren of the Accounting Department.

C. Soil Temperature Conditions at the Argonne National Laboratory

1. Average Annual Soil Temperature. The average annual soil temperature for each of the seven levels for the calendar years, 1953, 1954 and 1955 are shown in Fig. 56. Also shown are the 3-year means.

Fig. 56. Annual average temperature profile.

The scatter of points for the soil temperature at any level indicates the year-to-year variability. This variability is a result of the year-to-year changes in such factors as net radiation, soil moisture, and the advection of air masses of different temperature. The vertical temperature gradient implies that there is a net transport of heat into the deeper soil layers. Since there are other heat transfer processes operating, such as the percolation of rain water, the conclusion that there is a net transfer downwards may not be justified.

The average annual soil temperature at all levels is warmer than that of the air 5.5 feet above the ground. This indicates that there is a net transport of heat from the soil to the atmosphere during the year.

2. The Annual Cycle of Soil Temperature. The annual progression of soil temperature is shown in three sets of charts. In the first set (Fig. 57) are plotted the daily average soil temperatures for each of the seven levels studied for the calendar year 1955. Twenty-four hourly observations are used in computing each daily average. In the second set of figures, Fig. 58 and 58b, is shown the march of soil temperature for the three-year period from January 1953 through December 1955. These are based on decade averages. The decade is defined as follows: the first decade of any month consists of the period from the first through the tenth day; the second decade, from the eleventh through the twentieth day; and the third, from the twenty-first day to the end of the month. In the third set of charts, (59a and 59b) is shown the variation of soil temperature based on monthly averages.

Fig. 57. Average daily soil temperature 1955

Fig. 58a. Annual progression of soil temperature at indicated levels based on 10-day averages

Fig. 58b. Annual progression of soil temperature at indicated levels below surface based on 10-day averages

a. The Daily Average Soil Temperature Charts. The response of soil temperature to individual weather situations is very pronounced at the 1-cm, 10-cm, and 20-cm levels, and is still evident at the 50-cm level. Only extended periods of unseasonable temperature deviate the annual progression at 100 cm.

During January and February, the data show little day-to-day variability of soil temperature in the upper layers of the soil. Air temperature data indicate that such a quiet period is not observed in the atmosphere. This lack of variation in the soil is due to the very large heat of fusion of water, 80 calories per gram. Such a stable period near freezing has been called the "zero curtain" by Sumgin.⁽²⁾ The data at 10 cm and 20 cm show little decrease in amplitude or phase shift behind the 1-cm level. The expected change of phase in amplitude becomes apparent at 50 cm and below. At the 10-foot level, the warmest period is mid-September; the coolest, mid-April; and the amplitude of the temperature cycle has been reduced to about 13° F. At 1 cm, the amplitude is 59° F, and at 10 cm it is 57° F. At 29 feet, there is a very marked phase shift; the warmest temperatures are observed in February and the coldest in August. The annual range of temperatures has fallen to only 2.8° F.

b. The Decade Average Charts. It is evident from these charts that the fluctuations due to changes in air mass or precipitation falling into the soil are not smoothed out by the decade averaging technique. It is obvious from Fig. 58b that only at 10 feet and 29 feet are the curves nearly sinusoidal. The charts also show little phase shift and amplitude decrease from the 1- to 20-cm levels. Below this depth, the phase shift and amplitude decrease becomes marked.

c. The Monthly Average Charts. These charts (Fig. 59a and b) are based on three-year averages for the period January 1953 through December 1955. Due to the averaging process the data become quite smooth. There again it may be pointed out that the phase shift down to 20 cm is relatively small, and the amplitude decrease is also quite small; however, it is measurable. The data below 20 cm show the classical amplitude and the phase shift changes with depth as the time of year changes.

3. Time-Depth Representative of Isotherms. In this chart (Fig. 60) the data have been plotted on linear depth versus time scale. It was necessary to break the scale between 10 and 29 feet to conserve space. The numbers plotted represent the decade averages of soil temperature. Each number represents a three-year average of the soil temperature for the decade indicated. The sloping dashed lines indicate the time of maximum and minimum soil temperatures at the various depths. These lines show that the lag increases with depth. Since heat is conducted from warm to cool regions, the direction of heat flux at various times of the year can be

Fig. 59a. Annual progression of soil temperature based on 3-year averages for each month
Jan. 1953 - Dec. 1955

Fig. 59b. Annual progression of soil temperature based on 3-year averages for each month
Jan. 1953 - Dec. 1955

Fig. 60. Isotherms in time-depth plane. Dashed lines connect maximum or minimum values.
Based on 10-day averages of data obtained during period January 1953 - December 1955.

inferred from these charts. It is also of interest to note that in the spring and fall the orientation of the isotherms is nearly vertical which of course means that the heat flux during these seasons is at a minimum. The sharpest gradients occur during the warmest and coldest parts of the year.

4. Average Hourly Soil Temperatures. In Fig. 61 are shown the average hourly air and soil temperatures for January, April, July and October of the year 1954. These months are representative of the four seasons.

During January, there is relatively little variation of soil temperature at the 1-cm level. This is the coldest month at the 1-cm and 10-cm levels in the soil and also in air at 5.5 feet above the ground. At 20 cm and below, the diurnal cycle is fairly well damped out. The very small daily range of soil temperature is partly due to the result of freezing and thawing of water in the snow and partly due to the blanketing effect of a snow cover when present. As shown in the vertical temperature gradients, the heat stored in the soil is now returning to the atmosphere.

April is a typical transitional month; the temperatures of the air and of the upper layers of the soil are rising rapidly. At the 10-foot level, the temperature has just reached its lowest level while at 29 feet, the temperature is falling. The vertical temperature gradients are quite small. The diurnal wave is pronounced at the 1-, 10- and 20-cm levels but is almost damped out at the 50-cm depth and is not found at the 100-cm level.

July, usually the warmest month of the year, is a period in which the heat flux is at a maximum. The data indicate that the 1-cm temperature is considerably warmer than the air at night and around noon. The air-temperature curve has a flatter maximum than the 1-cm soil temperature curve. This is the result of the intense turbulent transport of heat upward in the atmosphere during the day. At the 50-cm level the daily cycle has been reversed, but it is undetectable at levels below this.

October is typical of the transition from summer to winter. The temperature is falling rapidly in the upper layers but is about static at the 10-foot level and is rising at 29-foot level.

One remarkable feature of these charts is that the soil at 1 cm is usually warmer than the air during the daily cycle; the maximum difference occurs at night. This indicates that the ground is giving up sensible heat to the atmosphere throughout the entire day during most months.

5. Air- Minus Soil-Temperatures. Figures 62a and 62b show the difference between the average hourly air and the 1-cm and the 10-cm soil temperatures by months for the year, 1955. The charts show that the

Fig. 61. Hourly average air temperature and soil temperatures at indicated levels.

Fig. 62a. Average hourly difference: air- minus 1-cm soil-temperature ($^{\circ}\text{F}$)
January - December 1955

Fig. 62b. Average hourly difference: air- minus 10-cm soil-temperature ($^{\circ}\text{F}$)
January - December 1955

soil is a heat source for the atmosphere both day and night during the winter months and during the night for the warmer part of the year. In all months but April 1955, the average daily temperature of soil is warmer than that of the air. This month was an extremely warm month - almost 9°F warmer than the average.

6. Additional Soil Temperature Studies. The reduction of the soil temperature data by IBM techniques for this period is nearly complete. It is planned to use multiple regression techniques to uncover the relationships between soil temperature and other meteorological parameters such as wind, sunshine and rainfall. Another phase of this analysis consists of applying numerical integration techniques to see if an estimate of the soil moisture can be determined from the soil temperature data alone. This work is being done in cooperation with Dr. E. H. Bareiss and Dr. E. M. Keberle of the Applied Mathematics Division.

METEOROLOGY

II. CONCENTRATION OF ATMOSPHERIC RADON

H. Moses and G. A. Zerbe

A. Introduction

Measurements of atmospheric radon used in these studies were made under the direction of A. F. Stehney and H. F. Lucas. The technique used for obtaining the radon concentration measurements is described in a previous semiannual report.⁽³⁾ The studies described in this report are based on observations which began in August 1956 and have been continued through November 1957. Radon samples were obtained daily during the workweek, Monday through Friday, for the three eight-hour periods: 0045-0845, 0845-1645, 1645-0045.

B. Seasonal Effects

Concentrations of atmospheric radon appear to undergo an annual cycle. This is indicated by the data presented in the curves of Fig. 63a-c. In preparing these curves, the data were separated into three groups corresponding to the three eight-hour measuring periods. These graphs show the monthly average values of radon concentration and the monthly maxima and minima.

In the nighttime and early morning hours, the period from 0045 to 0845, the radon concentrations are higher than those in the remaining two periods. Figure 63a shows that the highest mean value for this eight-hour period occurs in September, and the lowest monthly average occurs in April. The monthly range of radon concentration (monthly maximum minus monthly minimum) is also largest in the fall months and smallest in the spring. The relatively high concentrations found in the late summer and fall months may be attributed to the dryness of the ground, the relatively strong inversions, and light winds. During the spring the ground is generally wet, thus inhibiting the release of radon. The winds are relatively strong, thus contributing to rapid dispersion.

In the second eight-hour measuring period covering the daylight hours, all three curves -- the maximum, mean, and minimum -- exhibit lower values than in the previous chart. The monthly averages are lowest in May and highest in August.

The curves for the late afternoon and evening show values approximately intermediate to those of the other two eight-hourly periods. One might expect that the curve of monthly average radon concentration, when

Fig. 63 (a) to (c). Monthly maximum, mean, and minimum values of radon concentration in $\mu\mu\text{C/liter}$ for the period September 1956 - November 1957

averaged over a period of three to five years, would be a smooth sinusoidal curve similar to those of monthly average air temperature or monthly average soil temperature. (See Section I) The curves such as those of Fig. 63a-c representing only a little over one year of data may be expected to deviate from a smooth curve due to random fluctuations in the large-scale weather patterns.

C. The Effect of Wet Ground

The amount of radon measured in the atmosphere over an eight-hour period is a function of a number of variables. Some of these are wetness of the soil, stability of the atmosphere, wind speed and direction, and atmospheric pressure. In this section, an attempt is made to show, qualitatively, the effects of wet ground upon the atmospheric radon concentration.

To study these effects, the radon measurements were separated into two categories. In the first were placed those measurements which were taken when no precipitation occurred within the eight-hour sampling period, or within any of the two preceding eight-hour periods. In the second category were placed those measurements which were taken when 0.10 inch or more precipitation fell within the sampling period or within any of the two preceding eight-hour periods. If precipitation occurred in amounts less than 0.10 inch, the data for the eight-hour period in which it occurred and those of the two subsequent eight-hour periods were not considered.

Relative frequency distribution of radon concentration for each of the three eight-hour sampling periods and for dry and wet ground conditions are presented in Fig. 64a-c. These graphs are extensions of studies described in the last semiannual report.⁽⁴⁾ From these graphs, it appears that the effect of wet ground in reducing the radon concentration is quite pronounced in the early morning period. During the daylight hours wet ground is not associated with lower radon concentrations since the two frequency distributions are very nearly the same. Radon concentrations for the period 1645-0045 are lower with wet ground than with dry, but the difference is less than for the eight-hour period 0045-0845.

The presentation of this section is somewhat biased since double the amount of data is used for the period from August 20, through November. Since the ground is relatively dry during this period, the effect of wet ground is greater than in such months as April or May. For this reason the wet-ground effect is somewhat magnified although the general qualitative picture is changed only little.

To determine the effect of meteorological variables such as atmospheric stability, wind speed, air pressure and rainfall etc. upon the concentration of atmospheric radon, a multiple regression study is now being carried out with the aid of the IBM computing group in the Accounting Department. This study should provide a more valid determination of the contribution of each of the elements to the concentration of radon than can be obtained by scatter diagrams or relative frequency distributions as presented above.

Fig. 64 (a) to (c)

Relative frequency distribution of radon concentration, August 1956 - May 1957. The ground was considered dry when no precipitation occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods. The ground was considered wet when precipitation of .10-inch or more occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods.

METEOROLOGY

III. INSTRUMENTATION

H. Moses and F. C. Kulhanek

A. Automatic Data Processing System

1. Introduction. The automatic data processing system for use with the soil temperature measurements began successful operation in October 1957. This equipment was purchased from the G. M Giannini and Co., Inc. In this report are described the broad specification of units which comprise the system now in operation. Figure 65 shows a schematic diagram of the components and how they fit together.

Fig. 65. Schematic diagram of automatic data processing system for soil temperature measurements.

2. The Basic Soil Temperature Measuring Equipment. As indicated in a previous report,⁽¹⁾ soil temperature measurements are made at eight different locations beneath the ground surface. These are at: 1, 10, 20, 50 and 100 cm and 10 and 29 ft and at 100 cm at a second location. The thermal elements are 100-ohm copper resistors known as Thermohms. A Leeds & Northrup Micromax wheatstone bridge recorder is used to

measure the resistance values thus providing a soil temperature measurement. The data are programmed by means of an Automatic Electric Type 45 stepping switch actuated by a motor-driven cam-operated auxiliary switch. In Fig. 65 the stepping switch is shown as the input scanner.

3. The Analog-to-Digital Converter. The analog-to-digital converter is a Giannini Type-100 disc encoder which is attached to the Micromax recorder shaft turning the slidewire. Its function is to convert analog values of soil temperature as represented by the angular shaft positions into digital values. The encoder provides counts from 0 to 599 in a modified decimal binary code. This encoder range corresponds to the two soil temperature ranges of the recorder, from 0 to 60°C and from -30 to 30°C. Readings to the nearest of 0.1°C may thus be obtained from the encoder. In the colder part of the year, when the lower range is used, 300 counts are subtracted electrically from the encoder to provide the correct soil temperature values.

4. The Translator and Tape Punch Units. The translator unit (Model K-106 manufactured by the G. M. Giannini & Co., Inc.,) consists of a complicated arrangement of relays, thyratrons, diodes and other electrical components. Its function is to translate the output of the encoder into impulses needed to actuate a Commercial Controls Model 2 paper tape punch. Five-channel paper tape similar to that of conventional teletypewriters is used.

5. Programmer and Timer. The function of the programmer, Giannini Type PC-105 is to switch the necessary circuits for providing the information desired on the tape. This information consists of tape identifying codes, date and time information, the soil temperature readings, a reference reading, and other codes such as space and end-of-line (carriage return) symbols.

There are actually two synchronous motor-driven timers in this system. The first one is an Eagle Polypulse Timer which provides a short pulse (about 100 milliseconds) to activate the control unit. Upon receipt of this pulse the tape punches the information as set up by the programmer, encoder and translating unit. Such a pulse is received every two minutes beginning at one minute past the hour and ending at seventeen minutes past the hour. The second timer controls the hour and date. It actuates stepping switches to provide the correct punching of the date and time a few minutes before the sequence of soil temperature readings begins.

6. The Data Processing Panel. Figure 66 shows an external view of the Data Processing Unit. It is composed of 5 slide-in chassis. The uppermost panel contains the digital clock showing the day of the month and the hour. A selector switch for setting the number of days in the month is also mounted on this panel. The five buttons below the date-hour numbers are reset buttons. Power to this section is controlled by the toggle switch on the extreme right.

Fig. 66. Console and tape punch
of soil temperature data
processing unit.

The second panel contains manual rotary switches for inserting the year and month. Two digits are used for each parameter. The last two selector knobs on the right are unused at the present time.

The middle of the panel rack houses the programmer. This panel contains a reset button to set the cycle to the beginning point. This is used in testing or in case of a power failure. When the "record" button is pushed a pulse is sent to the translator unit which initiates the punching of the information currently contained in the encoder and programming units. The toggle switch on the extreme left is a power on-off switch.

The remaining two chassis are those of the translator. Only a "record" button and a power switch are brought out in this unit. The "record" button is a duplicate of the "record" switch on the programmer chassis.

The Commercial Controls Model 2 Tape Punch is located on top of the cabinet housing the data processing unit. The tapes provided by this unit are sent monthly to the IBM Service Bureau for transcription to cards.

Work is continuing on the problem of converting the remainder of the meteorological measuring system to automatic recording on paper tape. The application of automatic techniques to some of the other meteorological parameters will be more difficult since the values are rapidly fluctuating; this necessitates averaging or integration. Another difficulty is presented by the fact that the voltage output of some of the meteorological sensor elements is not linearly related to the value of the parameter measured.

The use of automatic data processing equipment together with the availability of the automatic computing facilities such as the IBM 650 or 704 and the Argonne National Laboratory computer GEORGE, allows us to do more in less time. Once the automatic system is completed we shall have at our disposal improved capabilities for better service to the Laboratory and greater contributions to the field of micrometeorology.

B. Installation for Measuring Atmospheric Moisture

1. Introduction. Information on the distribution of moisture in the lower layers of the atmosphere is not only of importance for engineering design and other scientific work at the Laboratory but also is of value in determining the diffusive capacity of the atmosphere. The measurement of atmospheric moisture content at several different elevations allows the determination of the eddy moisture flux which is strongly influenced by the intensity of turbulent mixing. A comparison of moisture flux with heat, momentum, and radon fluxes will provide useful information, now unavailable, on the relations among the mixing or exchange coefficients for each of these quantities.

2. Location of Moisture Measuring Elements. Moisture-measuring equipment, using the Dewcel as the sensing element, was purchased during this six-month period. The installation of this equipment is nearly complete for five locations above the ground: 5.5, 19, 37, 75 and approximately 120 ft. The last four locations are on the Meteorology Tower, and the lowest one is underneath the Meteorology Tower. At the present moment the exact elevation of the uppermost Dewcel, mounted on the tower cabin, has not been measured. The Dewcel element is protected from rain, dust, and other undesirable atmospheric elements by a weather hood. A photograph showing the Dewcel as mounted with the weather hood on the 19-ft level of the Meteorology Tower is shown in Fig. 67.

3. Description of the Dewcel. The Dewcel, manufactured by the Foxboro Company, consists of a cylindrical tube about 4" long and about 1/4" in diameter. The tube is covered by a wick of woven glass tape upon which gold wires are wound in bifilar fashion. The tape is impregnated with lithium chloride and the gold wires are connected to a 25 volt A.C. power source.

Fig. 67. Mounting of Dewcel and weather hood at 19-foot level on meteorology tower.

The operation of the Dewcel depends upon the fact that the conductivity between the parallel gold wires is a function of the amount of moisture absorbed by the hygroscopic lithium chloride. If the vapor pressure of the ambient atmosphere is greater than that in the immediate vicinity of the film of lithium chloride and water, water will be absorbed by the lithium chloride. As this occurs, the conductivity between the gold wires will increase, thus allowing more current to flow and raising the temperature of the Dewcel element. This will continue until the equilibrium vapor pressure about the lithium chloride-water film is equal to that of the ambient atmosphere. If the reverse is true, i.e., the vapor pressure about the film exceeds that of the atmosphere, the vapor pressure flux will be in the opposite direction, and water will be given up with the result that the conductivity will decrease and the temperature of the element will fall. The Dewcel thus acts as an automatic self-regulating control to vary the current flow between the gold heating wires as required to maintain an equilibrium temperature which is a measure

of the ambient absolute humidity. A resistance thermometer, located inside of the cylindrical tube, measures the equilibrium temperature of the Dewcel elements. The electrical resistance of this thermometer is recorded on a Brown Electronik Potentiometer which is calibrated to read the value of the ambient dewpoint directly. The recorder has been calibrated to read dew points from -30 to 90°F .

The electrical circuitry used with the Dewcel and resistance thermometer is similar to that used with the Thermohms in the measurement of soil temperature. Figure 68 shows a schematic diagram of the Dewcel installation. In this figure may be seen the power supply for heating the bifilar gold windings, the connecting cables between the Dewcel elements, the programmer, and the recorder. The programmer is an Automatic Electric Type 45 stepping switch controlled by a synchronous timing motor.

Fig. 68. Schematic circuit diagram of moisture measuring equipment using Dewcels

4. The Dew Point Chart Trace. In recording the data on the Brown strip chart, the following pattern was adopted. Two-minute dew-point readings are made for successively higher levels beginning with

the 5.5 ft level. After the dewpoint at the top level has been recorded, a reference resistor is placed in the circuit for two minutes. This reference resistor has the function of separating one atmospheric sounding from the next, and also provides an indication of the recorder's accuracy. Actually, two reference resistors will be used. At the end of one series of readings, a reference resistor equivalent to the temperature of -20°F will be inserted into the circuit. After the completion of the second series of readings, another reference resistor equivalent to a reading of 80°F will be used. This arrangement facilitates the reading of the recorder charts.

Measurements of dewpoint at the 5.5 ft level were begun in December 1957. The measurements at the other levels will begin as soon as calibrations are completed. It is expected that the system will go into operation very shortly.

METEOROLOGY

IV MODEL SMOKE DIFFUSION EXPERIMENTS

G. H. Strom* and H. Moses

A. Introduction

Experiments to produce, on a reduced scale, diffusion phenomena occurring over the Argonne Site were carried out in the New York University Wind Tunnel during this six-month period. The objective of these experiments is to investigate the capability of the wind tunnel validly to model those atmospheric phenomena which lead to the observed dispersal patterns resulting from the emission of stack gases. Satisfactory progress was made in adapting the instrumentation and developing the techniques for carrying out this work.

B. The Wind Tunnel

The Meteorological Wind Tunnel at the New York University, College of Engineering, has a working section which is approximately forty feet long, three and a half feet high, and seven feet wide. One side of the test section contains glass panels for visual and photographic observations of the experiments. Wind speed in the test section is accurately controlled over a range from 0 to 15 ft./second.

Thermal conditions within the tunnel are controlled by floor and ceiling heating units and by a harp of electric heating wires arranged in a vertical plane perpendicular to the tunnel axis. The electric heating wires, spaced ten per inch, are stretched horizontally across the airstream at the entrance to the test section. The electric current to individual wires may be controlled to produce a vertical gradient of heat, and thereby control the temperature gradient of the airstream as it enters the test section.

The velocity profile at the test section entrance may be controlled with properly spaced strings stretched horizontally across the airstream. The velocity profile is modified as the air moves along the test section by the roughness of the floor, which is controllable, and the obstructions to air flow formed by the model itself.

For the production of smoke, air is metered through a Flowrater and passed through a metal chamber containing oil vapor. This oil vapor is produced by electrically heated vaporizers. Helium is also metered through a Flowrater before being added to the air-smoke mixture in the tube leading to the model.

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The addition of helium is used to reduce the density of the model gas effluent. Previous tests at the New York University Wind Tunnel have shown that this means of reducing the stack gas density is a permissible substitution for the application of heat.

C. Theoretical Considerations

In previous work on modelling atmospheric diffusion problems, it was found that useful results may be obtained if the model is geometrically similar to the full scale case, and the following relations are maintained:

$$\left(\frac{\rho s}{\rho}\right)_M = \left(\frac{\rho s}{\rho}\right)_F$$

$$\left(\frac{Vs}{V}\right)_M = \left(\frac{Vs}{V}\right)_F$$

and $\left(\frac{V^2}{gd}\right)_M = \left(\frac{V^2}{gd}\right)_F$, Froude's number

The subscripts M and F refer to the model and full scale respectively.

ρs = mass density of stack gas
 ρ = mass density of ambient air
 Vs = stack gas ejection speed
 V = wind speed
 d = stack diameter
 g = acceleration due to gravity

In order to achieve complete dynamic similitude, other relations would have to be observed such as the Reynolds number $\left(\frac{\rho V d}{\mu}\right)$, where μ is the dynamic viscosity) and some parameter involving a buoyancy term which would be dependent on the thermodynamic stability of the ambient fluid.

In cases where models have sharp edges, it can be established that the flow characteristics are unaffected by Reynolds numbers over the range of flow velocities used. Although the stack model used in these experiments is cylindrical, it is believed that the viscous effects as indicated by Reynolds numbers are still of second order in importance. However, this will have to be firmly established before these experiments are completed.

Previous work with wind tunnels also leaves no doubt that the thermal structure of the ambient fluid exerts a marked influence on the flow characteristics and consequently, the diffusion rate of the stack effluent.

These experiments are designed to measure carefully these variables in the wind tunnel. One of the aims of these experiments is to learn how to scale the thermal structure in models validly by comparing the vertical temperature gradients in the wind tunnel with those in the atmosphere.

D. The Model Experiments

A scale factor of 1/8 inch equals 1 foot or 1:96, was adopted in the model of the Meteorology Stack shown in Figs. 69b and 70b. Models were also made of the Meteorology Building, 181, and the Meteorology Tower, but these are not shown in these photographs.

Two Crown Graphic cameras were used to photograph the smoke plume. The first camera, taking a side view of the smoke plume, was located 43.12 inches from the glass paneling and 17.34 inches above the floor of the wind tunnel. The other camera, which took a plan view of the smoke from the top, was located 10.96 inches downwind of the stack and 34.81 inches above the floor. The positions of the cameras with respect to the stack and with respect to stations measuring air speed and temperature may be seen in Figs. 71a and b. The numbers in parentheses beside the words Camera XZ or Camera XY refer to the focal length. A right-handed Cartesian coordinate system was used; X was taken downwind, Y crosswind, and Z upward.

In making these smoke photographs, the lenses were held open continually. A speed photoflash unit was used to flash a Sunflash-V8 Flash tube, manufactured by the Kemlight Laboratories, Chicago, Illinois. The circuitry was arranged to provide a light flash of about 4 microsecond duration at intervals of every 1.5 sec.

Since Froude's Number $\frac{V^2}{gd}$ is held constant for the model and full scale and g remains the same, it is easy to show that $T_m = T_f \sqrt{K}$ where T_m and T_f represent time in the model and full scale and K is the length ratio of geometric scaling factor. Since the geometric scale factor is assumed to 1/8 in. = 1 ft. or 1/96, $T_m = T_f \sqrt{1/96} = 0.102 T_f$.

In the series of experiments made with the Experimental Stack at the Argonne National Laboratory, with which the tunnel measurements are to be compared, 16 photographic exposures of a smoke plume were made on one film, 15 seconds apart. It is therefore evident that to maintain the same Froude's Number, this time interval would have to be multiplied by a factor of 0.102. Sixteen exposures of the model plume were therefore taken 1.5 seconds apart during each run.

Fig. 69. Comparison of multiple exposure smoke plumes in atmosphere and wind tunnel -- stable atmospheric conditions

- a. Actual multiple exposure smoke plume at Argonne site.
- b. Model multiple exposure smoke plume at NYU wind tunnel.

Fig. 70. Comparison of multiple exposure smoke plumes in atmosphere and wind tunnel -- unstable atmospheric conditions

COURTESY OF N.Y.U. 3 1/2 x 7 FT WIND TUNNEL
12-1-57 E J K

Fig. 71a. Detailed tunnel layout for smoke diffusion experiments

COURTESY OF NYU 3-1/2 X 7 FT WIND TUNNEL
12-1-57 EJK

Fig. 71b. Tunnel layout for model smoke diffusion experiments.

In Figs. 69 and 70 are shown samples of smoke plumes photographed in full scale and in the wind tunnel. These are multiple exposure photographs consisting of sixteen exposures, and have been adjusted to equivalent length scales. Note that the stack heights are the same in both sets of pictures. It is apparent that although the eddy structure is different, the rate of plume widening is comparable.

There are still a number of experimental difficulties which have to be overcome before valid comparisons can be made between the model and full scale. Some of these are:

(1) It is likely that the camera inside the tunnel taking a plan view of the plume will have to be moved outside of the tunnel. This will involve cutting a hole in the ceiling. The reasons for this change are to obtain a sharper image of the smoke plume and to assure a minimum of airflow interference.

(2) The tunnel lighting has been uneven since only one flash lamp was used. It is planned to arrange several such lamps in a line along the plume to obtain more uniform lighting.

(3) In some of the runs, it was observed that the plume showed rapid dispersion in spite of the presence of a stable lapse rate in the region from a few inches above the floor to above the plume. In these cases, however, the floor temperature was somewhat warmer than that of the air in the region of the plume. It is believed that the rapid dispersion was caused by a mass overturning of sections of the airstream, since the airstream showed long-term temperature fluctuations. To eliminate this difficulty, additional temperature control equipment was installed.

(4) An examination of the airstream speed profiles in the tunnel showed that they differed in a crosswind direction in some cases, and in others deviations from the expected configuration occurred, i.e., velocity minima and maxima occurred at intermediate elevations indicating a disturbance. These unnatural profiles may arise from pressure differentials created at the heating units. Steps are being taken to eliminate these difficulties.

A considerable amount of painstaking work is being done to measure accurately and control the temperature and wind profiles. It appears that the result of these efforts should provide valuable information on how to validly model the dispersion process.

METEOROLOGY

V. THE ARGONNE METEOROLOGICAL MODEL TOWING TANK

P. Frenzen

A. Introduction

A program of experiments designated to study the influence of gravitational stability upon turbulent diffusion utilizing the Argonne meteorological model towing tank has begun in the period covered by this report. Earlier publications in this series have summarized planning and tank construction phases of the project⁽⁵⁾ as well as preliminary equipment trials and development of methods of observation and analysis.⁽⁶⁾

To date, further modifications in the apparatus have reduced residual mean flow components and made it possible to photograph the turbulent motions of tracer particles for longer periods of time. In addition, photographic techniques have been improved, and digitized read-out equipment has been employed to advantage in extracting particle displacement data from enlarged photographs of the experiments. A successful manual computation of a Lagrangian autocorrelation curve for a selected set of experiments led to the preparation of a digital computer program for the routine performance of this calculation. By the end of this report period, the machine program had been completed and tested.

B. Apparatus

The difficulties with residual mean flow pulsations and with the installation of a moving upper lid designed to restrict the surface waves which produce them have been described in the previous report of this series. The form and magnitude of these flow variations are shown as the dashed curve in Fig. 72. Although they are consistent in similar experiments and can readily be determined and then subtracted from individual measurements to obtain the turbulent velocity fluctuations alone, it was still desired to reduce them since they were of the same order of magnitude as the turbulent components themselves. Further, smaller net mean flows would allow tracer particles to remain in the field of view for longer periods of time, increasing the lengths of trajectories which could be observed.

The three-eighths-inch bar, two-inch mesh grid used in these trials was originally mounted in a rectangular aluminum frame whose members presented only one-eighth-inch sections to the fluid as the grid advanced, but which measured three inches along the tank (cf. Fig. 47, p. 78, ANL-5755) Prof. D. Fultz (University of Chicago, Department of Meteorology and consultant to the Radiological Physics Division) suggested that the channels

Fig. 72. Observed residual mean flow variations

formed between these plates and the walls of the tank might be complicating the flow. A visual check with ink tracers revealed that this was indeed the case; large amounts of fluid were streaming back past the grid through these conveniently ducted passages. This not only accelerated the growth of the boundary layers at the tank walls, but it also allowed the grid proper to carry additional fluid forward in the central regions. The relatively large positive residual mean flow components shown in Fig. 72 resulted.

When a frame built of bars identical to those in the grid was substituted for the aluminum plate, an experiment with otherwise similar conditions produced the residual flow components plotted as the thin solid line in Fig. 72. The net positive drift was replaced by a net negative drift of about the same magnitude, this now representing, in the absence of the channels supplied by the plate frame, the slow return of fluid primarily carried forward by the boundary layer under the moving lid.

To reduce this backflow, the boundary layer was decreased by removing half the length of the lid. A recent measurement of the remaining mean flow components under this five-foot lid is plotted as the heavy solid line in Fig. 72. Camera difficulties cut off the run at the point indicated,

but the limited data obtained indicate that the net drift may have been largely eliminated. If further measurements bear this out, the principal limit to the length of particle trajectories that can be followed will be the capacity of the camera.

An indication of other difficulties that may arise, however, is given in the right half of the thin solid curve of Fig. 72. At $x/M = 60$, the end of the ten-foot lid passed over the test area; the next point, $x/M = 75$, shows the stern wave disturbance which resulted. Interestingly enough, a similar disturbance is not evident in the limited test data with the five-foot lid (after $x/M = 30$). If this result proves to be correct, it may be associated with the passage of the end of the lid during the maximum positive drift phase of the residual flow variation.

C. Data Reduction

Consecutive velocities of individual particles along their trajectories are now obtained by the following, largely standardized procedure. Thirty-five millimeter negatives, exposed at one-second intervals through the rotating shutter described in the previous report, are enlarged on 11" x 14" sheets of plasticized, shrink-resistant paper (Kodak Resisto-Rapid) using a nondiffusing type, 35mm enlarger (Leitz Focomat).

Consecutive prints are then viewed on a blink-comparator built for the purpose. The reflection of one print is seen superimposed on another through a forty-five degree, half-silvered mirror. By switching the separate lights illuminating the prints, the operator can readily determine which group of points on one photograph corresponds to the points representing the same particle on the preceding photograph. Switching the lights back and forth between photos gives a definite impression of motion to the successive images of the same tracer particle.

After numbering all suitable tracer images on photograph number one ($t = 1$), the operator then transfers these numbers with the aid of the comparator to the corresponding images of the same particles on photograph number two ($t = 2$); numbers from photograph two are then transferred to photograph three, and so on. New particles are assigned numbers as they appear, and the numbers of particles which disappear are dropped from the list.

The blink comparator was originally built as a substitute measure after the experimental prints were discovered to be too opaque to be compared on a light table, but it has since been found superior in several respects. One is the impression it gives of particle motion described above. Another is its ability to superimpose two images, as on a light table, but with the added feature that either image can be switched off at will.

Particle displacements are then taken from the prints with the aid of a digitized coordinate reader (Benson Lehner Oscillograph Analyzer and Reader, "OSCAR"). Since the time interval between the first and final images of every tracer particle on each photograph was only two-thirds of a second, the scale of the reader is adjusted (with reference to a known length appearing on the prints) to one-and-a-half times the scale in the experiment. In this manner the machine is made to print out the measured displacements as velocities in millimeters per second.

From photograph number one, velocities of all particles are extracted in their numbered order; from photograph number two, velocities of the same particles are taken in the same order, and so on. Blanks are inserted when previously observed particles are found to be missing, and new particles are added in the order of their appearance. The result in OSCAR's output is a matrix of observed velocities, u_{ij} , for "j" particles at "i" times.

D. Analysis

As described in the previous reports of this series, the continuous sampling of particle velocities along relatively long trajectories in these experiments has been attempted in order to study the Lagrangian autocorrelation function $R(\xi)$, the key parameter in G. I. Taylor's basic equation for turbulent diffusion, under varying conditions of fluid stability.

In Taylor's equation(7)

$$\overline{x^2} = 2\overline{u^2} \int_0^T \int_0^t R(\xi) d\xi dt$$

where $\overline{x^2}$ expresses the dispersion of the diffusing particles and $\overline{u^2}$ is a measure of the intensity of turbulence. The Lagrangian function $R(\xi)$ is the average correlation of the velocity of a particle at any instant with its velocity at an interval of time ξ later:

$$R(\xi) = \frac{\overline{u(t) u(t + \xi)}}{\sqrt{\overline{u(t)^2}} \sqrt{\overline{u(t + \xi)^2}}}$$

Since $R(\xi)$ has been considered difficult to evaluate directly, experimental studies of turbulent diffusion have sometimes used the indirect procedure suggested by Taylor in his original paper, that of calculating the function from a doubly differentiated form of his diffusion equation:

$$R(\xi) = \frac{1}{2\overline{u^2}} \frac{d^2}{dt^2} (\overline{x^2})$$

This has been done by Kalinske and Pien⁽⁸⁾ in a water channel, by Mickelsen⁽⁹⁾ in a wind tunnel, and by Hay and Pasquill⁽¹⁰⁾ in the atmosphere, to name only a few investigators, but the method is subject to considerable uncertainty since it depends upon a twice differentiated, empirical curve.

On several occasions the Lagrangian function has been directly evaluated in the free atmosphere; in one such study Edinger⁽¹¹⁾ tracked soap bubbles, while in another Gifford⁽¹²⁾ employed zero lift balloons. But this writer knows of only one laboratory experiment, that by Vanoni and Brooks⁽¹³⁾ in 1955, in which an attempt was made to evaluate the Lagrangian autocorrelation function by direct measurement of particle trajectories in the homogeneous, decaying field of turbulence behind a grid.

Working in the 2.7-meter long test section of a water tunnel, Vanoni and Brooks photographed particles during the time required for one hundred flashes to be emitted by a strobe unit operating at 60 to 180 cps. Though the trajectories they obtained were long enough to show $R(\xi)$ falling to zero in about half their runs, the authors recommended that such observations be extended over longer trajectories and times. In this respect, the Argonne towing channel affords a distinct advantage.

Recognizing the influence of turbulent decay on Lagrangian correlation measurements in the lee of a grid, Vanoni and Brooks kept the effect "reasonably uniform" by using only one velocity product from each of a number of trajectories, these taken at $+\xi_i/2$ and $-\xi_i/2$ from a central time common to all trajectories and to all $R(\xi)$ determinations. In the Argonne experiments the use of decay corrections originally suggested by G. K. Batchelor⁽¹⁴⁾ enables all of the velocity data taken from decaying, turbulent trajectories to be used in direct evaluations of $R(\xi)$ since they restrict the evaluation neither to one velocity pair per trajectory nor to only those velocities measured far from the grid where both decay rates and turbulence intensities are small.

The Batchelor corrections (there are two) are based upon the mass of wind tunnel experimental data which show that the velocity scale V of the turbulence behind a grid decays as $t^{-1/2}$, while the characteristic length scale L (say, average eddy diameter) increases as $t^{1/2}$. These combine to define a time scale $T = L/V$ which is seen to increase as the ratio $t^{1/2}/t^{-1/2} = t$. At the same time, it has been observed that the turbulence during these changes remains statistically similar; that is, curves representing statistical functions (e.g., power spectra) describing the field during the decay retain approximately the same shape.

Batchelor points out that by merely adjusting the scale of the energy axis and relocating the origin of the wave length (or frequency) axis, any series of such curves taken at different times in decaying turbulence can be made to resemble data from a statistically stationary field

of turbulence. He suggests that one may, 1) cancel the velocity decay by multiplying the observed values by $t^{1/2}$, and 2) compensate for the increasing time scale by dividing the experimental time intervals by t . This redefinition $d\tau = dt/t$ integrates to $\tau = \ln t/t_1$ where the arbitrary constant t_1 essentially defines the units.

The velocity correction⁽¹⁾ is self-explanatory; the same cannot be said for the time scale adjustment.⁽²⁾ However, both the need for and the results of this latter correction can be seen in the following Table 49. The data are from the same experiments used in the manual and machine test calculations to be described in a later section; the correlation results quoted are those obtained by the computer (16 Dec. 57, Type A.)

Table 49

Obs. times (t), (t+ ξ)	Actual $\Delta t = (\xi)$	Corrected $\Delta \tau$ $\ln(t+\xi) - \ln(t)$	Correlation $R(\xi)$	No. of velocity pairs correlated
1,2	1	.69	.40	36
2,3	1	.40	.57	26
3,4	1	.28	.78	31
4,5	1	.22	.60	15
5,6	1	.17	.82	19
6,7	1	.15	.81	21
7,8	1	.13	.93	21
8,9	1	.11	.81	21
9,10	1	.10	.87	21
10,11	1	.09	.93	21

Note that even though the first Batchelor correction, i.e. that for velocity decay, has been performed on the observed velocity values, and even though the actual time intervals between the members of each velocity pair remain the same, the correlations still trend toward higher values at later times in the experiment. They behave as though the time intervals between successive observations were shrinking; and indeed, the Batchelor time correction defines just such shrinking time intervals.

All of the correlations obtained from these data at constant real time intervals consistently show this trend toward higher values at later times. On the other hand, in the nine cases in which the logarithmically corrected time intervals repeat along the trajectories used ($\ln 10 - \ln 8 = \ln 5 - \ln 4 = .22$, etc.), four exhibit essentially constant correlations at later times, one oscillates around a constant trend, two rise, and two fall.

Within the limitations of the relatively small amount of data available (numbers of velocity pairs available for each point in these trends are of the same order as those in the last column of Table 49), the Batchelor time correction appears to be a step in the right direction.

E. Calculations

Decay-corrected data from three experiments carried out under almost identical conditions were selected for a trial evaluation of the Lagrangian autocorrelation function. Approximate values for a dozen points along the curve were calculated by a somewhat simplified procedure; the same denominator was used in every $R(\xi)$ quotient, namely a mean square fluctuation determined from all the corrected data rather than the product of the root mean squares of only the particular velocities involved in each $R(\xi)$ determination individually. The results of these manual calculations are plotted as triangles in Fig. 73, the figures beside each point indicating the number of velocity pairs employed in the calculation of that correlation value.

Fig. 73. The Lagrangian autocorrelation function determined from data corrected for decay (Exp. 1, 2, 3; 05 Jul 57)

The curve thus determined was considered quite promising; it falls smoothly to zero as required, perhaps not as asymptotically as might be wished, but in the regions of longer corrected time intervals fewer velocity pairs were available and consequently the results began to scatter.

Since such curves are desired for each experiment in the program, and since the labor involved in even simplified calculation with limited data is considerable, the problem has been programmed for Argonne's electronic digital computer "GEORGE." The basic machine program performs the following operations:

- 1) Accepts the u_{ij} velocity matrix output of OSCAR as input;
- 2) Averages all velocities with the same observation time i ;
- 3) Subtracts these means to obtain a u'_{ij} matrix of turbulent fluctuations;
- 4) Determines the standard deviation (root mean square) of the fluctuations at each observation time and prints these out for a separate determination of the rate of decay;
- 5) Multiplies each entry in the u'_{ij} matrix by the square root of the observation time, $i^{1/2}$, to obtain a velocity-decay corrected matrix, u''_{ij} ;
- 6) Calculates correlations $R(\xi)$ for every possible time interval ($i_1 - i_2$, etc.) and averages them over the trajectories, j ;
- 7) Prints out the correlation values $R(\xi)$ along with the corrected time intervals (ξ) to which they correspond.

Using the same data as those selected for the trial manual calculations, GEORGE computed fifty-five correlations, each an average determined from as many as thirty-six velocity pairs. When repeated time intervals are combined, forty-one points on the Lagrangian autocorrelation curve remain. These are plotted as circles in Fig. 73; the filled-in circles indicate what are perhaps more heavily weighted points, averages of those corresponding to the repeated time intervals.

During the development of its final form, several variations of the GEORGE program were evolved. These can be selected as desired by the addition of certain specific machine commands. In one, the averaging of step (2) above is omitted, and a list of separately determined mean velocities is supplied as input data. Another program variation prints out the number of velocity products used in calculating each $R(\xi)$ value. Both of these steps were employed in the calculation of the points in Fig. 73.

The computer results define a curve which appears somewhat more realistic in the regions of higher correlation than does the manually calculated curve; $R(\xi)$ does not immediately drop from unity but rather shows a slight tendency to preserve itself for a short time. (The point is not trivial, for the presence of a cusp would complicate Fourier transformation of the curve.) However, considerable scatter develops in the correlations due to the relatively small number of products involved in each determination. Encouraging is the fact that the weighted points, calculated from more data, lie consistently closer to the approximate curve shown.

It is expected therefore, that the use of larger input data matrices (made possible by the more recent refinements in data reduction described above) will improve all the points accordingly. Moreover, the reduction in residual mean flow components should make it possible to follow some trajectories almost twice as long as those used in these calculations. Thus it will be possible to assess more accurately the significance of the trend toward negative correlations visible in the curves of Fig. 73.

In the near future, the GEORGE program will be expanded to include a calculation of a least-squares fit of the Lagrangian correlation curve to some functional form yet to be decided, pending consideration of a number of correlation curve examples. Power spectra of the turbulence fields can then be obtained by Fourier transformation of the functional representation. These will be used to assess the influence of fluid stratification changes upon the relative energies of turbulent eddies of different sizes.

F. Conclusion

One of the most promising aspects of the Argonne model experiments is the very real possibility that they can be directly compared with studies conducted in the prototype atmosphere. Such an experiment would involve photographing and measuring the trajectories of neutral density balloons during periods of differing atmospheric stability, a program paralleling that of the current studies in the towing tank. Evaluation of the Lagrangian autocorrelation function would then proceed in much the same manner, except for the omission of the Batchelor decay correction steps. Indeed one of the variations of the GEORGE program provides for just such an omission for possible future evaluation of atmospheric data.

Incomplete studies by others (notably Hay and Pasquill, loc. cit.) indicate that the Lagrangian correlation is preserved in the atmosphere for long periods, perhaps a minute or more, whereas in the model it has fallen to zero after two seconds of corrected time. The significance of this observation in the model (distorted as it is in both time and length scales) relative to a similar measure of the "time to zero correlation" in the atmosphere remains to be evaluated. In other words, the appropriate scaling factor, if any, remains to be determined. However, it is the variations of these times in both model and prototype, measured as a function of the stability of the fluid, that will be of principal interest in this investigation.

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STAFF ACTIVITIES

Am. Phys. Soc., Boulder, Colorado. September 5-7, 1957. "Identification of I^{138} as a Delayed Neutron Precursor," by G. J. Perlow and A. F. Stehney.

Bioassay and Analytical Chemistry Meeting. St. Louis, Missouri. Nov. 7-8, 1957. "Low Level Detection and Identification of Radioactivity in Humans," by C. E. Miller.

Radiological Society of North America. Chicago, Illinois. November 17-22, 1957. "The Relevance of the Skeletal Burden of Thorotrast Patients to the Problem of Chronic Toxicity of Bone Seekers in Man," by L. D. Marinelli.

A. F. Stehney conducted a course in Chemistry-310 (Radiochemistry) October-December 1957 at the Downtown Center of the University of Chicago.