1

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TABLE OF CONTENTS

	<u>Page</u>
The absorbance shift of irradiated organic solvents. Part III I. B. Berlman and W. R. Anderson	5
A simple container for scintillating solutions I. B. Berlman and L. D. Marinelli	8
The concentration of radon in the atmosphere Part I - Chronological variations H. F. Lucas, Jr., F. I. Ilcewicz and A. F. Stehney	10
Part II - Comparison with meteorological data F. C. Kulhanek, G. A. Zerbe and H. Moses	13
The thorium ²²⁷ accident - final comparison between experimental results and calculated values of retention and elimination L. D. Marinelli and P. F. Gustafson	16
The human spectrometer Introduction - C. E. Miller	21
I. <u>In vivo</u> measurements C. E. Miller and L. D. Marinelli	22
II. Background studies with the human gamma-ray spectrometer C. E. Miller	30
III. Studies on photomultiplier tubes C. E. Miller and O. J. Steingraber	32
Progress report: Radium toxicity R. E. Rowland, J. H. Marshall and J. Jowsey	39
Positive ion emission: Progress on the mechanism H. A. Schultz	40
The distribution of cerium ¹⁴⁴ and thulium ¹⁷⁰ in the dog skeleton J. Jowsey, J. H. Marshall, and R. E. Rowland	46
Quantitative autoradiographic measurement of calcium ⁴⁵ concen- trations in cortical bone J. H. Marshall, R. E. Rowland and J. Jowsey	48
An analysis of the spectrum of Ba ¹³³ with a view to its possible use in the determination of the RBE of alpha particles in the production of bone cancer J. H. Marshall	63

3

TABLE OF CONTENTS

	<u>Page</u>
Studies on background	
H. May	69
Drift velocity of electrons in gases	
J. C. Bowe	74
Meteorology	
The multiple exposure technique for studying atmospheric diffusion	
G. H. Strom and H. Moses	80
Diffusion studies conducted by means of the type K-24 aerial cameras	
Peter E. Wasko	82
Relationship between visual appearance and concentrations of smoke plumes	
F. I. Badgley and H. Moses	85
Fluctuations in temperature	
F. I. Badgley and H. Moses	88
Climatology	
H. Moses	91
Automatic data processing system	
H. Moses	94
Argonne meteorological model towing tank	
P. Frenzen	95
Soil temperature studies	
H. Moses	100
Publications	103
Staff activities	105

THE ABSORBANCE SHIFT OF IRRADIATED ORGANIC SOLVENTS. PART III*

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

In analyzing the absorbance curves of xylene for various radiation doses (see Fig. 1, Part II)* it is noted that the absorbance increase at a specified wavelength is proportional to total dose for doses equal to or less than 10^6 r, but for higher doses the increase is relatively smaller. Figure 1 illustrates this fact more clearly. In this figure are indicated the absorbance changes in xylene induced by various doses, plotted as ratios to the absorbance produced by 10^6 r in the region from 290-370 m μ .

Ultraviolet light and oxygen bubbling of xylene will produce absorption changes in the region from 290-400 m μ similar to those produced by x-rays and gamma rays. The absorbance curves vs. wavelength are similar to those shown in Fig. 1 of Part II. That the absorbance curves are not exactly alike is shown by Fig. 2, where the absorbance changes are plotted as ratios to the absorbance produced by 10^6 r of gamma radiation. The variation from constancy indicates the difference in the shapes of the two curves.

Previously it was noted that the absorbance (page 5, Part II) of an irradiated solution decreased with time. On repeating our results on a newer, more stable spectrophotometer no such decrease was noted with time. It is assumed, therefore, that the original decrease of the absorbance at a particular wavelength with time was instrumental rather than actual.

One of the mechanisms capable of explaining the decrease in the scintillation light output of a solution of xylene and diphenyloxazole (PPO) (Fig. 3, Part II) with large gamma-ray doses, has been postulated (Part II) to be the decrease in the number of scintillation molecules. A simple and conservative calculation has been made on the probable number of PPO molecules that could have been modified structurally by the radiation products of xylene. This calculation is based on the assumption that the PPO molecule, because of its weaker chemical bonds, is more easily attacked

*Part I appears in Report on Biological, Medical and Biophysics Programs, ANL-5518, p. 8 (1956); Part II appears in Radiological Physics Division Semiannual Report, ANL-5596, p. 3 (1956).

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Fig. 1. Absorbance changes induced in xylene by various doses plotted as ratios to absorbance produced by $10^6 r$ in the region 290-370 $m\mu$.

Fig. 2. Curve I is the ratio of the absorbance change produced by bubbling oxygen through irradiated xylene to the absorbance change produced by $10^6 r$ of gamma radiation.

Curve II shows the absorbance change produced by irradiating xylene with ultraviolet light (2537 Å) compared to the effect of $10^6 r$ of gamma radiation taken as unity.

by the radiation products than is the xylene molecule, and that the G factor (the number of modified PPO molecules per 100 ev) is 1.0.* The calculation is as follows:

$$1 \text{ erg} \approx 6.3 \times 10^{11} \text{ ev}$$

$$1 \text{ r in air} = 88 \text{ erg/gm (where } Z = 7.2 \text{ and assuming } W, \text{ the energy to produce an ion pair, is equal to } 34 \text{ ev)}$$

$$1 \text{ r in xylene} = 75 \text{ erg/cc (} Z = 7; \text{ density, } S = 0.86)$$

$$10^6 \text{ r} = 7.5 \times 10^7 \text{ erg/cc} \times 6.3 \times 10^{11} \text{ ev/erg} = 4.7 \times 10^{19} \text{ ev/cc.}$$

Assuming a G factor of 1.0 (where the G factor is the number of modified PPO molecules per 100 ev absorbed) then

$$4.7 \times 10^{17} \times 1.0 = 4.7 \times 10^{17} \text{ modified PPO molecules/cc.}$$

Noting that 2 g/l of PPO in xylene corresponds to approximately 5.5×10^{18} molecules/cc we infer that the 10^6 r radiation dose could produce approximately an 8 percent change in the number of scintillating PPO molecules. This change in the scintillator concentration is not sufficient to account for the large decrease in the light output at the lower radiation doses. If a larger value of G is assumed to account completely for the light output decrease at the lower radiation doses, then, if a linear relationship between dose and radiation products is assumed, the decrease in the light output at the larger radiation doses should be larger than that experimentally observed.

Warren L. Buck** has found that around PPO concentrations of the order of 2 g/l the degree of quenching of the light output by dissolved oxygen is a function of the scintillator concentration, the quenching being larger for the smaller concentrations. Since in our case the quenching of the light output by the dissolved oxygen is found to be greater for an irradiated solution than for an unirradiated one, it is felt that this independent finding is consistent with the view of loss of light emission through destruction of PPO molecules.

As mentioned above, the decrease in the light output, however, cannot be completely explained by the decrease in the PPO concentration alone; hence it must be assumed that other equally important effects are involved. Among these is the absorbance of the solution. From Fig. 1, Part II, it is noted that the absorbance increase in irradiated xylene extends beyond 400 mμ. Since the emission of PPO in xylene starts around 350 mμ the effects of absorbance on the light output are not negligible and cannot be neglected. Further elucidative studies are in progress.

*A conservative figure according to private communication of E. J. Hart and Sheffield Gordon, Chemistry Division, Argonne National Laboratory.

**Private communication (Physics Division, Argonne National Laboratory).

A SIMPLE CONTAINER FOR SCINTILLATING SOLUTIONS

I. B. Berlman and L. D. Marinelli

A container for organic scintillating solutions should be inert to the solvent and be highly reflective. In a previous article⁽¹⁾ a glass Pyrex cell was described. Recently a cell made of teflon⁽²⁾ has proven very satisfactory. Teflon is both inert to the solvent, xylene, and is very reflective in the wavelength region of the organic scintillators. It also flows under pressure so that a pressure seal is feasible. The teflon cell is machined from white solid stock into a right cylinder closed at one end. (See Fig. 3.) A shoulder is cut into the inside rim of the open end to retain a glass disk which is the window of the cell. The glass disk should be at least 1/8 in. thick and fire polished around the cylindrical edge. To avoid the use of cement, which is generally attacked by the solvent, a tapered fluorothene ring⁽³⁾ is used. By pressing this ring over the window end of the teflon container, the edge of the latter is forced against the glass disk, and a satisfactory seal is produced. The fluorothene ring should have an inner taper of at least 6°. An air space should be left above the solution to allow for thermal expansion of the solution.

This cell is easily disassembled. First the fluorothene ring is removed. Then by applying Scotch tape to the top surface of the glass window, the glass disk is pulled out of the teflon container.

Fig. 3. Diagram of teflon cell.

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1. Berlman, I. B. Report on Biological, Medical and Biophysics Programs, Part I, ANL-5518, p. 14 (Jan. 1956).
2. C. E. Miller has concurrently found teflon containers satisfactory for NAI crystals.
3. The tapered ring idea is due to A. J. Bragagnolo.

THE CONCENTRATION OF RADON IN THE ATMOSPHERE

Part I. Chronological Variations

H. F. Lucas, Jr., F. I. Ilcewicz and A. F. Stehney

In measuring natural amounts of body radium in humans by the radon method,⁽¹⁾ it is necessary to know what fraction of the radon collected in breath samples is derived from atmospheric radon previously absorbed by body tissues. Earlier work^(2,3) has shown that the rate of elimination of absorbed radon decreases with an apparent half-life of 10 to 15 hours during the period in which samples are collected. Thus, a knowledge of the concentration of radon in air breathed by subjects for one or two days prior to the analysis may be required for a satisfactory estimate of the correction for residual atmospheric radon.

The present report describes measurements which were made on a semicontinuous basis in order to evaluate the degree of variation in the concentration of radon in the atmosphere. Although it was quickly apparent that large variations occurred during a given day and from week to week, analyses have been continued in order to determine values during different seasons of the year and to provide data for a detailed correlation with meteorological data which is being carried out by the Meteorology Group of this Division. (See Section II of this report.) The data given here are in the nature of a progress report in order to summarize the results obtained up to this time.

Air samples were obtained through an intake duct, the end of which was located three feet above the ground and one foot from the east wall of a one-story building (D-203). Air was drawn through Drierite and Ascarite in order to remove water and carbon dioxide, and the radon was absorbed on charcoal in a trap immersed in a dry-ice cooled bath.

After completion of sample collection, the radon was transferred to a ZnS scintillation counter⁽⁴⁾ for measurement. Sampling was conducted continuously during the normal work week, i.e., from Monday morning to Friday afternoon. Three 8-hour samples were obtained each day: the "morning" sample from 00:45 to 08:45; the "day" sample from 08:45 to 16:45; and the "evening" sample from 16:45 to 00:45. Since the air was sampled at a uniform rate, each sample represents the mean radon concentration during an 8-hour period. The total volume of sample for each run was about 100 liters and counting rates ranged from 50 to 600 counts per minute. All samples were counted to a 90% confidence level of 2%. Sampling operations were done on a semiautomatic basis, and the total operator time required was less than two man-hours per day.

Fig. 4. Eight-hour averages of the concentration of radon in the atmosphere during the week of October 29 to November 2, 1956.

- △ Sampling period from 00:45 to 08:45 (Morning, M)
- Sampling period from 08:45 to 16:45 (Day, D)
- Sampling period from 16:45 to 00:45 (Evening, E)

Fig. 5. Weekly averages of the concentration of radon in the atmosphere during the period of August 20 to December 1, 1956.

- Sampling period from 00:45 to 08:45
- Sampling period from 08:45 to 16:45
- Sampling period from 16:45 to 00:45

The concentration of radon for each sample obtained during one week is shown in Fig. 4 as an illustration of the large variations which occurred each day. In general for a 24-hour period, the morning samples had about twice the concentration of radon the day samples had, and the evening samples were of intermediate value. In addition, significant changes in the concentration of radon for each sampling period occurred during the week.

In order to indicate the longer-term variations in the concentration of radon for the period studied, weekly averages for each sampling period were calculated and plotted in Fig. 5. The relative values between daily sampling periods are similar to those shown in Fig. 4 although it is evident that the radon values for the week illustrated in Fig. 4 were unusually high. The mean radon concentration in air for each of the three sampling periods is given in Table 1, together with the minimum and maximum values for each period. The smallest value obtained for an 8-hour sample was $0.090 \mu\mu\text{c Rn}$ per liter, and the largest value was $1.37 \mu\mu\text{c Rn}$ per liter. The average for all samples was $0.41 \mu\mu\text{c Rn}$ per liter of air.

TABLE 1

Summary of values for the concentration of radon in the atmosphere during the period of August 20 to December 1, 1956

Sampling period	Micromicrocuries of radon per liter of air		
	Mean	Minimum	Maximum
08:45 - 16:45 (Day)	0.272	0.090	0.67
16:45 - 00:45 (Evening)	.40	.142	.83
00:45 - 08:45 (Morning)	.55	.184	1.37
All	0.41		

References

1. Stehney, A. F. and H. F. Lucas, Jr. International Conference on Peaceful Uses of Atomic Energy, Vol. 11, Paper 852 (United Nations, 1956).
2. Harley, J. H., E. Jetter, and N. Nelson. Elimination of Radon From the Body, Report No. 3, New York Operations Office, AEC (1951).
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THE CONCENTRATION OF RADON IN THE ATMOSPHERE

Part II. Comparison with Meteorological Data

F. C. Kulhanek, G. A. Zerbe and H. Moses

To ascertain the effect of meteorological variables upon the concentrations of radon reported above, the 8-hour averages of the radon content and following meteorological data were plotted as a function of time in Figs. 6a and 6b.

A. Stability. The stability is measured as the difference in temperature between the 144-foot and 5.5-foot levels. Positive values indicate inversion conditions. Two-minute averages taken each hour were used to plot the curves of Figs. 6a and 6b.

B. Atmospheric Pressure. Pressure values are instantaneous on-the-hour readings.

C. Wind Speed at the 150-foot and 19-foot Levels. These readings represent 10-minute averages taken each hour and centered about the hour.

D. Rainfall. The values represent the total rainfall accumulated in each hour.

An examination of Figs 6a and 6b shows rather strikingly the correlation between atmospheric stability and the magnitude of the atmospheric radon concentration. It may also be noted that not only is the relationship between the actual concentrations good, but the change in the radon value between successive readings appears to be well correlated with stability measurements. For example, during the early morning hours when an inversion is present the concentration of the radon almost invariably increases, conversely during the afternoon period, when the atmosphere is unstable and vertical mixing takes place, the radon concentration decreases.

Another striking relationship is the effect of rainfall on the radon concentration. After rainfall, the amount of radon concentration in the atmosphere is lower than on corresponding times on days in a dry period. Apparently the water in the top layers of the soil inhibits radon release to the atmosphere. One would expect that the amount of radon released from the ground would be related to the changes in atmospheric pressure. The data presented fail to show an obvious relationship.

14

Fig. 6a

Fig. 6b.

Fig. 6. Time variation of atmospheric radon concentrations and meteorological elements: pressure, stability, rainfall, and wind speeds at 150- and 19-foot levels above ground.

THE THORIUM²²⁷ ACCIDENT - FINAL COMPARISON BETWEEN EXPERIMENTAL RESULTS AND CALCULATED VALUES OF RETENTION AND ELIMINATION

L. D. Marinelli and P. F. Gustafson*

In two previous reports,^(1,2) there were presented preliminary data, embodying measurements and calculations of both gamma radiation in vivo and activity of excreta, on a human accidentally contaminated with Th²²⁷ Cl₄ through a puncture wound. The accident occurred two days after the separation of Th²²⁷ from its Ac²²⁷ parent. A more complete and accurate paper, supplanting these reports, has been accepted for publication in the open literature.⁽³⁾

It is the purpose of this note to compare the experimental results reported in the latter with theoretical computations which have been completed since that time.

The experimental data of Ref. (3), pertinent to this comparison, namely gamma-ray activity in vivo and activity of excreta, are presented herein in tabular form for convenience of the reader (Tables 2 and 3). Details of the theoretical work will be available shortly in a laboratory report⁽⁴⁾ covering the various aspects of gross metabolism of radium grown within the body from the decay of a thorium parent. The assumption is made therein that this radium is retained according to the power law formula

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

where $R_C(t)$ is the fraction of the injected dose that remains in the body at time t (in days). When radium is grown from a metabolically stable thorium parent present in the body (as is actually found in practice a few days after administration - see Tables 2 and 3), this assumption leads to the following expression for the number $R(t)$ of Ra atoms present at time t for each atom of Th originally injected

$$R(t) = \lambda_T e^{-\lambda_T t} \int_0^b e^{-(\lambda_R - \lambda_T)u} R_C(u) du \quad (2)$$

where: $u = t - \tau$ and λ_T and λ_R are the decay constants (per day) of thorium and radium respectively. The daily elimination rate $E(t)$ of radium follows from the differential equation of chemical balance within the body:

$$\frac{dR(t)}{dt} = \lambda_T e^{-\lambda_T t} - \lambda_R R(t) + E(t) \quad (3)$$

*Division of Biological and Medical Research

**In calculating the elimination rate as tabulated in Ref. (2) p. 25 the term $\lambda_T e^{-\lambda_T t}$ in expression (3) above was erroneously omitted.

and is given explicitly by

$$E(t) = R_C(t) \lambda_T e^{-\lambda_R t} - \lambda_T e^{-\lambda_T t} + (\lambda_R - \lambda_T) R(t) \quad (4)$$

The values of $R(t)$ and $E(t)$ for $R_C(t) = 0.3 t^{-0.7}$ are shown in Table 4 as calculated by expressions (2) and (3), respectively, namely as atoms of Ra^{223} per atom of Th^{227} originally present in the body.

Comparison with the experimental findings (Tables 2 and 3) requires conversion of these units in μc of Ra^{223} and knowledge of the activity of Th^{227} originally present in the body. The latter was obtained by extrapolating at $t = 0$ the experimental findings *in vivo* (Fig. 7) and turns out to be $4.7 \mu c$, neglecting the excretion for $t < 12$ days in order to eliminate the uncertainties due to the removal of Th^{227} by excision (6th day) and Ca EDTA administration.

Fig. 7

The results of $R(t)$ in Table 2, multiplied by $4.7 \times \lambda_R / \lambda_T = 7.8$ can then be compared directly with the experimental values. As shown in Fig. 7 the agreement is satisfactory after the 20th day; agreement at earlier times cannot be expected because of the complicating circumstances mentioned above.

Comparison of the data on excretion, requires similar conversion of the theoretical values of $E(t)$, namely multiplication by the factor 7.8 used before. The experimental values of $E(t)$ as given in Table 3 are deficient in the sense that the values on feces, by far the most important route of

elimination, are not only few, but represent extrapolation from incomplete samples. Proof of their inadequacy is the sharply varying ratio of feces to urine (27.5 to 1.0) which is at variance with the relatively constant ratios obtained by all investigators reporting complete collection of excreta (Table 5). Fortunately, however, the urinary collection was assayed more often and it was more nearly complete. The experimental points are shown

18

TABLE 2

Total body activity after excision

Days*	$\mu\text{c Th}^{227}$	$\mu\text{c Ra}^{223}$	Days*	$\mu\text{c Th}^{227}$	$\mu\text{c Ra}^{223}$
6	$(5.64 \pm 1.40)**$	$(.93 \pm .23)$	40	$1.03 \pm .08$	$.17 \pm .05$
12	$2.92 \pm .07$	$.38 \pm .05$	49	$0.62 \pm .04$	$.10 \pm .05$
19	$2.36 \pm .06$	$.26 \pm .05$	97	$0.10 \pm .01$	$.02 \pm .05$
33	$1.28 \pm .08$	$.18 \pm .05$			

*Days after separation of Th^{227} .

**Values in parenthesis refer to body burdens which would have been obtained in vivo if the proportion of Ra^{223} and Th^{227} in the excised tissue were representative of the partition existing in the body. Errors indicated above refer only to standard errors in counting. Accident occurred two days after separation.

TABLE 3

Daily excretion of Th^{227} and Ra^{223} *

Th^{227}		Ra^{223}	
Day**	μc	Day**	μc
<u>Urinary Excretion</u>			
2	2.9×10^{-1}		
6	2.2×10^{-2}		
7	2.0×10^{-2}	7	6.9×10^{-3}
10	3.1×10^{-3}	10	5.6×10^{-3}
13	3.9×10^{-3}	13	8.4×10^{-3}
14	1.1×10^{-3}	14	3.1×10^{-3}
27	5.6×10^{-4}	27	2.3×10^{-3}
48	1.2×10^{-4}	48	2.1×10^{-3}
70	4.7×10^{-5}	70	2.2×10^{-4}
159	1.5×10^{-6}	159	2.6×10^{-5}
221	1.9×10^{-7}	221	2.5×10^{-6}
<u>Fecal excretion</u>			
7	4.8×10^{-3}	7	1.9×10^{-1}
36	2.0×10^{-4}	36	2.4×10^{-2}
48	1.1×10^{-4}	48	6.9×10^{-3}
72	1.7×10^{-4}	72	2.4×10^{-2}
221	1.2×10^{-6}		

*Based on daily outputs of 1250 cc of urine and 25 g dry feces.

**Days after Th^{227} separation.

Fig. 8

the thorium parent before scientifically sound figures for maximum permissible levels can be calculated.

in Figure 8. The calculated urinary excretion curve is shown by the solid line: this was obtained by using the values of $E(t)$ (Table 4), multiplied by 7.8 (conversion to μc of Ra^{223} and $4.7 \mu\text{c}$ of Th^{227}) and by 0.03. The latter represents the average fraction of radium eliminated which is present in the urine of patients at various times after ingestion (Table 5). The agreement is better than can generally be expected from the accidental nature of the exposure.

It should be realized, however, that this information, though valuable in specific instances of internal thorium contamination, must be supplemented with more detailed data on the localization of

TABLE 4

Theoretical values of $R(t)$ (retention) and total elimination $E(t)^*$

t (days)	$R(t)$	$-E(t)$	Ratio
2.22	.0283	.0280	.99
4.45	.0346	.0277	.80
11.1	.0371	.0227	.61
22.2	.0297	.0152	.51
33.4	.0214	.0101	.47
44.5	.0149	.00668	.44
77.9	.00456	.00193	.42
111.0	.00134	.000557	.41

*Units of atoms of Ra^{223} per atom of Th^{227} originally present in the body, under the assumption that a single injection of Ra is retained according to the expression

$$R_C(t) = 0.3 t^{-0.7}$$

20
TABLE 5

Fraction of total Ra elimination present in urine*

Author	Reference	Time interval	**U/T (averages)	Route
Seil, et al.	(5)	0-4 days	.033	I.V.
Aub, et al.	(6)	28 d - 10 yrs	.045	Various
Norris, et al.	(7)	~24 yrs	.017	I.V.
Looney, et al.	(8)	0-30 days	.026	Inhalation
Stehney, et al.	(9)	1,000 days	.031	
		Average U/T	0.03	

*Data in this table are limited exclusively to instances of complete collection of excreta, sufficient sensitivity of assay and absence of medication.

**U/T = ratio of urinary to total excretion of radium.

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THE HUMAN SPECTROMETER

Introduction

C. E. Miller

During the past 6 months, the Human Spectrometer has been utilized for various tasks which may be grouped in three general areas: in vivo measurements, background studies, and studies of specific radioactivity.

(I) In vivo measurements have been made on subjects carrying body burdens of Sr^{85} , Ra^{226} and Te^{129} . In addition, the normal radioactivity of healthy, unexposed subjects - both employees of Argonne National Laboratory (control subjects) and subjects from Sweden, Denmark, France, England, Japan, Indonesia and India - has been determined. Cs^{137} and K^{40} ratios have been measured and compared to the values obtained previously.

(II) The gamma-ray background of the facility has been re-examined and some additional shielding tests performed. The background count at low energies, i.e., below 150 kev, can be reduced by as much as 50 per cent by the addition of 1/8 in. of lead around the crystal. Earlier tests had demonstrated the gamma-ray background to be very stable and reproducible. Slight long-term variations which did result could be correlated with changes in barometric pressure. More recently, however, during periods of south wind, increases of 10 per cent in the integral background and increases of 100 per cent in particular energy bands have been noted and studied. These fluctuations are to be attributed to reactor gases originating approximately one half mile south of our building.

(III) Both the noise and radioactive backgrounds of several 5-inch DuMont photomultipliers have been scrutinized. This study has been undertaken because the conflicting results that have been published by various authors concerning magnitude and time-variations of the noise background of phototubes due to temperature changes, etc., are of little help in the attainment of the low, stable backgrounds which are so essential to accurate counting of small numbers of quanta of feeble energy. The specific radioactivity of numerous samples of glasses and their constituents have been measured in an attempt to identify the primary source of radioactivity entering as a raw material into the fabrication of the glass used in the manufacture of phototube envelopes.

I. In Vivo Measurements

C. E. Miller and L. D. Marinelli

In the following sections dealing with in vivo measurements of Sr^{85} , Te^{129} , and Ra^{226} , reference will be made to the chair position and to the one-meter arc technique. The techniques will not be discussed here since the pertinent information on the arc technique can be found in the open literature⁽¹⁾ and the chair position is discussed in Ref. (3).

A. Strontium-85

Two patients who had ingested tracer amounts of Sr^{85} at the Argonne Cancer Research Hospital under the care of Dr. R. J. Hasterlik have been measured in the iron room. Only a description of the measuring techniques and sensitivities obtainable will be mentioned here.

Sr^{85} has a physical half-life of 65 days and emits a gamma ray of 513 kev. In order to obtain the net spectrum of Sr^{85} in the body of a subject it is necessary to subtract from his gross spectrum the spectrum due to both K^{40} and Cs^{137} present in his body. This would be a simple task if the patient's normal spectrum had been obtained at a time previous to his ingestion of Sr^{85} .

Since this was not done, the net K^{40} and net Cs^{137} spectrum of an unexposed normal (control) subject of the same height and weight was used as follows. The control subject's net K^{40} spectrum was normalized to yield under the K^{40} photopeak an area identical to that existing under the K^{40} peak of the patient's spectrum. By simple subtraction, the Cs^{137} plus Sr^{85} spectrum of the patient resulted; then the net Cs^{137} spectrum of the control was normalized to yield under the Cs^{137} photopeak the same area as that of the Cs peak in the patient's spectrum. Subtraction of the normalized Cs peak yielded the true in vivo Sr^{85} spectrum of the patient. The spectra shown on Fig. 9 were obtained by this technique for subject WSM, discussed below.

The shape of the in vivo Sr^{85} spectrum thus obtained was compared with the spectrum obtained from a Sr^{85} source, buried in a Presdwood phantom of appropriate thickness placed at the same distance. The Sr^{85} spectrum determined in vivo was identical to the spectrum of Sr^{85} in the phantom throughout the relevant energy region, implying that the subject was not contaminated with any other isotope and that, furthermore, the phantom represented the patient's body insofar as scattering and absorption were concerned. The subject's Sr^{85} body burden could then be calculated from the counting rates and the amount of Sr^{85} used in the phantom.

Fig. 9. Gamma ray spectra of subject WSM.

Subject RM received $50 \mu\text{c}$ of Sr^{85} orally on 2/15/56 and was examined on 12/14/56 or 302 days later. This subject's body burden could be determined quite accurately on the 1-meter arc using only two 30-minute runs. In addition, he was run in the standard chair in order to determine the arc-to-chair ratio, since the latter position yields a higher counting rate than the arc due to closer spacing of the crystal to the body. The factor was found to be 3.6 to 1 in all channels. Subject RM had 121 grams of K, 5.5×10^{-9} c of Cs^{137} and 9.4×10^{-8} c of Sr^{85} .

Subject WSM received $10 \mu\text{c}$ orally on 11/28/55. On 10/25/56, or 332 days later, his gamma-ray spectrum was obtained utilizing the standard chair technique. His body contained 121 grams of K, 5.5×10^{-9} c of Cs^{137} and 1×10^{-8} c of Sr^{85} . This subject was not run using the one-

meter arc technique because the lower activity would have necessitated times longer than the patient was willing to give.

One interesting fact was noted. In the case of a bone-seeker, such as Ra^{226} , practically the same counting rate is usually observed whether the person on the one-meter arc faces toward or away from the crystal. Subject RM, instead, yielded a Sr counting rate 25 per cent higher when his back was toward the crystal: this finding can be taken as presumptive evidence that the material was not uniformly distributed throughout his skeleton.

If necessary, it is possible to measure as low as 2×10^{-9} c of Sr^{85} with perhaps a 20 per cent error. The accuracy obtainable will depend upon the availability of previously obtained K^{40} and Cs^{137} spectra for subjects of the same weight and build. If spectra from subjects whose builds are quite different from the patient's build are used, rather large errors may result since the counting rates for the Sr^{85} are found as the differences among several large numbers. Considerably smaller amounts could have been measured with greater accuracy if Cs^{137} were not present, for the K^{40}

spectrum can always be measured to any desired accuracy by means of K^{42} . Data on the metabolic features of Sr^{85} of these patients will be reported by Dr. R. J. Hasterlik of the Argonne Cancer Research Hospital.

B. Radium-226

The Health Division and the Radiological Physics Division of Argonne National Laboratory and the Argonne Cancer Research Hospital are conducting a joint study of individuals containing Ra^{226} above background levels. The Radiological Physics Division is responsible for all necessary measurements of radioactivity. Three subjects have been studied in the last 6 months, and several cases have been scheduled for the future. Two subjects, WF and EW, accumulated the Ra burden while painting radium watch dials. The third patient, subject No. 120,⁽²⁾ drank radium water as a child for a period of two years, namely from 24 to 26 years ago. The total body burdens found are as follows:

Subject WF - $5.2 \mu c$; Subject EW - $1.2 \mu c$; Subject No. 120 - $0.15 \mu c$.

The measuring technique (chair, arc, collimated scan, etc.) used on a particular case is dictated mainly by the total body burden, as ascertained from the results of our preliminary arc reading and, to a lesser extent, by the clinical history of bone changes, if any have been recorded. The body burdens of the three subjects above were determined using the one-meter arc technique and the appropriate crystal. The 4 in. x $1\frac{1}{2}$ in. NaI(Tl) crystal was used for subjects WF and EW and the 8 in. x 4 in. crystal was used for subject No. 120. The two subjects, WF and EW, individually yielded the same spectra and counting rates whether facing toward or away from the crystal, thus indicating that the material was fairly uniformly distributed within the skeleton. Their burdens were calculated using the equation developed by Marinelli. This equation used the subject's average thickness, the absorption coefficient of radium gamma rays in Presdwood, the counting rate of $1 \mu c$ of radium in air at 1 meter, and the counting rates obtained with the subjects facing toward and away from the crystal on the one-meter arc. This technique depends upon the subject's average thickness and requires the determination of the absorption coefficient of radium through Presdwood.

In order to evaluate independently the accuracy of this technique, their burdens were also determined by a method which might be called the spectro-metric trial and error approach. A radium source is placed at a position corresponding to the center of the body when curved on the one-meter arc, namely, at one meter minus half the thickness of the subject, or very close to 90 cm. Presdwood is then placed in front of the source, that is, between the source and the crystal, and behind the source. The effect of the Presdwood behind the source is to scatter gamma rays, emitted away from the crystal through angles of 90° to 180° back to the detector. Varying the amount of Presdwood behind the source essentially affects the pulse spectrum only at

the low energies, whereas the Presdwood in front of the crystal attenuates the photopeaks and tends to fill up the dip normally present between a photopeak and its associated Compton peak. With some practice, the shape of the subject's spectrum can be quickly matched quite accurately if the higher energy region of the spectrum is used as a guide to obtain the correct thickness of the front section of Presdwood and the ratio of the high energy band to the low energy band is used to determine the thickness of the back section of the phantom.

Once the shape of the spectra of both patient and phantom are matched, the Ra (B+C) body burden is given directly by the source strength in the phantom times the ratio of the counting rates in the region 0.2-2.2 Mev; the Ra²²⁶ burdens equal the Ra (B+C) values divided by 0.31, the radon retention factor.

The results obtained with this technique checked the value found by Marinelli's equation within 2 per cent. It is of decided interest that these techniques, both aimed at substituting a point source in a scattering medium for a definitely irregular distribution in the human body, do yield practically identical results, though they are based on "equivalence" assumptions dependent on different parameters, namely, equivalent thickness and depth reached by geometrical consideration on the one hand and spectral equivalence on the other.

In the course of these measurements the activity located in subject EW's knees, shins, ankles and head was measured by scanning with a collimated crystal.

Subject No. 120's body burden was measured using the 8 in. x 4 in. NaI(Tl) crystal on both the one meter arc (facing toward and away from the crystal) and in the standard chair (position No. 1)(3) in order to determine the arc-to-chair ratio for radium. Subject 120's counting rate was approximately 25 per cent higher when facing the crystal than when lying with his back to the crystal, thus indicating a nonuniform skeletal distribution. Consequently, the activity in this subject's knees, shins, ankles, and head was also recorded in a fashion identical to that used for EW. The activity in subject 120's knees, ankles and head was lower than that of subject EW's by the factor equal to the ratio of their body burdens. Subject 120's shins, however, seemingly contained almost as much radium as subject EW's shins although the body burdens were in the ratio of 1.8. The arc-to-chair ratio varied with the energy bands; this may be due to the peculiar nonuniformity in the Ra²²⁶ distribution in the skeleton of this patient.

Further studies will be performed on Subject No. 120 in an attempt to determine the amount of mesothorium, if any, which might have been mixed with the original radium solution. In addition, the skeleton, and the long bones in particular, will be scanned more carefully with a collimated detector. This subject is of great interest since it is the only case we have studied in which, in the absence of any definite therapeutic objective, the subject received the material as a child.

C. Tellurium-129 Inhalation Accident

During August, two employees submitted routine urine specimens to the Bioassay Section of the Industrial Hygiene and Safety Division which, when ashed and counted, yielded high beta activity that persisted in check samples collected the following night.

Subject A, whose urine yielded the highest activity, was counted in the steel room on 8/17/56 in both the standard chair and on the one-meter arc. The element was identified as Te^{129} and the body burden estimated to be $1.9 \mu\text{c}$, using both Marinelli's equation and the trial-and-error spectrographic technique discussed in the Ra^{226} section of this report. This man was then scanned with a collimated $2\frac{1}{2}$ in. x $2\frac{1}{8}$ in. NaI(Tl) scintillation counter and the material found to be located principally within the chest. Subject B's burden was found to be $0.815 \mu\text{c}$ as of 8/24/56, and it also was located in his chest. Once the contaminating nuclide was identified, review of the operational log book set the most likely date of the contamination at July 16, 1956; thus, the first measurement on subject A was made on the 32nd day after the exposure. Employees A and B submitted complete urine and fecal specimens eliminated from the 35th through the 39th day, and the activities were quantitatively analyzed by gamma-ray spectrometry. Approximately $1\frac{1}{4}$ per cent of their total body content was eliminated each day, equivalent to a metabolic or biological half-time of 55.6 days. This value, combined with the decay constant of the isotope (half-life = 41 days) yields an effective half-life in vivo of 23.6 days.

The body burden of Subject A was determined on the one-meter arc on the 32nd, 39th, 46th, 63rd and 115th day and with the standard chair technique on the 115th and 151st days. The body burdens estimated in vivo fell on a straight line on a semi-log plot with an effective half-time of 22 days.

Subject B's burden was determined, using the one-meter arc technique, on the 39th, 45th, 63rd and 115th day, and an effective half-life of about 22.5 days found. These two values are in fair agreement with the 23.5 day half-time estimated from the excretion rates on the 35th - 39th days, and, as in previous tests, lend considerable confidence in the correctness of our in vivo measurements.

On the 45th and 63rd days, the spectra of both individuals were determined while on the one-meter arc, with and without a $1/4$ in. thick sheet of lead interposed between his chest and the counter. The difference between the two readings is due to the attenuation of those gamma rays being emitted only from the chest area. By plotting the differences found on the two dates, the effective half-time in the lung was found to be of the order of 15 days.*

*This value is nearly one half that found for RaSO_4 inhalation.(4)

In the above cases, therefore, the material was evidently inhaled and deposited in the lungs; hence the 22-day effective half-life, constant from the 35th day through the 151st day, is representative of inhalation exposures only. Namely, the lungs acted as a reservoir and released the material into the rest of the body. There is reason to suspect that the material was not entirely an insoluble compound, since 60 per cent of the amount excreted was present in the urine and only 40 per cent in the feces, whereas a truly insoluble compound would have been present only in the feces. During the course of this work several tests of self-consistency were necessary as the net activity of Te^{129} approached that of the K^{40} and Cs^{137} present in the subjects' bodies. This statement is self-evident when one considers that: (a) the Te^{129} gamma ray (0.465, 0.695 and 1.12 Mev) scintillation spectrum is astride the spectra of the other two; b) 1 μc of Te^{129} in vivo yields about 780 c/m within the energy band of 40-760 kev and 170 c/m in the 365-728 kev energy band, and c) the average $\text{K}^{40} + \text{Cs}^{137}$ human burden yields 140 and 40 c/m, respectively, in these bands.

The shape and magnitude of the K spectrum could be determined quite easily since the size of the K^{40} gamma photopeak (1.46 Mev) is independent of the magnitude of the others and has been previously determined in this laboratory for subjects of various anatomical builds.⁽³⁾ However, the size of the Cs^{137} peak could not be determined early in the experiment since its photopeak (661 kev) was not clearly separated from that of the 695 kev gamma of Te^{129} .

The normal ($\text{Cs}^{137} + \text{K}^{40}$) spectrum of subject A was determined in two ways. By the first approach, the gamma-ray spectrum of a normal, unexposed individual of the same build, and having the same amount of K^{40} , was subtracted from subject A's gross spectrum. The resulting spectrum could be assumed to be due essentially to Te^{129} . The Te^{129} determined in this fashion showed an effective half-life of 22 days. This value, however, could be slightly in error since subject A could have a Cs^{137} burden different from that of the control subject despite the latter's identical K^{40} content.

The second approach consisted of taking the difference between the gross spectra of subject A taken 39 days apart. If the Cs^{137} and K^{40} content of the individual has remained constant in the interim (our experience on our Cs^{137} controls confirms this assumption), the spectral difference will represent the in vivo Te^{129} spectrum of the subject in question, and bear to the amount of Te^{129} present in the first of the two dates a ratio equal to $(1 - e^{-693 \times 39 / T_{\text{eff}}})$ where T_{eff} is the effective half-life of Te in the body. If the choice of T_{eff} is correct, the subtraction of the Te^{129} spectrum from the gross spectrum should yield the $\text{Cs}^{137} + \text{K}^{40}$ spectrum. The correctness of the latter could be judged by its smoothness in the region of 465 kev. This procedure confirmed the value of $T_{\text{eff}} = 22$ days obtained at earlier times when the activity of Te^{129} was substantially higher than the normal activity of the body.

The K^{40} component of the resulting $Cs^{137} + K^{40}$ spectrum was then subtracted by normalizing a previously determined K^{42} spectrum for a subject of the same height and weight^(3,5) yielding the in vivo Cs^{137} spectrum. Subject A's net total gamma-ray spectra on two days referred to above are shown in Figure 10 together with the $K^{40} + Cs^{137}$ spectrum thus determined. The separate Te^{129} and Cs^{137} spectra are shown in Figure 11.

Fig. 10. Gamma ray spectra of subject A.

Fig. 11. Subject A's net in vivo Cs^{137} and Te^{129} spectra.

An analysis similar to the one mentioned above can be evolved from the difference between the initial gross spectrum and the gross spectrum obtained at the end of the 39-day interval in question. Without any assumptions as to T_{eff} , but keeping in view the essential smoothness of the $K^{40} + Cs^{137}$ spectrum at 465 kev, an empirical factor of proportionality to the Te^{129} spectrum can be obtained which will yield by proper subtraction the true normal $K^{40} + Cs^{137}$ spectrum. All these procedures indicated that subject A had a body burden of 1.1×10^{-8} c of Cs^{137} . The Te^{129} burden of B was considerably less than A's and consequently it was of greater importance to correct his net total spectra for the K^{40} plus Cs^{137} component.

D. Cesium-137 in Humans

It was reported in the January 1956 Semiannual Report⁽⁵⁾ and elsewhere⁽⁶⁾ that most human beings contained a slight amount of Cs^{137} , a fission product due to world-wide contamination by nuclear detonations. In July 1956⁽³⁾ we reported that the burden had increased approximately

21

60 to 100 per cent in 4 subjects on whom consecutive runs had been made since July, 1955. The Cs^{137} contents of the four original subjects plus eight additional unexposed "control" subjects have been followed from February 15, 1956 to December 15, 1956.

The data previously taken on the four subjects have been carefully analyzed and they are shown in Fig. 12, where the Cs^{137} contents are expressed in units of $10^{-5} \mu\text{c}$ of Cs^{137} per gram of potassium. It is evident that, although the ratio varies with the individual, in each case a definite increase was experienced from the summer (or fall) of 1955 to the spring of 1956 and that, thereafter, no substantial variation has ensued. Less extensive readings on another group of eight persons, also from our laboratory, dating from October 1955 to August 1956, have shown substantially the same trend in the Cs^{137}/K ratio. The potassium content of all our subjects has remained practically constant throughout this period.

Fig. 12.

Since Cs^{137} has a relatively short average life in the body, we may infer from these results that, if equilibrium conditions of intake and elimination in the food chain have prevailed, the rate of Cs^{137} fallout has not varied much in the Chicago area throughout the year 1956.

The Cs level of numerous subjects from various foreign countries has been determined during their visits to our Laboratory. All Europeans have shown much the same Cs/K ratio as our control group. Outstandingly lower have been those individuals from Japan, India and Indonesia who include little or

no meat and milk in their diet. Two non-meat-eating, non-milk-drinking Indonesian subjects showed no detectable burden of Cs. Two other individuals from India, who had been drinking milk and eating meat during approximately 50 days previous to their measurement, yielded Cs/K ratios about 50 per cent of our control value.

Of interest are some values of the Cs^{137}/K ratio in meat and milk included in Fig. 12. Although incomplete for a satisfactory evaluation of the trends of Cs^{137} contamination of the food supply, these findings on

milk are clearly lower than those reported for powdered milk by Sievert.⁽¹¹⁾ The discrepancy may well be due to contamination with short-lived fission products in Sievert's samples; we have no spectrometric evidence of their existence in ours.

In a subject (RER), repeatedly tested in March-April 1956, the Cs^{137}/K ratio in the urine was found to be $1.07 \pm 0.07 \times 10^{-5} \mu\text{c Cs}^{137}/\text{g K}$, whereas in vivo this ratio was 2.2 ± 0.4 . These values, taken together with the values reported above for meat and milk, strongly imply that, if steady conditions of intake and elimination prevailed:

- a) subject RER derived somewhat less than 65% of his K from cattle products (since, at equilibrium, the Cs/K ratio should be the same for food as for excreta). This is consistent with the dietary habits of the individual, and,
- b) some discrimination, at the human kidney clearance level, seems to take place against Cs^{137} , since the urine Cs^{137}/K ratio is only 0.49 of the same ratio in vivo. This hypothesis is consistent with the findings of Threefoot, et al.⁽¹²⁾ in dogs where the Cs^{134}/K kidney clearance ratio averaged 0.48.

II. Background Studies with the Human Gamma-Ray Spectrometer

C. E. Miller

The gamma-ray background of the 8 in. x 4 in. NaI crystal, when located within the Human Spectrometer Cave, is 1200 c/m in the energy band of 40 kev to 1.8 Mev and 500 c/m for all pulses above 1.8 Mev. Surrounding the crystal with 1/16 in. or 1/8 in. virgin lead reduces the gamma-ray background at the lower energies by approximately 250 c/m. The addition of a greater thickness of lead does not reduce the background further, indicating that the local gamma-ray background contains degraded low energy gammas easily absorbed by small thicknesses of high Z material.

The pulse height spectrum of high energy events has the shape shown by Fig. 13. The peak falls at an energy of about 80 Mev which is approximately the amount of energy that would be dissipated by high energy mesons transversing 4 in. of NaI. A few pulses per minute are observed which are equivalent to 500 Mev electrons. A gamma-ray spectrometer must thus be able to sort 500 Mev pulses without producing false pulses if the lowest backgrounds possible are to be attained. A reflection of only 0.1 per cent of the initial pulse, occurring after the dead time of the analyzer, would result in a false background peak at 80 kev. As larger crystals become available, the size of the cosmic-ray-produced pulse will increase, and more stringent demands will be placed on the electronic system.

Fig. 13.

The gamma-ray background in the above-mentioned energy band had been examined for reproducibility on different dates. Five consecutive 50-minute background runs on 4/30/56 yielded counting rates of 1212, 1208, 1208, 1211, and 1204 c/m or an average value of 1209 ± 2.5 c/m. The following day, 5/1/56, four consecutive 50-minute runs yielded values of 1255, 1251, 1257 and 1249 or an average rate of 1253 ± 2.5 c/m and again all fell within the expected statistics. Although the average total counting rates differed by 44 c/m, the difference was spread over the energy band of 40 kev to 1.8 Mev, hence it did not yield sufficient statistical results to attribute the difference to any particular isotope. Futile attempts were made to correlate it with barometric pressure and cosmic rays with the hope of identifying the cosmic ray spectrum.

The 980-minute overnight background taken from 12/4/56 till 12/5/56 gave an integral counting rate of 1455 c/m. The spectral difference between these readings and the usual lower ones obtained at other times yielded an A^{41} spectrum. It was calculated that 3×10^{-9} c of A^{41} were present in the $8m^3$ steel room, namely a concentration of $0.375 \mu\mu c$ per liter of air. The wind directions were checked and found to be from the south, mainly parallel to the line joining the reactors to our building air intakes. These dates were found to correspond to the dates of the first critical experiments of EBWR.

The wind directions were then checked for the previously mentioned dates. The wind had not been from the south from 4/25/56 until the night of 4/30/56 - 5/1/56, thus accounting for the lower background on 4/30/56. The wind direction changed from SE-S during the night of the 30th and into the S-SW by 9:30 A.M. remaining S-SW for the next two days.

The reason A^{41} was not observed during the runs made before 12/5/56 is undoubtedly due to the physical arrangements of the original research reactor and of our building. The research reactor has a holdup

room permitting the short half-life isotopes to decay before release. Secondly, no air is supplied directly to our basement wing housing the steel room. However, outside air does leak into the basement from air ducts passing through the basement area. An air pump circulates basement air through the steel room, thus resulting in an additional holdup time.

A correlation between seemingly spurious high backgrounds obtained at various times by our apparatus and wind directions indicated also that a source of activity lies to the south of our building.

III. Studies on Photomultiplier Tubes

C. E. Miller and O. J. Steingraber

A. Uniformity of response over photocathode surface

Low energy gamma rays interact in the first few millimeters of a NaI(Tl) crystal. The use of a thick crystal will not increase the detection efficiency but will only raise the background leading to a less sensitive spectrometer on account of the lower S/B (net count from source/background of detector) ratio. Successful use of a thin crystal depends greatly on photocathode uniformity since, due to its proximity to the photocathode surface, the quanta of light are not predominantly scattered throughout the crystal volume but strike the cathode directly in greater number than is the case for a thicker crystal. A great variation in local cathode response leads to the production of different photopeaks per monoergic gamma ray, depending upon the location of the event, and ultimately it results in very poor energy resolution.

The photocathode uniformity of several tubes has been examined and the effect of various shield potentials on electron collection from the photocathode checked. In lieu of a light pulser, the light obtained from Am^{241} gammas striking a NaI crystal was used as a light source. Crystals $4\frac{1}{4}$ in. x $\frac{1}{4}$ in. thick were canned in various ways and optically coupled to the tube in question. The collimated beam was then moved around the crystal surface. In order to avoid variations in crystal efficiency, or losses in the crystal due to edge effects which may be present in these tests, crystals $1\frac{1}{4}$ in. x $1\frac{1}{4}$ in. are in the process of being canned with the source permanently sealed within the can. This arrangement will utilize the same crystal at all points on the photocathode surface and insure the same sized light pulse at all locations.

The collection efficiency at the edge of the 5 in. photocathode surface, judging from peak heights, is about 80 per cent of that at the center. Varying the shield voltage affects the response to a limited degree, as long as the shield voltage is slightly higher than the first dynode potential. The peak heights drop considerably when the shield is connected directly to the first dynode

since numerous electrons are collected on this structure. A high noise level is present if the shield is at the same potential as the photocathode potential, due most likely to the high electric stress across insulated gaps.

In the 16-in. photomultiplier, besides the center to edge effect, a more important defect was noted, i.e., the photocathode sensitivity of some areas was only 60 per cent of that at adjoining areas. The 8 in. x 4 in. crystal, when mounted on the 16 in. phototube, yielded a Cs^{137} spectrum with two separate photopeaks, demonstrating that the light from a single scintillation is not sufficiently spread on such a wide cathode area by the MgO packing which encases the large crystal.

B. Phototube noise

The maximum sensitivity (i.e., lower detectable limit) of a counter is determined by the background counting rate of the system whether these background counts are due to residual radioactivity, photomultiplier tube noise, or amplifier noise. When using NaI(Tl) crystals as detectors, the radioactive background predominates for energies above 30 kev even in a well shielded cave, but the phototube noise is likely to be the controlling factor at lower energies. The sensitivity of a low energy scintillation counter thus depends upon the noise level of the particular phototube used, and the success or failure of a project may depend entirely upon individual luck in securing a low noise tube.

In order to clarify the nature of noise fluctuations, a series of tests was performed on a group of 5-inch DuMont tubes as received from the manufacturer, without guard rings. A group of six tubes was checked in the following manner: A NaI(Tl) crystal was connected to the tube and the H.V. adjusted so the 59.7 kev Am^{241} peak fell in channel 120 of the 256 channel analyzer. The crystal was removed and the noise spectrum obtained immediately after and at later times, while keeping constant the particular voltage used for each tube.

Different effects were noted with different tubes. In general, the noise decreased with time, and three new tubes yielded essentially the same background. The initial noise background depended upon the tube's previous history. That is, exposing the tube directly to daylight (not through a window pane) or to a fluorescent light (rich in ultra-violet) produced a very high noise level requiring perhaps 24 hours to return to normal, whereas exposure to indirect incandescent light had little, if any, effect. It seems apparent, therefore, that the type of illumination used in exposure may explain the disparity in views expressed by different workers.⁽⁷⁾

Only one tube demonstrated the hysteresis effect of gradual increase in noise as described by Ziegler and Seliger.⁽⁸⁾ In the case of this tube, it was noted that switching the voltage off and on had no effect, once the noise

had built up. However, if the tube was removed momentarily from the socket and then replaced, the noise level was down to its lowest value but would again increase with time. Two physical operations were performed simultaneously on this tube; the H.V. divider was soldered directly to the phototube pins⁽⁹⁾ and a guard ring was painted on the outside of the tube. The hysteresis effect was eliminated and the noise pulse counting rate was reduced to 1/4 that ever observed for any other tube.

Other tests have been performed but are not completed; it is expected to report them in the future in greater detail.

C. Sources of radioactivity in photomultipliers

The radioactivity contained in the photomultiplier's component parts accounts for a sizable percentage of the gamma-ray background in the Human Spectrometer. Appropriate tests had demonstrated that the activity was located principally in the glass envelope and in the tube base, whereas the inner structure proved to be relatively clean. The serious handicaps presented by this radioactivity have been emphasized elsewhere.⁽⁹⁾

Dr. Bernard Linden, Allen B. DuMont Laboratories, supplied samples of the various glasses used in the tube envelope for the face plate, graded seal, stem and base. In addition, a sample of Kimball lime glass and a sample of lead glass were obtained from our stock room. The following results were obtained on the basis of radioactivity measurements.

<u>Date:</u>	<u>Sample</u>	<u>Per cent K by weight</u>
5/19/56	0080 soda lime glass	0.2 (plus a small amount of Ra)
	0012 soda lime glass	3.84
	7052 glass	2.4
	9010 glass	1.73
	Kimball lime glass	4.5
	Lead glass	4.5
	Tube basing compound	Considerable amount of K and Ra

Following the receipt of the above results, Dr. Linden submitted two additional samples of glass which, when counted, yielded the same net counting rate as sample No. 0080 above. However, the new samples contained about 10^{-13} c Ra/g of glass while the lime glass contained about 0.2 per cent K mixed with traces of Ra. The results on the above two samples were rather alarming in that they contained about the same proportion of radium as most sands.

In order to identify more closely the sources of radioactivity in the glass, samples of the different raw materials which were used in its manufacture were requested from DuMont. Dr. Sheldon and Dr. Hood of Corning Glass, via Dr. Linden of DuMont, sent samples of 3 sands and samples of 8 other materials which are used in the production of various glasses.

37

The results of these analyses, listed in Table 6, were sent to Dr. Linden and are discussed in DuMont Progress Report No. 23.⁽¹⁰⁾ Since the availability of this report is very limited and the matters discussed therein (lower half of page 8 and all of page 9) are pertinent to our subject, we take the liberty of reproducing in toto the above-mentioned portion:

... "The first three samples listed in Table 1 were samples of sand which are used in glass. The other samples, with the exception of the potting compound, are components which are included in the glass with the sand. It is of interest to note that ordinary sand has the lowest radioactivity of the 3 sands tested. Sand No. 6 which showed a very low background is sodium carbonate, which is used in the manufacture of ordinary lime glass. Sand No. 17, which showed an extremely high activity, has an antimony component. This component can be eliminated from the glass and it is estimated that this would result in an improvement by a factor of 2 at the most in the radioactivity of the glass.

"It was pointed out by Dr. Hood that in the manufacture of lime glass, a certain amount of feldspar is included as a source of aluminum oxide and silica. This feldspar contains some potassium. By elimination of feldspar in the glass approximately another factor of 2 may possibly be attained. Since ordinary sand makes up about 70 per cent of lime glass by weight, the most that can be hoped is to approach the basic activity level of this sand in glass itself. Another possibility, which is somewhat remote, is to find a source of sand which has a lower background than that supplied from Corning.

"This is the present status of the investigation into radioactivity background due to glass in multiplier phototubes. It is now up to the various users of these tubes in low level counting applications to decide whether an improvement by a factor of between 2 and 4 will help them enough to justify the cost of a special melt of glass."

Our reaction to Dr. Linden's statement is as follows: A reduction by a factor of 4 in the radioactivity of phototubes would be very welcome indeed. A typical 5 in. phototube adds about 200 c/m to the background of an 8 in. x 4 in. NaI crystal. The elimination of 150 c/m would be more valuable than the 15 per cent reduction of total background indicates because the majority of these events fall in the low energy region of the spectrum. Moreover, the absence of photopeaks (due to the elimination of feldspar and Sb_2S_3) would result in a smoother background spectrum and in a greater probability of identifying the remaining sources of contamination.

TABLE 6*

Specific radioactivity in samples of material used in the manufacture of glass

Sample	c/m from total sample	Activity	Weight	Activity/g or %/g
Ordinary sand	132.0 c/m	1.66×10^{-10} c Ra	1,005 g	1.66×10^{-13} c Ra/g
Special sand	187.6 c/m	2.3×10^{-10} c Ra	965 g	2.4×10^{-13} c Ra/g
Pulverized sand	171.7 c/m	2.1×10^{-10} c Ra	1,004 g	2.1×10^{-13} c Ra/g
Sand No. 7 (Sodium nitrate)	96.7 c/m	2.6 g of naturally occurring K	1,005 g	0.26% K
Sand No. 6 (Sodium carbonate)	essentially background		1,009 g	
Sand No. 14 (Al hydrate)	12.9	less than 0.184 g K	1,003 g	0.0267 K
Sand No. 17 (Sb sulfide)	1,031.0 c/m	in excess of 10^{-9} c of something?	1,031 g	
Sand No. 19 (Feldspar)	4,486.6 c/m	2×10^{-9} c Ra, + 11.6 g K	1,005 g	2×10^{-12} c Ra/g + 1.16% K/g
Sand No. 224 (Raw dolomite)	125.9 c/m	1.52×10^{-10} c Ra	1,090 g	1.4×10^{-13} c Ra/g
Sand No. 93 (Ca carbonate)	1,861.4 c/m	2.3×10^{-9} c Ra	1,005 g	2.3×10^{-12} c Ra/g
Sand No. 229 (Sodium sulfate)	36.7	$.45 \times 10^{-10}$ c Ra	1,021 g	4.5×10^{-14} c Ra/g
Potting compound	16.0	almost clean	949 g	

*Data transmitted to DuMont on August 12, 1956.

37

The fact remains, however, that quartz and vycor have been found to be radioactively clean and have been used successfully as windows on crystal cases; hence, the elimination of practically all radioactivity from photomultipliers could be accomplished by sealing either of these materials to a metal envelope. It is hoped that suitable techniques for accomplishing this will be developed.

Acknowledgment

The authors take pleasure in acknowledging the cooperation and assistance of Mr. J. E. Rose in many aspects of these problems; special thanks are due to Mr. R. Selman for skillful canning of some of our crystals.

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37

PROGRESS REPORT: RADIUM TOXICITY

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

The microdistribution studies of radium deposition in humans now include material from eleven subjects, three of whom received their burdens therapeutically, the other eight having been employed as dial painters for varying lengths of time.

In addition to the studies outlined in the previous progress report (ANL-5596, July 1956) the diffuse distribution of activity throughout the bone is being evaluated both qualitatively and dosimetrically and will be compared with the familiar hot-spot distributions.

The diffuse distribution is determined from autoradiographs made with NTA plates and no-screen x-ray plates, as well as with the usual stripping film technique. The loss of radon from the bone while it is being autoradiographed is under study for the various types of autoradiography employed. It is hoped that this effort will permit the correct evaluation of the dose received by the bone in vivo, from the number of alpha tracks the emulsions record.

40

POSITIVE ION EMISSION: PROGRESS ON THE MECHANISM

H. A. Schultz

Introduction

For gaseous diffusion studies of interest to the Laboratory, the halogen sensitivity of positive ion emitters can be utilized most effectively if the mechanism of operation is understood and controlled.

Transient Initial Ion Currents

The study of the mechanism has continued. An appropriate ion emitter⁽¹⁾ is operated in a reaction vessel containing a selected gas at atmospheric pressure. If the gas contains a trace of chlorine, a transient increase in ion current appears when the equipment is first started, but the effect of chlorine disappears rapidly.⁽²⁾

In order to study the reaction effectively, it is desirable to understand and control smaller but similar transient effects which occur in the absence of chlorine. The previous report gave an analysis of this problem and a summary of the progress that had already been made.⁽²⁾ Subsequent studies of transient ion currents have lead to two significant advances.

1. An improved method of inert gas (argon) preparation has been adopted.
2. It has been shown that when specially prepared gas is used, the transient behavior of the ion current is directly related to the transient thermal effects in the boat and its leads.

In the present argon-processing train, the gas is first passed through two dry ice-alcohol traps to reduce the water vapor content, then through a tube containing calcium chips heated to approximately 325° C. Calcium is well known for its "getter" properties, taking up all gases except those of the inert group.⁽³⁾ Good results were also obtained with calcium or barium "mirrors" inside a tube at room temperature, but it was necessary to regenerate these surfaces periodically by the evaporation of fresh metal from a filament.

In the study of positive ion sources it has been found that those which are sensitive to halogens in a reproducible manner are in general also sensitive to the temperature of the source of ionizable material. Consequently, in the design of the present boat and its associated equipment, the problem of temperature control has been given prime consideration. The boat has been made very light, with massive leads, so that its thermal time constant (heat capacity/rate of dissipation) will be small, and so that the leads will not serve as

41

auxiliary sources of ionizable material. The special power supply has been designed so that the resistance between the boat leads will be maintained constant, and therefore the mean temperature of the boat will be maintained essentially constant.⁽²⁾

Control of the mean temperature provides adequate control over the ion current under steady state conditions. However, the hottest part of the boat provides a disproportionately large share of the ionizable material, since the ion current is an exponential function of the temperature, and since the hottest part of the boat is nearest to the filament.⁽¹⁾ Accordingly, if the central temperature were to overshoot before coming to equilibrium, an overshoot in ion current would result, even though the mean temperature were held constant. The overshoot in temperature at the center of a luminous boat has been observed directly with an optical pyrometer.

The boat is nonluminous during ion current studies, and there is no convenient way to make temperature measurements on the central portion. However, it is comparatively easy to measure the temperature of one of its ends by use of a thermocouple. Experiments have established that the transient portion of the ion current and the transient portion of this temperature disappear in the same time interval, the ion current decreasing as the end temperature increases, so that the logarithm of the ion current is essentially a linear function of the end temperature. The decay is not a simple exponential function of time with a single time constant. The change in ion current corresponds to a change in effective central temperature approximately equal and opposite to the change in end temperature. The experimental data, coupled with the physical considerations outlined above, leave little doubt that there is a direct relation between the transient ion current and the transient end temperature. Heat conduction down the leads is the most probable cause.

The effect must either be eliminated, or be taken into account in subsequent work. One new boat design has been subjected to preliminary tests, with promising results.

Effect of Water

An investigation was undertaken to determine the effect of water as an additive to the dried argon in the reaction chamber. In the vapor phase, it had very little effect upon the ion current for relative humidities up to nearly 100% at room temperature. However, if a high concentration of vapor was allowed to remain in the chamber overnight while the equipment was shut down, a very large, slowly decaying transient effect was produced when the boat was turned back on. It required hours or even days to regain stability. This effect was due to the properties of the alkali compound in the boat. While the equipment was cool, the water vapor was collected by the alkali. If the filament alone was started, a normal background ion current was obtained. If the boat was started also, the salt disintegrated explosively as the water was released and shot out tiny particles of alkali, some of which adhered to the filament.

42

Thus, the effect of the boat was irreversible. Once initiated, the transient ion current persisted until all the excess alkali had been evaporated from the filament. The clean-up could be accelerated by raising the temperature of the filament.

Effect of Extended Range of Filament Temperature

It has been reported previously that the ion current is essentially independent of the temperature of the ionizing filament above a certain threshold temperature, provided the ionizable material is supplied by a separately controlled source.⁽¹⁾ Filament temperatures to 1250° C had been investigated.

Preliminary investigations at still higher filament temperatures, in the range to 1520° C, indicate that above a second threshold the ion current again increases with filament temperature.

This result is of considerable importance to the evaluation of the mechanism of operation. If further investigation confirms these early results, then one must reject the hypothesis that the plateau in the ion current vs. temperature relation is due to the ionization of every ionizable particle reaching the filament, and one must also re-evaluate other working hypotheses. Here, therefore, is a fertile region for investigation.

Participation in Air Pollution Symposium

A paper entitled "Determination of Gaseous Organic Halide Tracers in Air by Positive Ion Emission Techniques" was presented at the American Chemical Society Meeting, Atlantic City, September 20, 1956.⁽⁴⁾ This paper emphasized the desirability of a gaseous tracer,⁽⁵⁾ and showed that the positive ion technique was promising.^(6,7,8) It included a brief summary of the progress that has been made in the study of the mechanisms.^(1,9)

During the preparation of this paper, a simple method was sought by which all the data from a particular Freon field test might be used in the determination of values of Sutton's parameters C_y and C_z in his equation for the dilution of effluent from a source⁽¹⁰⁾

$$C(x,y) = \frac{2Q}{C_y C_z u x^{2-n}} \exp \left\{ \frac{-1}{x^{2-n}} \left(\frac{y^2}{C_y^2} + \frac{h^2}{C_z^2} \right) \right\} \quad (1)$$

The concentration is $C(x,y)$ at point (x,y) , x measured downwind from the source, y crosswind; Q is the rate of emission; u the average wind velocity; and h the height of the source. The dimensionless parameter n is determined from anemometer data using the equation

43

$$\left(\frac{u}{u_1}\right) = \left(\frac{z}{z_1}\right)^{\frac{n}{2-n}} \quad (2)$$

where u , u_1 are the average wind velocities at heights z , z_1 , respectively.

For the case of a ground source ($h = 0$), $\log x^{2-n} C(x,y)$ is a linear function of y^2/x^{2-n} . A graphical treatment of the data from field test No. 4(7,8) yielded values $C_y = 0.39$, $C_z = 0.23$ meters^{1/8}, about twice Sutton's recommended values for a ground source under "average conditions" $C_y = 0.21$, $C_z = 0.12$ meters^{1/8}. The goodness of fit is shown in Fig. 14, which was shown at the meeting. (Two errors were found by going back to the original recorder charts; 1.56 ppm should have been 1.00 and 0.10 should have been 0.16.)

Fig. 14. Atmospheric Diffusion test No. 4(7,8) Concentrations of Freon 12 in ppm obtained with source and sampling bottles 30 in. above the ground; emission rate 3.2 g/sec; sampling period 4 min. Values in parentheses were calculated from equation (1) using $n = 0.25$ obtained by equation (2) and $C_y = 0.39$, $C_z = 0.23$ meters^{1/8}, obtained by a graphical treatment of the experimental data.

For an elevated source ($h \neq 0$), an extension of the above method is suggested, using successive approximations. Several methods were explored briefly, using data from test No. 7, for a source at the 37.5 foot level of the meteorology tower. In this case, the concentrations were simply too spotty to be represented in a reasonable manner by a single pair of parameters C_y , C_z .(7,8)

40

Changes of Facilities

This project has been moved into a portion of the space formerly occupied by Dr. W. P. Jesse, in order to make the previous space available for other work. During the moving, several changes were made in the equipment, some of necessity, because of differences in the two rooms, others for convenience. Components which had been built into the old laboratory over a period of years were rearranged, as they were built into the new laboratory all at the same time. The gas-handling system was recleaned during the moving. After the equipment had been reassembled, satisfactory operation was attained readily.

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THE DISTRIBUTION OF CERIUM-144 AND THULIUM-170 IN THE DOG SKELETON

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

Cerium and thulium lie respectively near the beginning and end of the yttrium-like series of rare earths. Yttrium is deposited in a characteristic pattern in the skeleton, not associated with calcifying areas, nor yet with uncalcified organic matrix, but on bone surfaces.⁽¹⁾ Cerium and thulium may be expected to show a similar pattern of deposition inasmuch as they belong to the same series as yttrium, but may show slight differences since they are at different ends of a series.

Four puppies were injected intraperitoneally with either Ce^{144} (carrier-free) or Tm^{170} (low specific activity) as the chlorides, and killed 24 hours or 7 days after injection. The tissues of the body were analyzed separately for retained activity and the femur and tibia used to make macroscopic and microscopic autoradiographs.

The results of the analysis of soft tissue distribution of the two isotopes may not be compared too stringently since the thulium could only be obtained at a low specific activity (0.8 mc/mg), and the large amount of nonradioactive carrier tends to push the rare earth elements towards the reticulo-endothelial system,^(2,3) with the result that the liver retains a high proportion of the injected dose. A considerable amount of the thulium also remained at the site of injection. However, there are obvious similarities and differences between the metabolism of these two elements.

Both elements are excreted ineffectively in both urine and feces, only a little over 10% of the dose being removed in 7 days. Percentagewise the liver, gut, carcass and skeleton retain the most significant fraction of the dose at both 24 hours and 7 days after injection, but the retention per gram of tissue, or the specific activity, is highest in liver and skeleton, and 24 hours after injection, in the intraperitoneal fluid.

Analyses show an increase in specific activity of both isotopes in the skeleton 7 days after injection when compared with values at 24 hours. The additional activity comes from the liver, kidney and intraperitoneal fluid which all show a decrease in specific activity in this time interval. The average specific activity of the bones of the thulium animals increases by approximately 25 per cent but does not reach the value attained by the liver. Cerium appears to accumulate in the skeleton at the expense of the liver and intraperitoneal fluid to an extent which increases the specific activity of the bones by a factor of two, which brings the value to one above that for the liver. Although there is certainly a transfer of both elements

47

from the soft tissues to the skeleton, the difference between the amount transferred of the two elements may be merely a reflection of the relative amounts of carrier in the injection solutions.

An analysis of the retention of the two isotopes in the different bones shows that femur and humerus have a specific activity higher by 25 per cent than the tibia-fibula and radius-ulna, a difference which decreases with time. The rest of the skeleton has significantly lower specific activity than the long bones. These differences may possibly be explained by the relative proportions of bone to soft-tissue and marrow in the tibia-fibula and radius-ulna which are both divided, compared with the femur and humerus.

The autoradiographic studies have not yet been done; these will show the localization of the two isotopes, whether they both show a yttrium-like distribution, whether they are different in their distribution and whether or not they are retained by the "bone protein" a site of deposition suggested by some authors studying the metabolism of the lanthanum.⁽⁴⁾

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4

QUANTITATIVE AUTORADIOGRAPHIC MEASUREMENT OF CALCIUM-45 CONCENTRATIONS IN CORTICAL BONE

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

I. Introduction

The quantitative evaluation of the rates of calcification of Haversian systems and of periosteal and endosteal bone requires accurate measurements of the amounts of Ca^{45} present in microscopic regions of cortical bone. Some of the linear dimensions of these regions are comparable to the range of the beta radiation emitted by this nuclide, hence none of the simplifying assumptions applicable to either point sources or macroscopically homogeneous volumes of activity can be used with impunity. Similar difficulties have been met in the past by quantitative autoradiographic techniques of greater or lesser complexity, according to the nature of the problem involved. Thus, beta-particle autoradiograms were grain-counted by Nadler, LeBlond, and Bogoroch⁽¹⁾ in their investigation of the relative radioiodine concentrations in thyroid follicles of different size. A complicated but absolute method for quantifying beta-particle autoradiography has been described by Odeblad⁽²⁾ who uses a microphotometer to measure the darkening of autoradiographic plates. Odeblad calibrates his film with a beta-ray source in air and then calculates the effects of the beta-particle absorption, scattering, and distribution from the actual tissue sample. Dudley and Dobyns⁽³⁾ describe the use of a thick plaster of Paris radiator in the quantitative determination of radiation dosages from Ca^{45} in bone. The present method extends and applies the work of Dudley and Dobyns to the measurement of Ca^{45} concentrations in cortical bone. A partial application of this method to the study of bone grafts is in publication.⁽⁴⁾

The subject of this report is a method for the microanalysis of autoradiograms applicable to radioactive sources with activity concentrations which fluctuate widely in directions parallel to the plane of the emulsion but which remain unchanged in the direction perpendicular to it for distances comparable with the beta-particle range. Combined with serial section autoradiography⁽⁵⁾ the method provides a powerful tool for the analysis of Ca^{45} in cortical bone.

II. Method

The method involves:

1. Embedding a bone containing Ca^{45} in a medium with the same density as that of cortical bone (a mixture of Bioplastic and plaster of Paris).

49

2. Cutting a cross section with a thickness of 150 microns. Since within this range more than 95 per cent of the Ca^{45} beta rays are absorbed, this represents practically a "thick" section.
3. Autoradiographing both the top and bottom surfaces of the section using autoradiographic plates.
4. Microradiographing the section with X-rays at unit magnification.
5. Superimposing the two autoradiograms in turn upon a 100 X-enlarged tracing of the X-ray microradiogram using an Eastman Contour Projector. The autoradiograms contain sufficient detail to secure unambiguous alignment with the X-ray to within 10 microns, so that the structure containing the activity can be accurately identified. "Hotspots" or "hot areas" which do not appear identically on the two autoradiograms are dropped from the analysis. This procedure insures selection of deposits of activity invariant in the third dimension.
6. Analyzing both autoradiograms under a microdensitometer with a very small aperture ($30\ \mu$ in diameter).
7. Calibrating the densitometer with similar autoradiographic plates exposed to a thick plaster of Paris radiator containing uniformly-distributed Ca^{45} of known activity per gram of radiator. At least one calibrator plate is developed in the same tray with the autoradiograms.

Three features distinguish the present method from other quantitative autoradiographic techniques:

First, the source is thick to Ca^{45} beta particles. The advantages of a thick source are that film darkening* is independent of source thickness and this makes possible the following method of standardization.

Secondly, the film darkening produced by the source is compared with that produced by a radioactive standard material in contact with the same film. The radioactive standard is a uniform mixture of the beta activity with a known weight of plaster of Paris. No attempt is made to construct a model distribution with which to compare the source activity because such a complication is unnecessary in the present method. Instead, the plaster of Paris radiator is made thick to the beta particles in all dimensions in order to provide a direct measurement of the darkening produced

*Film darkening, $D = \log_{10} \frac{I_0}{I}$ where I = intensity of transmitted light.

"Darkening" will here refer to film, "density" to (g/cm^3) .

50

in a given film by a thick uniform Ca^{45} concentration in contact with it. This method of standardization avoids calculation of the absorption, scattering, or solid angle of the beta particles. Accurate calculations of electron penetration, as shown by the work of Spencer,⁽⁶⁾ are at best very elaborate even when applied to homogeneous media, and their extension to the boundary conditions presented by contact autoradiograms are as yet not feasible.

Thirdly, the concentration parameters for a certain class of non-uniform sources are obtained directly from the uniform source standardization by exact mathematical arguments. Let the rectangular coordinates x and y lie in the plane of a contact autoradiogram with the z axis extending through the source material perpendicular to the film. Then the Ca^{45} concentrations which can be measured by the present technique are those which can vary in an arbitrary way with x and y but must not vary with z for a distance below the film comparable to the beta particle range. Experiments with dogs using serial section autoradiography⁽⁵⁾ have shown that the Ca^{45} concentrations in most Haversian systems and in periosteal or endosteal surfaces extend a millimeter or more in the direction parallel to the axis of a long bone, whereas the maximum range of Ca^{45} beta particles in bone is only 0.3 mm. Therefore, in Ca^{45} autoradiograms of cross sections through the shafts of long bones, most Ca^{45} concentrations satisfy the above criterion of invariability with depth; those few which do not are eliminated by the comparison of the pair of autoradiograms (vide supra).

The Ca^{45} concentration parameters measurable by this technique are (1) the specific activity or the activity per unit volume for any distribution which extends uniformly in all directions, such as the diffuse activity in bone, (2) the total activity per unit length of Haversian systems, irrespective of the radial distribution of activity, and (3) the total activity per unit area of periosteal or endosteal surfaces irrespective of the variation of specific activity across the surface.

III. Derivation of s , the Activity per Gram of a Uniform, Extended Source of Beta Particles

The absolute calibration of the present method depends upon the comparison of the film darkening produced by a thick uniform source in bone with that produced by a plaster of Paris radiator. In order to elucidate the effect of density (g/cm^3) upon this comparison, let us assume that we have two materials with identical atomic compositions and equal, uniform specific activities, s (millicuries per gram). Let one have a uniform density ρ and the other a uniform density $\rho' = 2\rho$. As discussed recently by Failla⁽⁷⁾ the pattern of beta-particle paths in the first material is the same as that in the second material except for a change in linear scale of a factor of two. This "expansion" principle follows from the fact that the

51

absorption and scattering processes for beta particles are atomic in nature, so that the requirement of "identical atomic composition" insures identical angular distributions in scattering, and insures path lengths inversely proportional to ρ .

Consider the path of a typical beta particle when these two materials are placed in contact with an autoradiographic film (Fig. 15). In the material of density ρ the beta particle is emitted from a volume element at A, scatters at B, traverses the emulsion at C, backscatters in the glass plate above the emulsion at D, and traverses the emulsion again at E. For the material with density $\rho' = 2\rho$, the angles of scattering for the corresponding particle will be unchanged and the path segments within the material will be half as long: $(A'B') = 1/2 (AB)$, $(B'C') = 1/2 (BC)$. Similar relations will hold for every pair of corresponding particle paths in the two materials. Therefore the distribution of particles (angles and energies) incident on the emulsion for the first time, as at point C, is identical for corresponding volume elements in the two materials. A volume element of the first material has eight times the volume and four times the mass of a corresponding element of the second material. Since the specific activities are equal, the number of particles emitted from respective volume elements per unit area of emulsion is the same in the two cases (a linear scale change of two means a factor of four in area).

Fig. 15 Comparison of the paths of corresponding beta particles emitted from corresponding volume elements of two materials with identical composition but different densities.

When we trace the path of a particle through the emulsion or backing, the expansion principle clearly no longer applies: the path lengths $C'D'$ and $D'E'$ certainly do not vary inversely with the density of the source material but remain equal to CD and DE , respectively. Therefore the distributions of exposed grains produced by beta particles originating in corresponding volume elements of the two materials will not be related simply by a change in linear scale. However, the total number of grains exposed by a given distribution of initially incident particles will be a fixed property of the autoradiographic plate, and hence the total number of exposed grains will be the same in each case.

The particle flux incident on the film is the sum of the fluxes from every volume element within beta-particle range of the emulsion, each of which is proportional to the specific activity of the source material. Therefore the resultant film darkening for an extended, uniform source depends directly on its specific activity and is independent of its density.

Now what is the effect of the atomic composition of the absorber? To a first approximation, the flux of beta particles transmitted by a material depends only upon its superficial density (g/cm^2), so that the darkening effect of a uniform source may be expected to be roughly independent of the atomic composition of the source material. However, as shown above, the comparison of a source with a radiator will give an accurate result only if the probabilities of electron energy loss and scattering per atom are the same.

Let us compare the atomic compositions of bone and plaster of Paris, taking the weight of dry, fat-free bone to be 65% hydroxyapatite and the remaining 35% to be organic matrix and other inorganic materials. (Table 7). A plaster of Paris radiator with $\bar{Z} = 12.1$, is seen to have an atomic composition closely equivalent to that of bone $\bar{Z} = 11.8$. The composition of bone is not constant, however, the relative weight of mineral and organic matrix varying during the process of calcification. If the above figures are taken to represent full mineralization, then at 75% mineralization (the lowest mineralization that covers a significant area on a micro-radiograph), the average atomic number of bone becomes about 10.7.

Table 7

Approximate composition of dry, fat-free bone

		Z	Weight	Z x Weight
$\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ 65% by weight	Ca	20	26%	5.2
	P	15	12%	1.8
	O	8	27%	2.2
	H	1	0.1%	0.0
Organic and other C, H, O, Na, etc. 35% by weight		7.5	35%	2.6
Average Z =				11.8

Hardened plaster of Paris (gypsum)

$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$		Z	Weight	Z x Weight
	Ca	20	23%	4.6
	S	16	19%	3.0
	O	8	56%	4.5
	H	1	2%	0.0
Average Z =				12.1

52

What order of magnitude of error is introduced in the present analysis by an increase in Z from 10.7 to 12.1? We note that the effect of back-scattering from a thick source mount increases the counting rate about 2% in a 2π counter for the above change in Z .⁽⁸⁾ The energy loss per unit path length for electrons of 70 keV (approximately the average energy of Ca^{45}) decreases about 2% for the same increase in Z .⁽⁹⁾ From measurements of mass absorption coefficients for beta rays in different elements⁽¹⁰⁾ one may estimate that the mass absorption of plaster of Paris may exceed that of bone from 2% to 5% depending whether the bone is 100% or 75% mineralized, respectively. Therefore, pending a direct measurement to be made in the future, it seems likely that the differences between the atomic compositions of bone and plaster of Paris introduce an error of not more than a few per cent into the analysis for Ca^{45} activity. Film darkening due to a uniformly-distributed, thick, beta-particle source may therefore be written

$$D = k s F \quad (3)$$

where

D = darkening corrected for non-linearity and for film background

s = specific activity of the source (mc/g)

F = exposure time (corrected for radioactive decay, if necessary)
(in days)

k = calibration factor (darkening g/mc-day) .

From the argument above, the value of k for autoradiograms made with the same film, the same beta emitter, and the same exposure and development procedure will hold for both bone and a plaster of Paris radiator, or

$$s = \frac{D}{k F} \quad (4) \quad \text{and} \quad k = \frac{D^*}{s^* F^*} \quad (5)$$

where the starred quantities refer to radiator exposures and the unstarred quantities to bone. Expression (5) provides the desired relation between the specific activity of a uniform source in bone and the known specific activity of the radiator in terms of corrected densitometer readings.

Although film darkening is not strictly linear with exposure, a calibration curve can easily be constructed from measurements of various exposure times and specific activities which allows one to correct a densitometer reading for linearity.

IV. Derivation of the Activity per Unit Length of a Line Source

Consider a long thin filament of beta activity embedded in a material of density ρ and exposed end-on, at right angles to an autoradiographic plate (Fig. 16). We will call such a distribution of activity a line source, ascribing to it negligible thickness, a length greater than the beta particle range, and a uniform activity per unit length, h .

The developed plate will show a "hotspot," the darkening of which can be measured in the plane of the film as a function of radial distance from the peak.

Let H be the peak darkening value for the line source, and let the function $u(r)$ describe the decrease in darkening as the radial distance from the peak position increases,

$$D = H u(r)$$

$$\text{where } u(0) = 1$$

$$u(R) = 0$$

R = maximum range of beta particles

(6)

Fig. 16 Contact autoradiogram of a line source.

Now what is the relation between peak darkening H and activity per unit length h ? Since all densitometer readings are corrected for linearity with exposure, the value of H for any line source in a given medium must be directly proportional to the exposure time F and to the activity per unit length h . Let us try the expression

$$H = \frac{q h F}{\rho} \quad (7)$$

where H = peak darkening over line source

h = activity per unit length (mc/cm)

ρ = g/cm³ of embedding material

F = exposure time (days)

q = unknown factor to be determined .

We make no assumptions here as to the nature of q or its dependence upon the density or atomic number of the embedding material.

The relation between the unknown factor, q , for a line source and the uniform source calibration factor, k , which was derived in Section III, can be found by mathematically constructing a uniform source out of an assembly of line sources, holding constant the density and atomic composition of the embedding material. (See Fig. 17.) Consider the effect upon the darkening at a point P of many line sources, each with the same activity per unit length, h , evenly spaced at right angles to the film surface.

Fig. 17 Construction for generating a uniform source from an assembly of line sources.

Each line at a distance r' from point P will contribute a darkening of $Hu(r')$. If there are n line sources per unit area of film surface, the increment in the darkening at point P due to those lines between r' and $r' + dr'$ from P will be

$$dD = n H u(r') 2\pi r' dr' \quad (8)$$

Substitute for H the relation in (7) above and

$$dD = \frac{q F}{\rho} n h u(r') 2\pi r' dr' \quad (9)$$

56

Now let the number of line sources per unit area n approach infinity and h approach zero in such a way that the product $n h$ remains constant. The assembly of line sources will then become effectively a uniformly distributed source with specific activity $s = n h / \rho$. If we substitute s for $n h / \rho$ in (9) and integrate over the whole surface around P , we obtain

$$D = q F s \int_0^{\infty} 2\pi r' u(r') dr' \quad (10)$$

The only variable within the integral is r' , the radial distance from point P . Now if we substitute r for r' we obtain an integral with exactly the same value as that in (10):

$$D = q F s \int_0^{\infty} 2\pi r u(r) dr \quad (11)$$

but now the integral refers to a single line source with the profile function $u(r)$.

Comparing (11) and (4), both of which apply to uniform sources, we see that

$$q = k/A \text{ where } A = \int_0^{\infty} 2\pi r u(r) dr \quad (12)$$

Substituting (12) into (7) we obtain the desired expression for the activity per unit length of a line source embedded in material of density ρ :

$$h = \frac{H\rho}{kF} \int_0^{\infty} 2\pi r u(r) dr \quad (13)$$

Notice that the integral A is a measure of the area of the hotspot as seen on the film. If the hotspot were a perfectly resolved disk of radius b , $u(r) = 1$ for $r < b$ and $u(r) = 0$ for $r > b$, then the area A would be simply πb^2 . The actual A is a weighted average of area elements, the weighting factor being the function $u(r)$, namely, the profile of a hotspot due to a line source.

From the discussion of Section III, it is apparent that the line source profiles, $u(r)$, for materials of different densities will not be related simply by a change of scale, because the presence of the film disturbs the homogeneity of the medium seen by the beta particles.

In the above derivation D must be linear with beta-particle exposure. However, this requirement does not restrict the general nature of the derivation, for as long as film darkening is a single-valued, increasing

57

function of exposure, it is possible to associate a linear D value with each exposure. In other words, for D to be linear we need only assume that the actual film darkening is additive.

Note that the construction of a uniform source from an assembly of line sources holding density constant automatically takes into account the effects of angular distribution and energy spectrum of the beta particles upon the darkening of the film. A line source is the simplest element of a uniform source in contact with a plane film, so it must produce the same distribution of particle paths through the emulsion.

V. Activity per Unit Length of Arbitrary Distributions Generated from Line Sources (Haversian Systems)

Expression (13) can be generalized to apply to any distribution of activity which can be generated by line sources, that is, to any distribution which is invariant in the direction perpendicular to the film for a distance comparable to the beta-particle range. Rewriting (13) so that H appears inside the integral,

$$h = \frac{\rho}{k F} \int_0^{\infty} H u(r) 2\pi r dr \quad (14)$$

Rewriting (14) in rectangular coordinates, the activity per unit length for a line source becomes

$$h = \frac{\rho}{k F} \iint D(x,y) dx dy \quad (15)$$

where

$D(x,y)$ = the two dimensional darkening distribution for a line source
(identical with $H u(r)$) ,

The integration is over the whole film surface for which $D(x,y) > 0$, just as before.

Consider, as in Section IV, a large assembly of line sources, but this time as their number approaches infinity let the product of their activity and concentration vary with position on the film, dropping to zero eventually in each direction. The continuous darkening distribution thus generated will be the sum of the distributions due to each line source taken separately

$$D(x,y) = D_1(x,y) + D_2(x,y) + \dots$$

and the activity per unit length of the entire distribution will be the sum of the individual h values.

Therefore the activity per unit length of a two-dimensionally arbitrary distribution of activity is formally similar to (15):

$$h = \frac{\rho}{k F} \iint D(x,y) dx dy \quad (16)$$

where now, h = total activity per unit length in the entire distribution.

$D(x,y)$ = any darkening distribution which is invariant in direction z and is bounded by zero values in the $x y$ plane. If the darkening is not required to fall to zero, the integral will not converge. In other words, the darkening distribution must be resolvable from neighboring distributions and from the edges of the film or embedding material.

Expression (16) can be evaluated by adding a series of densitometer readings spread over the distribution (as shown in Fig. 18):

$$h \approx \frac{\rho(\Delta x)(\Delta y)}{k F} \sum D_{ij} \quad (17)$$

where

- h = activity per unit length (mc/cm)
- ρ = density of material (g/cm³)
- k = calibration factor from radiator (5)
- F = exposure time corrected for decay (days)
- Δx = horizontal spacing on the film between successive densitometer readings (cm)
- Δy = vertical spacing of readings (cm)
- D_{ij} = typical densitometer reading corrected for linearity and background.

The sum is over all D_{ij} values not zero.

Note particularly that the area (Δx) (Δy) that enters (17) is not the densitometer aperture. The only requirement on the aperture size or shape is that it be small enough so that the true film darkening does not vary significantly within the aperture. The same requirement must be made on Δx and Δy to insure an accurate integration.

The above requirement on aperture size stems from the fact that the light transmitted by the film depends exponentially upon film darkening. Therefore the average light intensity seen by the densitometer bears no relation to the average darkening if there is much variation of D within the aperture. For Ca^{45} distributions in bone it is found that the aperture diameter must be $\geq 30\mu$. With larger apertures the densitometer readings for hotspots become dependent upon aperture size.

57

Expression (17) is evaluated with a recording microdensitometer with micrometer indicators on the microscope stage.

Once a number of typical distributions have been measured as in Fig. 18, values of the summation can be plotted as a function of two or three quickly measurable parameters.

Fig. 18 Autoradiogram analysis for activity distributions that vary with x and y but are constant within betaparticle range in the z direction (perpendicular to film).

Expression (17) is being used for the analysis of Ca^{45} in Haversian systems, whose radial distributions of activity are not uniform enough or large enough in extent to be treated as uniform sources by expression (5).

60

VI. Activity per Unit Area of Surfaces (Endosteum and Periosteum)

The activity per unit area of a surface can be derived directly from expression (16). Let the active surface be perpendicular to the film.

(Fig. 19.) Take rectangular axes x and y in the plane of the film, and let the yz plane be parallel to the active surface. Then we may find the total activity per unit area in all yz planes of a distribution of activity which is arbitrary in the x direction but remains constant in the y and z directions for a distance comparable to the beta-particle range.

Fig. 19 Autoradiogram analysis for activity distributions that vary with x but are constant within beta particle range in the y and z directions.

Since by our choice of axes we have made the $D(x,y)$ values for such distributions independent of y , we may replace $D(x,y)$ in expression (16) by $D(x)$. Setting dy equal to unit length and dividing through by it, we obtain the total activity per unit area for the distribution,

$$L = \frac{\rho}{k F} \int D(x) dx \quad (18)$$

This is evaluated by adding a single row of densitometer readings (Fig. 19)

$$L \approx \frac{\rho \Delta x}{k F} \sum D_i \quad (19)$$

where L = activity per unit area (mc/cm^2)

Δx = spacing between successive densitometer readings (cm)

ρ = density of material (g/cm^3)

D_i = typical densitometer reading corrected for linearity and background

k = calibration factor from radiator (5)

F = exposure time corrected for decay (days)

Expression (19) is being used to obtain the total Ca^{45} activity per unit area deposited on a growing bone surface following a single injection of the isotope. Combined with the specific activity of the blood integrated over the period of the experiment, expression (19) provides a measurement of growth rate (cm/day for the advancing surface).

Summary

A method has been developed for the evaluation of concentrations of beta particle activity from thick section autoradiograms. The method is particularly applicable to the study of short-range beta emitters in bone. A cross-section of bone that is thick to the beta particles is sandwiched between two autoradiographic plates. Only those distributions of activity which produce nearly identical exposure patterns on the two plates are analyzed. The film darkening from these distributions is compared with that from a thick uniform plaster of Paris radiator with a microdensitometer. Simple relations between the activities and the exposure of uniform, one-dimensionally arbitrary, and two-dimensionally arbitrary distributions of activity are derived from the postulate that the distribution does not vary with depth within a beta-particle range from the surface of the film. The activity per unit depth for any distribution which satisfies this invariance requirement can be quickly obtained with an accuracy of a few per cent. The growth of Haversian systems, periosteal and endosteal surfaces, and diffuse calcification in bone is being studied with this technique.

62
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AN ANALYSIS OF THE SPECTRUM OF Ba^{133} WITH A VIEW TO ITS POSSIBLE USE IN THE DETERMINATION OF THE RBE OF ALPHA PARTICLES IN THE PRODUCTION OF BONE CANCER

J. H. Marshall

Comparison of the carcinogenic action of radium and strontium is complicated by the fact that not only the nature but also the range of the emitted particle is different. One can only speculate as to whether the differences in toxicity arise from the high specific ionization of the alpha particle as compared with the beta particle, or from the different dosage distributions in the neighborhood of hotspots caused by the short range of the radium alphas as compared with the strontium betas. A preliminary survey of α - and β -ray emitting heavy elements has been made by Marinelli.(1)

The suggestion is herein made that equivalent doses of Ra^{226} and Ba^{133} be compared in experimental animals. Barium lies between strontium and radium in the second column of the periodic table and therefore it is likely to show a practically identical pattern of deposition in bone following intravenous injection.* Furthermore, Ba^{133} is an efficient source of 45 kev conversion electrons which have a range of about 16μ in bone. Therefore, Ba^{133} deposited in a given location may be expected to irradiate a surrounding volume of bone no greater than that irradiated by radium α particles, but the specific ionization for the Ba^{133} electrons will be only $1/100$ of that for the alpha particles.

Ba^{133} decays to stable Cs^{133} with an 8-year half-life, so there would be no problem arising from buildup of daughter activities or from radioactive decay during a toxicity experiment with short-lived mammals.

Details of Ba^{133} Decay Scheme

The decay scheme of Ba^{133} shown in Fig. 20 is that given by Goldhaber and Hill(2) supplemented by Hayward, et. al.(3) and Langevin.(4) The gamma-ray energies are averages of those reported in References (3) and (4). The gamma-ray intensities per disintegration and the electron capture branching ratio are essentially those given in Reference (3).

*It should be noted, however, that experimental evidence on this particular point is very meager.

Fig. 20 Decay scheme for Ba^{133} (data from Xe^{133} decay included because there is a common level in excited Cs^{133}).

The internal conversion coefficient listed below (number of electrons/number of gammas) for the 81 keV gamma ray has been measured both from Xe^{133} and from Ba^{133} decays:

$\alpha_k = 0.3$	$K/L = 10$	(5)
$\alpha_k = 1.8$	$K/LM = 6$	(6)
Total $\alpha = 3.5$		(3)
$\alpha_k = 1.47$	$K/LM = 4.9$	(7,8)

The values $\alpha_k = 1.6$ and $K/LM = 6$ have been used in the following analysis. The internal conversion coefficient of the high energy gamma rays was estimated to be $\alpha = 0.02$.⁽⁵⁾ From the level assignments and energies shown in Fig. 20, the 295 keV gamma should be magnetic dipole with $\alpha_k = .037$, the 360 keV gamma should be electric quadrupole with $\alpha_k = .020$; these internal conversion coefficients were taken from the values calculated by Rose.⁽⁹⁾ For the gamma ray given as 65 keV in Fig. 20, the value $\alpha_k = 2.4$ has been measured.⁽⁴⁾

In Table 8, the measured and calculated conversion coefficients above have been combined with the decay scheme of Fig. 20 to give the electron and photon spectrum expected from 8-year Ba^{133} . This tabulation is only approximate and reflects the uncertainties of the experimental data.

In Table 9 the estimated electron and photon components of the Ba^{133} spectrum are summarized together with their ranges in bone, the energy per disintegration emitted on the average in each component, and the probable relative contributions to the total dose delivered to a mouse bone. The entire energy of the conversion electrons has been assumed to be expended in the mouse bone. The fractional absorption of the penetrating X and gamma radiation in mouse bone was estimated by assuming a 1 mm average photon path in a material of $Z = 13$ and density 2 g/cm^3 .

On the basis of the present estimate, the dose absorbed in the bones of a mouse injected with 8-year Ba^{133} would be approximately:

- 55% due to electrons with range $\leq 16 \mu$
- 20% due to electrons with range $25-50 \mu$
- 15% due to electrons with range $250-500 \mu$
- 10% due to diffusely-distributed photoelectrons from X and γ rays.

These values of fractional dose must be regarded as provisional estimates accurate only to within about 10 per cent of the total dose. This uncertainty arises from the spread in measured values of the internal conversion coefficient for the 81 kev γ ray, from ignorance of fluorescence yield for L X-rays, and from the dependence of K X-ray and gamma-ray dosage upon the assumed average path in mouse bone.

It is interesting to compare the above dosage distribution with that of Ca^{45} (pure beta ray-emitter with $E_{\text{max}} = 250 \text{ kev}$). From the shape of the beta-ray spectrum one finds the total dosage to be divided as follows:

<u>Fraction of total dose</u>	<u>Energies of electrons</u>	<u>Maximum range for electrons of given energies</u>
13%	0-50 kev	20μ
30%	50-100	$20-70 \mu$
33%	100-150	$70-140 \mu$
24%	150-250	$140-300 \mu$

From the standpoint of β -ray range, therefore, Ba^{133} does offer, as compared to Ca^{45} , the advantage of greater energy dissipation within distances comparable to α -ray range in bone. The main advantage, however, would seem to lie in its half-life which is long enough to eliminate the administration of supplementary Ca^{45} (half life ≈ 160 days) necessary to keep the

66
Table 8

Type of transition		Internal conversion coefficient* α , and fluorescence yield W				Number per Ba^{133} disintegration				Energy in kev		
		α_k	$\frac{\alpha_k}{\alpha_{LM}}$	Total α	W_k	Transitions	Photons	LM Electrons	K Electrons	Photons	LM Electrons	K Electrons
81 kev γ	M1	1.6**	6**	1.9	-	1.00	.35	.093	.557	81	77	45
65 kev γ	M1	2.4**	(6) ^{††}	3	-	.04	.01	.004	.026	65	61	29
295 kev γ	M1	.037 [†]	(8) ^{††}	.04	-	.30	.29	.001	.010	295	291	259
360 kev γ	E2	.020 [†]	(8) ^{††}	.02	-	.70	.69	.002	.012	360	356	324
K X-rays following internal conversion		-	-	-	0.88	.61	.54		.07 [†]	32		32
K X-rays following Ba^{133} K capture		-	-	-	0.88	.1**	.09		.01 [†]	32		32
L X-rays following Ba^{133} L capture		-	-	-	-	.9**				5		5

* α = no. of electrons/no. of gamma rays.

** Measured value or average of values.

† Calculated from level assignments (Fig. 20) and Reference (9).

‡ Reasonable values from Reference (10).

† K Auger electrons

Table 9

	Energy	Range in bone ($\rho = 2$)	Energy per disintegration	Fraction of energy absorbed in mouse bone* (approx.)	Contribution to the total energy absorbed in mouse bone*, %
<u>A. Short Range ($\leq 16\mu$)</u>					
K Conversion	45 kev	16 μ	25.0 kev	1.0	48.0
electrons	29	7	0.8	1.0	1.5
K Auger electrons	32	8	2.5	1.0	4.8
<u>B. Intermediate Range</u>					
LM Conversion	77	43	7.0	1.0	13.4
electrons	61	28	0.2	1.0	.4
L X rays (assuming $W_L = 1.0$)	5		4.5	1.0	8.6
<u>C. Long Range (250-500μ)</u>					
K Conversion	259	300	2.6	1.0	5.0
electrons	324	420	3.9	1.0	7.5
L Conversion	291	360	0.3	1.0	.6
electrons	356	480	0.7	1.0	1.3
<u>D. Penetrating Radiation</u>					
K X rays	32		20.2	0.12	4.6
Gamma rays	65		.7	0.02	.0
	81		28.4	0.01	.6
	295		85.5	0.006	1.0
	360		248.0	0.006	2.9
					<hr/> 100

*Mouse bone is assumed to have an effective Z = 13 and an average photon path length of 1 mm.

68

dosage constant throughout the duration of the experiment. It should be noted that repeated Ca^{45} injections, besides the purely additional labor required, introduce an additional uncertainty in the comparison, since for equal average dose, the pattern of dose distribution at the histological level would be a function of time and, moreover, different from that obtained with a single injection of a longer lived isotope of the same element.

Carrier-free Ba^{133} can be produced by deuteron bombardment of cesium through the reaction $\text{Cs}^{133}(\text{d}, 2\text{n})\text{Ba}^{133}$. The yield from the Argonne cyclotron for an 8-hour bombardment with 100 μamps of 14-20 Mev deuterons is about 0.1 millicurie. Much higher activities would be obtainable by pile neutron irradiation if highly-enriched Ba^{132} were made available. From the foregoing it is obvious that the economic aspects of the experiment may overrule other factors in determining the advisability of its performance.

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67

STUDIES ON BACKGROUND

H. A. May

I. Use of Gridded Ion Chamber; Chamber Background

The developmental work on a gridded ion chamber has been discussed in previous reports.⁽¹⁾ The mean chamber background, averaged over numerous counts made during this period, was found to be 0.17 ± 0.02 c/m for the energy region above 5.0 Mev. The energy spectrum of background counts is shown in Fig. 21, in the region from 4.0 to 6.0 Mev. The curve represents a total of 1574 counts accumulated in 126 hours of counting time. Considering the decay rates of probable contaminating elements, and the consistent findings of other workers that most metals contain traces of radium, it seems likely that the peaks are due to Ra^{226} ($E = 4.791$ Mev) and Po^{210} (radium F, $E = 5.20$ Mev). Since these built-in "sources" are indeed thick, it is rather surprising that any semblance of a line is observable; the possibility of surface contamination, however, cannot be ruled out completely.

COUNTS / HR.
per 200 kev channel

Fig. 21

observed in other chambers of comparable dimensions.⁽²⁾ We believe that it must originate in electrical noise or other extraneous sources not yet identified.

In the quantitative analysis of traces of ThC' (Po^{212}) in the presence of RaC' (Po^{214}), the background in the energy region from 7 to 9 Mev is the limiting factor. Fig. 21 indicates that the counting rate drops off monotonically with increasing energy, but the low counting rates have so far prevented us from obtaining a spectrum with reasonably good statistics. The observed distribution for one 23 hour run is shown in Fig. 22. The histogram includes a total of 42 counts, giving a mean counting rate of $0.030 \pm .008$ c/m within this energy range. In the same period there were 115 counts of energy greater than 9 Mev: This rate is about ten times greater than that

Fig. 22

1-inch Pt planchets, and flamed. The resulting thick samples were not expected to give very good energy resolution; nevertheless, analyses by members of the Chemistry Division revealed well-defined peaks at 5.1, 5.4 and 6.1 Mev. The offending isotopes are believed to have been Pu^{240} (5.162 Mev) and Cm^{242} (6.110 Mev).⁽⁴⁾

Decontamination of the chamber was attempted by scrubbing all component parts with a hot detergent solution. In addition, the assembly of Lucite insulators and brass field rings was placed in the lathe, and a light surface cut was removed from all exposed surfaces. The system of tubing and valves for evacuation and gas flow was thoroughly flushed also. This procedure was deemed successful, since the background count was reduced to about 0.35 c/m, namely a hundredfold. This was considered adequate for present use.

II. Low Background Gamma-Ray Counting

The background counting rate of two NaI crystals has been previously reported⁽⁵⁾ where it was shown that for a 4 in. x $1\frac{1}{2}$ in. crystal, 27 c/m were detected in the energy region 80 kev to 0.8 Mev in coincidence with pulses from a ring of GM counters completely surrounding the crystal and 1-inch Hg shield. These counts are believed to be of cosmic origin. Two experiments have been carried out to further determine the origin and nature of this background component.

In the course of a planned experiment on detection of gamma radiation from mixed transuranic elements in vivo, a number of small Pu, Am and Cm sources were obtained,⁽³⁾ with activities of the order of 10^5 disintegrations per minute. Pulse height analyses were done, mostly with collimators giving 1 per cent geometry, to establish true isotopic content. After this series of analyses was concluded, it was discovered that the chamber background count had risen to around 35 c/m for pulses exceeding 5.0 Mev. Sources of the contaminant were prepared by wiping the interior chamber surfaces with filter paper, which was then wet ashed, evaporated on

1. Plastic coincidence detector

The slow rise and time jitter of GM pulses introduces difficulties in coincidence gating of the 256-channel pulse height analyzer now available. Plastic scintillators have the advantage of fast rise time, but they present problems of efficient light collection if large structures are used for good shielding geometry. A "nut bowl" was turned from a piece of scintillating plastic⁽⁶⁾ and the external surfaces aluminized to aid in light collection. It was mounted in an inverted position, and the upper surface optically coupled to a 16-inch photomultiplier.⁽⁷⁾ All pulses above tube noise were used to drive a coincidence-anticoincidence gate, with the pulses from an 8" x 4" NaI crystal immediately below (and partially within) being analyzed. Essential geometry and dimensions are shown in Fig. 23. It was hoped that some structure might be demonstrated in the coincidence spectrum. Results were disappointing; the coincidence counting rate was less than expected. This may be due to either insufficient thickness of plastic resulting in pulse height low enough to fall into thermal photomultiplier noise, or poor reflectivity of the evaporated aluminum coating. Shortcomings of the latter have been noticed also by Rowland of our laboratory and are consistent with observations elsewhere; moreover the H.V. power supply for this tube was later found to be unstable.

Fig. 23 Plastic scintillator coincidence arrangement

1. DuMont 16" photomultiplier.
2. Lucite light pipe.
3. Scintillating plastic "nut bowl" in .040 stainless steel.
4. 8" x 4" NaI (T) crystal and quartz light pipe in stainless steel.
5. DuMont type 6364 5" photomultiplier.

2. GM tube coincidence setup

It was thought desirable to repeat the measurements of cosmic-ray background, previously reported on two crystals, $2\frac{1}{2}$ in. diameter by $2\frac{1}{8}$ in. and 4 in. diameter by $1\frac{1}{2}$ in., with a modified arrangement of the GM tubes. For measurement of human activity *in vivo*, a ring of GM tubes surrounding the crystal is awkward and impractical. This experiment was undertaken to see if equal coincidence shielding could be obtained from a layer of GM tubes outside the iron shield, covering only its top surface. Seventeen GM tubes, having active dimensions of 2 in. by 31 in., placed in parallel (both physically and electrically) were found sufficient for the task.⁽⁸⁾ The shield used was not the 7' x 7' x 6' room, but a smaller enclosure

72

with inside dimensions 32" x 14" x 14" having the same 8" wall thickness. Thus two possible factors are introduced against direct comparison with the previous data, namely, difference in number of low energy quanta due to backscattering from walls, and difference in background of the shield itself. Nevertheless, the data summarized in Table 10 reveal fair agreement for the smaller crystal. The much greater differences for the 4" x 1½" crystal remain unexplained.

Table 10

Background counting rate. Energy range: 120 kev to 760 kev - Cols. 1 & 3
80 kev to 800 kev - Cols. 2 & 4

	2½" x 2⅛"		4" x 1½"	
	(1)	(2)	(3)	(4)*
Background, 8" Fe shield	100 c/m		154 c/m	
Anticoincidence with GM tubes	76			
Difference	24			
Observed coincidence rate	24		28	
Background, 8" Fe plus 1" Hg	70	75 c/m	92	127 c/m
Anticoincidence with GM tubes	58		78	
Difference	12		14	
Observed coincidence rate	14	15	16	27

*See Ref. (5)

The data of Table 10 were taken with a coincidence-anticoincidence gate duration of about 10 μsec. This is the minimum required to operate reliably the 24-channel Marconi analyzer used. The gating length was increased to determine whether the finite time required for scattering and diffusion of meson-produced neutrons (or other secondaries) with eventual radiative capture (or inelastic collisions) in the shield is of any significance in production of observed backgrounds. Table 11 compares the coincidence rate for gate widths of 10 and 100 μsec obtained during a run of 1000 min.

At the moment, this increase in coincidence rate, though statistically significant, remains unexplained as to identity of the physical agent. It cannot be accounted for by chance coincidences. The usefulness or irrelevance of a more elaborate coincidence arrangement to the lowering of the background can be inferred by inspection of the table.

Table 11

(4" x 1½" Crystal, 8" Fe and 1" Hg Shield)

Energy Region	(1) 10 μ sec pulse coincidence (c/m)	(2) 100 μ sec pulse coincidence (c/m)	(3) Anticoincidence (100 μ sec) (c/m)	(4) Ratio <u>Col. 3</u> <u>Col. 2</u>
65-365 kev	17.9	19.2	68	3.9
365-830 kev	4.5	5.2	20	3.87
0.83-1.27 Mev	2.5	3.6	11	3.06
1.27-1.64 Mev	1.2	1.3	5.7	4.4
1.64 Mev and above	18.3	28.9	38	1.32
Total	44.4	58.2		

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7. Experimental DuMont tube.
8. We are indebted to Mr. James Gray of the Chemistry Division (ANL) for loan of GM tubes required for these experiments.

74

DRIFT VELOCITY OF ELECTRONS IN GASES

J. C. Bowe

The drift velocity of electrons moving under the influence of a uniform electric field has been measured in nitrogen and the rare gases helium, neon, argon, krypton and xenon as a function of E/P_0 , where P_0 is the gas pressure referred to 0°C . The gases were circulated through a hot calcium purifier prior to and during the time of measurement. An all-metal, helium-tight system was used. The method of measurement and description of apparatus are contained in prior ANL reports, 5378, 5456, 5518 and 5596.

The data are compared with the experimental work of R. A. Nielsen⁽¹⁾ and the theoretical work of Graham and Ruhlig.⁽²⁾ Nielsen referred the gas pressures to 20°C . In order to make the comparison consistent, his data were replotted to 0°C . This results in a translation of his curves in the positive direction of the abscissa by 7.3%. In all instances, Nielsen's curves lie higher than the present data. He used an electrical shutter method and a thermionic source of electrons consisting of a plane grid of tungsten filaments. The gas pressures at which data were taken is not given.

Graham and Ruhlig (hereafter referred to as G & R) calculated the cross section for momentum transfer using a set of phase shifts derived by Westin from existing experimental scattering data. Average values of drift velocity were then calculated, using these values of cross section and a Druyvesteyn distribution.

Experimental Results

1. Nitrogen

Prepurified nitrogen, obtained from Matheson Co., having a stated purity of 99.996%, was used in these measurements. The calcium purifier was operated at 350°C . After purification, the pressure was found to be less by a few mm, indicating that there was some absorption. Nitrogen is a good gas to use for the purpose of testing the method of measurement and calibration since the results obtained with this gas by various workers are in agreement. The present experimental results given in Fig. 24 are lower. The two curves can be brought into coincidence by translating the ordinates of Nielsen's curve 11% along the abscissa. This indicates that the discrepancy between the two curves lies in the determination of E/P_0 . It is difficult to see how such a large error can occur in the present measurements. A

theoretical curve, calculated on the assumption that the collisions are elastic and using Normand's⁽³⁾ measured values of cross section, falls below the experimental curve. At $E/P = 2$, the calculated value of drift velocity is about $1/2$ the measured value.

2. Helium

The results in helium are given in Fig. 25. Research grade helium obtained from Matheson Co. was used. A drift velocity curve determined immediately after the gas was admitted to the chamber, was identical to subsequent curves taken after the gas had been circulated over the hot calcium turnings at 450°C . If Nielsen's curve is moved 11% to the right, it will fall along the open circles. The theoretical curve of G & R is seen to lie lower. However, this curve can be brought into agreement by a translation along the abscissa.

3. Neon

In Fig. 26, the experimental and theoretical curves in neon are presented. Reagent grade, tank neon obtained from Matheson Co. was used and circulated through the purifier which was kept at 450°C . Above $E/P_0 = .15$, there is a marked discrepancy between Nielsen's data and the present data. Agreement with the theoretical curve of G & R is very good. The fact that this curve is also in reasonable agreement with one obtained using the Compton equation⁽⁴⁾ for drift velocity with the ordinary kinetic theory cross section supports the scattering data in neon and the subsequent calculations of the transport cross section. These results show that excitation and ionization are not in evidence below $E/P = 0.7$ as was believed by Nielsen and G & R.

4. Argon

In Fig. 27, the experimental results in argon are presented. The discrepancy between Nielsen's curve and the present one is not as marked as was formerly believed prior to converting Nielsen's data to 0°C . Tank argon of 99.999% purity, obtained from the Cleveland Wire Works, was used in these measurements. The curve obtained immediately after the gas was admitted to the chamber, bears no relation to the one obtained after circulation through the purifier. (See ANL-5518.) Here again, the present results are lower than those of Nielsen. The curves cannot be brought into coincidence by a simple translation. The calculated curve of G & R is in partial agreement over the region where comparison is possible.

Fig. 24 Drift velocity of electrons in nitrogen. The two curves can be brought into agreement if the experimental curve of Nielsen is moved 11% along the abscissa.

Fig. 25 Drift velocity of electrons in helium. The experimental curve of Nielsen and the theoretical curve of Graham and Ruhlig are shown for comparison. The open circles are obtained by moving the ordinates of Nielsen's curve 11% to the right.

Fig. 26 Results in neon compared with the experimental curve of Nielsen and the theoretical curve of Graham and Ruhlig.

Note: Limits of the estimated experimental error are indicated by the short horizontal bars, except where they are of the same order of magnitude as the point size.

Fig. 27 Drift velocity of electrons in argon compared with the experimental results of Nielsen and the calculation of Graham and Ruhlig.

Fig. 28 Drift velocity of electrons in krypton.

Fig. 29 Drift velocity of electrons in xenon.

Note: Limits of the estimated experimental error are indicated by the short horizontal bars, except where they are of the same order of magnitude as the point size.

5. Krypton and Xenon

The experimental results in krypton and xenon are presented in Figs. 28 and 29. These gases were reagent grade obtained from Matheson Co. The gases were each transferred into glass flasks, having a freeze-out finger containing activated charcoal. Drift velocity curves determined prior to purification were identical to results obtained with the purifier operated at 450°C.

Discussion

On the basis of a simple model, the electron drift velocity is given by

$$W = (e/m) \quad (l/u) \quad (E/P)$$

where e and m are the charge and mass of the electron, l is the mean free path, E is the electric field strength and P is the gas pressure. While this equation is not adequate to describe the experimental data, it is nevertheless useful in providing a qualitative understanding of some of the factors which determine the shape of the drift velocity curve. According to this equation, W is proportional to the product of the electric field strength and the time between collisions. It also says that W is proportional to the mean free path (inversely proportional to the collision cross section) and inversely proportional to the square root of the energy. On the basis of this energy dependence of W , one would expect the drift velocity curve to show a definite upswing to higher values when E/P is large enough so that an appreciable number of electrons have sufficient energy to excite or ionize the gas molecules. The effect of these types of collisions is to reduce the average energy of the electrons and thereby increase the drift velocity. This feature is verified when experimental curves are compared with theoretical curves calculated on the basis of elastic collisions only. It also explains why molecular gas impurities, having low lying energy states, cause the drift velocity to be larger in the impure than in the pure gas. In the region of E/P where excitation and ionization cannot occur, a larger collision cross section results in a smaller drift velocity. The dependence on collision cross section, however, is complicated by the fact that this quantity is energy dependent and varies sharply in the region of a Ramsauer maximum. Thus, whereas one might intuitively expect the drift velocity curves in the rare gases to be similar, differences can be expected in accordance with the variation of the energy dependence of the collision cross section from one gas to the next.

An interesting illustration can be found in the case of neon. According to the calculation of G & R, neon does not exhibit a pronounced Ramsauer maximum, but, rather, has a cross section which varies slowly with energy. Nielsen, G & R, attribute the discrepancy between experiment and theory as being due to the onset of inelastic collisions for E/P in the region of $E/P = 0.3$. Support for this conclusion is found in the energy measurements made by J. S. Townsend some 30 years ago, and in the small cross section for neon. However, the present measurements indicate that inelastic collisions do not occur at values of E/P as high as 0.7. The agreement between the experimental curve and the one calculated by G & R, taking into account only elastic collisions, is excellent. Moreover, the curves are in reasonable agreement with the one calculated by using the Compton equation and a constant value of cross section.

There seems to be no experimental or theoretical data with which to compare the results in krypton and xenon. The fact that for these curves W is low, indicates that these gases have large collision cross sections. The similarity of these curves to each other and to argon, indicates that the variation of the cross section with energy in these gases is similar. Likewise, the dissimilarity in the curves for helium, neon and argon reflect the differences in the energy dependence of the cross section in these gases.

One might be inclined to question the results obtained in an all-metal system inasmuch as it is known that electron drift velocity in the rare gases is sensitive to the presence of gaseous impurities. However, when measurements were repeated several days or even weeks after the purifier was turned off, and the gas was allowed to remain in the chamber, identical results were obtained. Whatever gases are liberated by the metal do not affect the drift velocity measurements. It is believed that the evolution of gases from the metal is negligible and that consistent results are obtained because the chamber was made gas-tight and no gas evolving organic matter or soldering fluxes were tolerated in the chamber.

A paper reporting these results will be prepared for publication in the open literature.

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METEOROLOGY

The Multiple Exposure Technique for Studying Atmospheric Diffusion

G. H. Strom* and H. Moses

This technique was developed by the senior author while he was in residence at the Argonne National Laboratory during the summer of 1955.^(1,2) In conducting these studies, multiple exposure photographs of smoke plumes were made with two cameras; one camera photographed a side view of the plume, and the other a bottom view. A total of 100 useable picture pairs was obtained during the summer of 1955, and an additional group of 7 useable pairs was obtained during the summer of 1956. Computations to determine the space co-ordinates of selected points on the plume have been completed for 100 film pairs; 50 have been performed on a desk calculator and 50 have been computed on the AVIDAC.

In a first analysis of these data, values for the parameters n , C_z and C_y in Sutton's diffusion equation were calculated using the assumption that the boundary of the smoke plume represents a constant percentage of the concentration at the center line. As indicated in previous reports, we now doubt the validity of this assumption. It was later assumed that the boundary of the plume may be represented by a constant concentration surface.

Using this assumption, an attempt was made to obtain a least squares fit of Sutton's diffusion equation to the multiple exposure data, thus determining the values of n , C_z , and C_y . An additional parameter was also sought, namely, the distance from the stack to a virtual origin. The assumption of a virtual origin is necessary since Sutton's equation assumes a point source and the expansion of the plume is very rapid within the first 5 feet owing to aerodynamic effects of the stack itself and the jet effect of the effluent.

The initial attempts to obtain a least squares fit to the data proved unsuccessful. It was therefore considered expedient to abandon this line of attack in favor of an analysis technique developed by the senior author during his return to the Argonne National Laboratory in the summer of 1956.

In this technique it was assumed that the visual plume profile represents a constant "line" concentration. The "line" concentration is defined by the integrals $\int_{-\infty}^{+\infty} X dy$ or $\int_{-\infty}^{+\infty} X dz$ where X is the concentration, y is

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5/

perpendicular to the plume and z is upward. In his initial studies he assumed that the line concentration at the plume boundary was 1 per cent of the line concentration through the plume axis at 100 feet from the virtual origin. This was followed by additional work assuming that the plume boundary represented a constant line concentration irrespective of the distance from the source, but a series of reasonable values for the parameter was assumed.

Values for Sutton's parameters, n , C_z , C_y , and the distance from the stack of the virtual origin, based on assumptions of constant line concentration are now being calculated on the AVIDAC by the Division of Applied Mathematics.

One of the primary aims of our research is to determine the relation between rates of atmospheric diffusion and meteorological parameters. The geometric configuration of the smoke plume, as described by Sutton's parameters, n , C_z , and C_y , provides a measure of both space and time rates of diffusion. During the past summer some preliminary determinations were made of Sutton's parameter values under the assumption that the plume profile represented a constant line concentration equal to 1 per cent of the maximum or plume-axis concentration at 100 feet from the virtual origin. The values for n , C_z , and C_y fall within the range reported by other investigators. These were compared to simultaneous measurements of the following meteorological parameters which are known to affect atmospheric diffusion:

1. Atmospheric stability which is measured by the difference in temperature between the 144-foot and 5.5-foot levels above ground.
2. The wind speed.
3. The value of " n " determined from anemometers located at 5 levels ranging from 79 inches to 150 feet.

In addition, values of n , C_z , and C_y , determined from the plume characteristics, were correlated among themselves.

The smoke plume measurements actually provided two values for n ; one from the photograph of the plume in the vertical plane (n_v), and one from the photograph in the horizontal plane (n_h). A third parameter derived from the smoke pictures was the slope of the plume boundary in the region of 10 to 100 feet from the virtual origin. These slopes were designated as D_v for the data of the vertical profile and D_h for the data of the horizontal profile.

The following are preliminary results obtained from these analyses:

1. The C_z 's and the C_y 's correlated only poorly with the stability values.
2. The n_h determined from the horizontal pictures correlated well with the n_v determined from the vertical pictures.
3. Values for C_z and C_y correlated only poorly with the values for n obtained from the anemometers.
4. There was a fairly good correlation between the average of the horizontal and vertical n 's determined from the smoke plume and stability.
5. Rather good correlation was obtained between atmospheric stability and the quantities $D_v U$ and $D_h U$, where U signifies wind speed. $D_v U$ and $D_h U$ may be considered as measures of the rates of vertical or horizontal expansion of the smoke plume.
6. There appeared to be a fairly good relationship between the ratio of $\frac{D_v}{D_h}$ and stability.
7. D_v and D_h correlated well with atmospheric stability.

Diffusion Studies Conducted by Means of the Type K-24 Aerial Cameras

Peter E. Wasko

Smoke Runs The following smoke runs with the 110-foot stack were made during August, September, and October using three type K-24 aerial cameras:

1. August 15, 1956	(two runs)
2. August 27, 1956	(one run)
3. September 24, 1956	(one run)
4. September 25, 1956	(one run)
5. September 28, 1956	(two runs)
6. October 5, 1956	(two runs)

In these runs cameras A and B (see Fig. 30) were positioned, one on each side of, and 200 feet from, the datum plane. The datum plane is the vertical plane in which the smoke plume was expected to be. The x-axis of a right-hand Cartesian co-ordinate system with the origin at

FIELD SET-UP FOR USE OF K-24 CAMERAS

Fig. 30

the center of the top of the smoke stack is in the datum plane. The A and B camera axes with elevation angles of 30° were in a vertical plane intersecting the datum plane at right angles, 80 feet from the smoke stack. The C camera was placed with its axis in the datum plane either 40 feet or 125 feet from the smoke stack. In 40-foot position, the axis of the camera is vertical, while in the 125-foot position, it is slanted 30° from the vertical. When the sun does not shine directly on the camera's lens, it is preferable to place the C camera in the 125-foot po-

sition. At the 125-foot location, the C camera has a range of view in the horizontal plane through the top of the smoke stack of about 110 feet in the x direction, whereas, at the 40-foot location, this range is about 80 feet. Moreover, with the C camera in the oblique position, the analysis of the C camera data with the Photogrammetric Plotter is somewhat simplified since the elevation angle of the projector's axis may remain the same as that used in analyzing data from the A or B cameras. In analyzing the data from the C camera pointing vertically upward, a reorientation of the projector's axis becomes necessary.

Geometrical considerations showed that in the case of the C camera with its axis in the oblique position, the plume must deviate by less than 20° from the datum plane in order to be photographed for the entire 110 feet. Also, for any deviation of the plume from the datum plane the plume could not be photographed by the C camera from directly below. A mathematical analysis showed that the overlay grid used on the A and B cameras could be used on the C camera with its axis in the oblique position. However, a numerical relabeling of the grid lines would be necessary.

Photogrammetric Plotter The images obtained on the projection screen, which consisted of crystolene prepared tracing paper (N 198LX, manufactured by the Keuffel & Esser Company) backed by a sheet of $1/8$ inch plexiglas, were at times quite diffuse. This difficulty was eased by the addition of a translucent beaded screen (type WDL, manufactured by the Klearcite Screen Company) to the papered side of the above screen. It was found, too, that outlining the plume on the negatives with china-marking crayon made it easier to find the edge of the projected plume, especially on over-exposed negatives. As a result, the speed of data reduction increased.

84

Data Reduction The smoke run in the present series selected for detailed analysis was the second run of October 5, 1956. Frame numbers 1-45, 46-76, and 77-102 were exposed, respectively, at time intervals of $3\frac{3}{4}$ seconds, $7\frac{1}{2}$ seconds, and 15 seconds. This procedure allowed for the selections of frames at integral multiples of $3\frac{3}{4}$ seconds and reduced the number of frames required to get an adequate sample at each time interval. However, oscillations in plume characteristics with a period equal to the period selected for analysis would be missed.

Data were read from the projection of the smoke plumes on the screen of the Photogrammetric Plotter. The plumes of the A and B camera films were analyzed at 5-foot intervals (except the first and second points from the stack which were at $2\frac{1}{2}$ -foot intervals) along the x-axis for some 30 points. Such small intervals were taken to insure that any changes in the plume's characteristics would not be missed. It was expected, too, that obtaining the data at constant time and space intervals would facilitate subsequent numerical analysis and machine computations. Sixty-eight plumes projected by the A and B projectors were reduced. The plumes as photographed by the C camera will be reduced next.

Computations The reduced data were submitted to the IBM Tabulating Section for computing essential diffusion characteristics. These computations are presently being programmed for the IBM computer, Type 650. December 17, 1956 was reserved for performing the first stage of these calculations at the IBM office in downtown Chicago.

When the calculations are complete, we should have information on the following:

1. The height of rise of the plume. The height of rise is given by the z-position of the plume's axis.
2. The thickness of the plume. The thickness is taken as the component of the z-difference between the top and bottom of the plume perpendicular to the plume's axis.
3. The downwind relative change of the plume's thickness per unit distance.
4. The indices of meander of the plume. These indices are:
 - a. The ratio of the distance from the smoke stack along the plume's axis to the projected distance of the plume's axis in a horizontal plane.
 - b. The ratio of the distance along the plume's axis to the distance along the x-axis.

5. The azimuth and elevation angles of segments of approximately 5 feet along the plume's axis.

6. The tilt of the plume about its axis. This is determined by the difference of the y-positions of the top and bottom of the plume.

7. The oscillation of the position of the plume's axis with time in spherical co-ordinates at fixed radii.

Relationship Between Visual Appearance and Concentrations of Smoke Plumes

F. I. Badgley* and H. Moses

The problem of relating the visual appearance of a smoke plume as determined by photographic techniques and the concentration of the material in and around the plume is of fundamental importance. A number of studies to determine the diffusion parameters using photographic techniques have been made both here and elsewhere.⁽³⁾ Data of this type would serve to put such studies on a more solid foundation. There are other practical facets for which data of this type would be useful. It is a familiar fact that the discharge from a stack may often be detected by an odor even when the visual discharge is far removed. Also, the seriousness of air pollution is often judged by the visual appearance of smoke trails while instruments measure the chemical content of the air and little is known of the relation between the two.

The senior author, while at Argonne during the summer of 1956, undertook this investigation, wherein the problem was attacked by emitting oil fog smoke and SO_2 from the same nozzle and attempting to measure the SO_2 concentrations at the boundary of the smoke plume.

Sulfur dioxide was measured by means of a Titrilog, manufactured by the Consolidated Engineering Corporation; and the visual tracer used was oil fog which was generated by a type M2 Army Smoke Generator. Sulfur dioxide was chosen as a tracer because equipment for detecting it was available; it is gaseous and, therefore, presents no settling out problem; it is cheap, and, with the Titrilog, can be released safely in concentrations large enough to be measured.

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The Titri-log used in these experiments was borrowed from the Northern Illinois Gas Company. This instrument depends on oxidizing sulfur to the hexavalent form by use of bromine as the oxidant. The bromine concentration in a liquid cell is kept at a constant level by means of the electrolysis of potassium bromide. Oxidizable sulfur introduced into the cell reduces the bromine content. This reduction is sensed by a pair of electrodes resulting in an increase of electrolyzing current to re-establish the proper bromine concentration. The magnitude of the required current is a measure of the oxidizable sulfur concentration. Compounds of sulfur other than sulfur dioxide cause interference but not in a manner serious to this study.

Sulfur dioxide was dispensed from 150-pound SO_2 cylinders through 3/4-inch galvanized pipe, a Fischer-Porter Flowrator to meter the flow, and a flexible rubber tube to the point where it mixed with the visible smoke. See Fig. 31. There was some chemical reaction present in the pipe and the tubing.

Fig. 31. Apparatus for studying relation between visual appearance and concentration of smoke plumes.

17

This would be serious for permanent installations but was no problem for this short term project. A standard U. S. Army type M-2 Smoke Generator was coupled to a 22-foot length of 3/4-inch pipe by means of a flexible metal tube. The pipe was well insulated with standard Magnesite pipe insulation so that the oil vapor produced by the generator would not condense in transit. At the end of the pipe the sulfur dioxide was mixed with the oil vapor in a simple nozzle made from a 3/4" x 3/4" x 1" tee fitting. The mixture, on escaping to the air, cooled and formed the dense white fog or smoke characteristic of the M-2 generator.

The entire pipe section was hinged at its lower end so that the discharge point could be swung up to 15 feet above the level of the smoke generator; legs were provided to hold it in this position. In practice, during the summer's work, this feature was not used, all smoke discharge was made with the pipe in lowered position, near ground level. See Fig. 31.

It was originally planned to sample the SO₂ and oil fog plume by the sampling bottles developed by Schultz and Dodd, similar to those used in sampling Freon 12.⁽⁴⁾ This technique was abandoned because it was found that the sulfur dioxide reacted with the rubber in the balloons and the stoppers. It was found that the sulfur dioxide, even in the absence of rubber, is not very stable and oxidizes fairly easily to sulfur trioxide, which is not detectable by the Titrilog. This oxidation proceeds at a rate which is dependent on the amount of sunlight and upon other variable constituents of the atmosphere which act as catalysts. Furthermore, adsorption of sulfur dioxide on glass proved to be a problem at low concentration. However, it did prove possible to develop a calibration technique for the Titrilog using polyethylene balloons. In this technique the sulfur dioxide mixture was introduced into the polyethylene balloons. The whole assembly was encased in aluminum foil to avoid any chemical action and the air was purified by filtering through activated charcoal before being mixed with the sulfur dioxide. Without these precautions, which can be used in calibration but not in sampling, the method becomes useless.

Because of these difficulties it became necessary to develop an entirely different sampling technique, i.e., one not involving balloons or storage problems. The method finally adopted involved pulling an air sample from the desired level above the ground through a one-inch diameter stainless steel and Tygon tube, using a small motor-driven suction generator. A portion of this sample was then bled directly into the Titrilog. This method is direct but is rather slow and clumsy when a large number of points need to be sampled. Using this technique, samples from ground level to 24 feet above the ground were obtained.

Pictures of the smoke plume were taken using the multiple exposure photographic technique described in a previous semiannual report.⁽⁵⁾ Pictures of the smoke plume for these studies were taken obliquely from

above at a position on the Meteorology Tower and from the side. During each test, which lasted 4 minutes, 16 instantaneous exposures were made on each of the films at 15-second intervals, giving a composite picture of the smoke plume from each viewing point. During the same period, Titrilog recordings were made continuously.

The following is a summary of the procedure which proved successful:

1. Oil vapor was produced by an M-2 smoke generator and led through insulated pipe.
2. Sulfur dioxide was mixed with the oil vapor and the mixture released into the air.
3. The resulting smoke plume was photographed.
4. The concentration of sulfur dioxide in and around the plume was measured by direct sampling into a Titrilog.

A total of about 25 runs were made using this technique. Because of the variability of the winds, it is likely that less than one half will provide useful data. These are now being worked on by the senior author at the University of Washington.

Fluctuations in Temperature

F. I. Badgley and H. Moses

In the course of measuring atmospheric stability--the temperature difference between two levels on the Meteorology Tower--and ambient air temperature, various patterns of short-period temperature fluctuations were observed. Of special interest were the amplitude minima in the temperature fluctuations which occurred most frequently on cloudless days shortly after sunrise and shortly before sunset. An example of such minima is presented in Figures 32A and 32B. The letters "a" and "b" in Figure 32A are placed near the temperature fluctuation minima. These periods of fluctuation minima have been called "siesta" periods--periods when the recorder pens are "resting."

An examination of the records reveals that the temperature fluctuations may range from less than 0.1°C to as much as 5°C . This is evident from Fig. 32A. The duration of the "siesta" period may vary from minutes up to an hour.

Fig. 32A. Ambient temperature
April 19, 1951 Station C-
Argonne National Laboratory

Fig. 32B. Temperature lapse rate
April 19, 1951 Station C-
Argonne National Laboratory

Both the temperature and temperature difference measuring equipment have relatively low lag. Copper-constantan thermocouples made of size 36 wire are used in conjunction with Brown Elektronik potentiometer recorders to obtain these data.

A second activity engaged in by the senior author during his stay at Argonne this summer was the investigation of these minima in temperature fluctuations. Both these which occur before sunset and after sunrise and others which occur during cloudy conditions were studied. To eliminate the possible effect of water in the thermocouples during precipitation, the study was confined to periods of no rain. Data for about one year were examined.

This investigation yielded a very surprising result, viz., that the minima in temperature fluctuations occur when the atmosphere is isothermal, i.e., the vertical temperature gradient is zero in the layer from 5.5 feet to 144 feet. A priori reasoning would have lead one to expect that "siesta" traces should occur when the vertical temperature gradient was adiabatic or .98°C per 100 meters. From theory, in an adiabatic atmosphere, parcels of air displaced vertically are always at the same

90

temperature as their surroundings. Our results, however, did show that the most frequent or modal vertical temperature gradient was the adiabatic lapse rate. The data were based on one minute averages.

The results of this study are summarized in Fig. 33. Several interesting features are depicted in this chart in addition to the minimum. First there appears to be a direct relationship between the amplitude of the fluctuations and the magnitude of the temperature gradient under unstable conditions. Second, during inversion conditions the fluctuations are relatively insensitive to changes in temperature gradient. Further investigation is needed to explain these phenomena.

Fig. 33. Fluctuations in lapse rate as a function of lapse rate.

Climatology

H. Moses

The report on 5 years of meteorological measurements entitled, "Five-Year Climatological Summary--July 1949 - June 1954," which is to appear as ANL report No. 5592, has been completed and is now in press. As indicated in the previous semiannual report, this climatological summary is divided into three parts: In the first part are presented data concerning single meteorological variables; in the second are given data showing the relationship between two or more weather elements; and in the third, a comparison is made between the meteorological measurements at the Chicago Midway Airport and those at the Argonne Meteorology Laboratory.

This five-year report is rather extensive; it consists of 348 tables and 21 figures. It is believed that this work will serve as the useful tool in planning operations by engineering and scientific groups of the Laboratory, especially where problems of atmospheric diffusion or air pollution are concerned, and will also serve as a useful body of data for climatological research.

With the increase in the number of reactors built in areas surrounding cities, meteorologists and others will have to use standard Weather Bureau observations to ascertain the meteorological features in the vicinity of the reactor sites. The information contained in this report will be useful as a guide to indicate the degree to which measurements in an urban area such as those at the Chicago Midway Airport correspond to those at a nonurban location 10 miles away. Significant differences were found to occur in some cases. A summary of some of the results of the third part of the report are presented below:

1. Temperature From Fig. 34, it is apparent that a significant difference in average temperature is present. The average of 5 years of hourly observations shows that the Chicago average was 2.8°F higher than that of the Argonne Meteorology Laboratory. The data also show that at times, though rarely, the temperatures differed by as much as 10°F.

2. Relative Humidity A frequency distribution of the differences in relative humidity is shown in Fig. 35. These data show that the dispersion in the relative humidity differences is greater than that of the temperature differences. The average relative humidity difference based on 5 years of data at the two places was 6.3 per cent--the average relative humidity was higher at Argonne.

92

TEMPERATURE AT CHICAGO MINUS TEMPERATURE AT ARGONNE
FREQUENCY DISTRIBUTION OF DIFFERENCES IN TEMPERATURE
BETWEEN THE CHICAGO MIDWAY AIRPORT AND THE ARGONNE
METEOROLOGY LABORATORY

CHICAGO RELATIVE HUMIDITY MINUS ARGONNE RELATIVE HUMIDITY (In per cent)
FREQUENCY DISTRIBUTION OF DIFFERENCES IN RELATIVE HUMIDITY
BETWEEN THE CHICAGO MIDWAY AIRPORT AND THE ARGONNE
METEOROLOGY LABORATORY

Fig. 34

Fig. 35

CHICAGO WIND SPEED MINUS ARGONNE WIND SPEED (MPH)
FREQUENCY DISTRIBUTION OF DIFFERENCES IN WIND SPEED
BETWEEN THE CHICAGO MIDWAY AIRPORT AND THE ARGONNE
METEOROLOGY LABORATORY

Fig. 36

3. Wind Speed The wind speeds curve (Fig. 36) also shows a fairly large dispersion. At times the differences in the speeds between the two places were appreciable. The average difference based on 5 years of data shows that the wind speed at the Chicago Midway Airport was higher by 2.1 miles per hour. This difference in part may be explained by the difference in the elevation between the two anemometers. The anemometer at the Argonne Meteorology Laboratory was located at 18.75 feet while the one at the Chicago Midway Airport was located at 37.75 feet above ground.

4. Wind Direction Frequency distributions of wind direction at the Chicago Midway Airport for observations of the cardinal wind directions and calm conditions at the Argonne Meteorology Laboratory are presented in Fig. 37. These data show that there is a relatively high percentage of observations with the wind from one direction at the Argonne Meteorology Laboratory and from an appreciably different direction at the Chicago Midway Airport.

Fig. 37. Frequency distribution of wind direction at Chicago Midway Airport for indicated wind directions at Argonne Meteorology Laboratory.

In the 5-year climatological report the data are presented in much greater detail; for example, there are separate tabulations for each group of like months, that is, 5 January's, 5 February's, etc. Also, there are monthly frequency distributions for each hour of the day. A variety of other types of information is presented. The information presented here is merely a general summary.

Automatic Data Processing System

H. Moses

During the previous year automatic data processing methods for use with the continuously operating meteorological equipment were investigated. Instead of purchasing a complete system at once, it was decided to purchase equipment that would automatize only one portion of our meteorological instrumentation. This will allow us to gain experience with this kind of equipment and will aid us in the selection of the remaining sections. It was decided to begin with the automatic processing of the soil temperature data. This choice was made because the soil temperature equipment requires no averaging and no compensation for nonlinearity. Specifications for a system which would punch the soil temperature measurements on paper tape have been written; it is expected that the bids will have been solicited before January 1, 1957.

A. Soil Temperature Measuring Equipment Soil temperature data are obtained for 8 different levels beneath the ground surface. Leeds and Northrup 100-ohm copper thermohms are used as the thermally sensitive elements and feed into a Leeds and Northrup Micromax Recorder located in the Meteorology Building. The soil temperature data from the different levels are programmed by means of a stepping switch operated by a motor driven cam actuating a micro switch. Soil temperatures are measured at the following levels below the ground surface: 1 cm, 10 cm, 20 cm, 50 cm, 100 cm, 10 feet, 29 feet, and 100 cm at a second location.

B. Components of the Automatic Data Processing System The data processing system will consist of the following components:

1. Analog-to-Digital Converter This will either consist of a disc type encoder mounted on the slide wire contactor shaft of the Micromax recorder or an apparatus which will convert the resistance values of the thermohms directly to a digital output.

2. Translator Unit This unit will translate the output of the Analog-to-Digital Converter to provide suitable impulses to operate tape punching and printing apparatus.

3. Tape Punch A conventional tape punch or combination of printer and tape punch will be used. The tape will be 8-channel chad-type paper tape.

4. Programmer and Timer This unit will switch in the necessary circuits for tape-punching and printing the codes, date-time groups, and soil temperature data.

5. Control Panel The control panel will contain a series of push buttons and switches for selecting the year and month, and other coded information, and for regulating such functions as resetting the date, perforating the tape at will, and inserting necessary information into the tape. Indicator lights will also be incorporated to indicate whether recording is in process, whether the power is on, or whether a power failure had occurred since the system was last monitored.

C. Soil Temperature Information The programming of the soil temperature information will be in accordance with the following schedule:

<u>Program position</u>	<u>Soil temperature level</u>	<u>Time of measurement minutes past the hour</u>
1	1 cm	1
2	10 cm	3
3	20 cm	5
4	50 cm	7
5	100 cm	9
6	10 ft	11
7	29 ft	13
8	100 cm (second location)	15
9	Reference	17

Argonne Meteorological Model Towing Tank

Paul Frenzen

Introduction

In recent years, G. H. Strom of New York University has demonstrated that the problem of stack dispersal of waste gases can be successfully investigated by means of laboratory models.⁽⁶⁾ Results of sufficient reliability to dictate desirable design features for steam-power plant installations have been obtained in a low velocity wind tunnel. Similar wind tunnel studies of atmospheric diffusion phenomena have been made by others, including R. H. Sherlock and his colleagues, at the University of Michigan. The method brings into play all of the advantages of laboratory

16

experimentation over natural observation; it enables the response of a model system to be studied over a large variety of controlled conditions before the design of a prototype is "frozen" and funds are committed for its construction.

It is evident that such preliminary study of the design of large, radioactive gas emitting installations will become increasingly important. For although modern reactors are essentially sealed off from the outside atmosphere, chemical reprocessing plants required for the economical utilization of nuclear fuels are not. And these, even if located in remote areas to minimize exposure of the general public, should be designed to avoid local concentrations of waste radioactive gases in order to protect their own personnel. Since a major portion of the activity arising from the storage and processing of irradiated fissionable materials is in the form of isotopes of noble gases (Kr^{85} and Xe^{133}) which cannot be isolated by anything other than expensive low temperature adsorption techniques, the problem of stack dispersal will be especially important.

As an alternative to wind tunnel studies, trials of a water channel have begun at ANL. Water immediately presents an advantage over air as far as Reynolds number similarity considerations are concerned; for since the kinematic viscosity of water is one order of magnitude smaller than that of air, the same degree of dynamic similarity* can be obtained in water with only one-tenth the velocity-length scale product required in air. Compressibility effects are not important since the Mach number appropriate to the prototype remains small, i.e., wind velocities in the atmosphere are considerably less than the speed of sound.

Experimental Equipment

The equipment has been simplified by designing the facility as a towing tank (see Fig. 38) instead of a continuous flume. Models (effluent sources, turbulence-producing screens, etc.) are to be towed through water initially at rest rather than to remain fixed in a moving stream. This considerably reduces the cost of the installation to probably something less than five per cent of that of wind tunnels which have been used for similar experiments. However, it limits the types of experiments which can be done by introducing an error in the model's lower boundary condition. The vertical shear is zero whereas in the lower layers of the atmosphere the velocity of the wind usually increases approximately logarithmically with height. Certain modifications planned for later experiments which will make possible the modeling of more realistic wind profiles are described below.

*Exact Reynolds number similarity in meteorological models is neither attainable nor, fortunately, necessary; the point here is the degree of similarity attained.

97

Fig. 38. The Argonne meteorological model towing tank. The towing carriage is shown on the rails in running position; the driving mechanism is visible on the floor below. Scale can be estimated from the intermediate legs which are three and one half feet long and five feet apart.

The completed tank is thirty feet long, twenty-six inches wide, and eighteen inches deep. At the normal operating depth of twelve inches, it contains about five hundred gallons of water. It rests upon two five-inch steel channels, thirty feet long, which are supported on edge by seven "H" frames built of Unistrut verticals and steel channel cross members. The tank proper has half-inch plate glass sides and a half-inch aluminum bottom thermally isolated by micarta pads from the steel channel frame. Virtually the entire bottom of the tank is accessible from below for the eventual installation of heating or cooling elements. Aluminum was selected for its corrosion resistance and heat conduction and the plate glass sides to facilitate observation in addition to providing a measure of thermal insulation. All joints were sealed with a synthetic, rubber compound (3-M Brand EC-801 plus 807 hardener).

Cold-rolled steel rails along the full length of the upper edges of the tank provide a track for a five-foot long carriage from which various models can be suspended. The carriage is driven by an endless, stainless steel control cable which passes completely around and under the tank. Below one end of the tank it winds around a spirally grooved driving sheave geared to an electric motor-hydraulic transmission combination. Continuously variable speeds up to five feet per second are possible with the present arrangement; this range may be readily changed by a simple substitution of gears. The speed control lever of the hydraulic transmission is automatically and smoothly returned to the null position by an adjustable dog which can be set to operate at any point during the passage of the carriage along the rails. Thus a model, after being drawn at full speed for almost the entire length of the tank, can be smoothly and positively decelerated to a stop in the last few feet. A war-surplus Vickers model AA16801A transmission is used. It is driven by a half-horsepower, 3450 rpm capacitor start-induction run motor, and delivers plus or minus four hundred rpm continuously variable through zero at the output shaft.

Quantitative observations will be made photographically. For the purpose, a 35 mm Robot Royal camera is being equipped with a solenoid tripping arrangement for remote operation. The Royal offers the additional advantage of burst operation, automatically taking as many as 24 frames in succession at speeds up to 8 frames per second. This feature will be used to study transient phenomena such as turbulent decay rates. In general, two kinds of photographs are planned, i.e., short-time exposures of the motions of small particles suspended in the water (streak photographs) and successive instantaneous exposures of the paths traced by plumes of ink from various sources. Of special interest will be the comparison of both kinds of photos taken in the same experiment, for here the diffusion patterns can be directly correlated with simultaneous, quantitative measurements of the turbulent field.

Experiments Planned

Early experiments will aim at developing internal illumination and tracing techniques for streak photographs. A study of the rate of decay behind rectangular meshes is planned, not only because of its simplicity but also because there is available in the literature a large body of similar measurements in wind tunnels to which these results can be compared. Further, these trials will be preliminary to the study of decay rates in stratified fluids. At an early date an ink source will be added "downwind," and techniques for observation and measurement of diffusion will be studied.

It is probable that useful engineering data from model buildings with stacks will not be obtained without correcting the wind profile condition mentioned above. Several methods of accomplishing this have been planned. In the first, a ten-foot-long aluminum plate will be suspended below the carriage to provide a moving floor on which to base the models. Roughness elements of various kinds will be added at the upstream end of the plate to speed the growth of the boundary layer necessary for the desired wind profile. Under nonstratified (lapse) conditions when neutral equilibrium prevails, this method will be especially simple, for then the plate can be hung just below the top surface of the water with models and roughness elements mounted upside down on its lower face.

A second method creates a longer fetch over which the wind profile can be established and in which the experiments can take place. The entire lower boundary would be made to move with the models and roughness elements by installing a wide belt (standard aluminum window screen) moving along the bottom of the tank. Secondary circulation would be minimized by installing false end plates in the tank separating the regions of belt entrance and exit from the main body of water. Continuously supplying some water with the belt as it enters and removing it as the belt exits would minimize the disturbances propagated through the openings at the bottom of the end plates through which the belt passes. However, this latter refinement may not be necessary because of the short life of each individual experiment.

The third modification would convert the towing tank to a continuous flume by installing a twenty five-foot-long false floor at the half-depth level. Driving propellers, deflecting vanes, and stilling meshes and screens would be installed in the resulting loop. Something between fifteen and twenty feet of one leg would be available for a test section; however, at best it would be only nine inches deep.

The results of measurements of decay rates behind screens in a stratified fluid will probably be of considerable theoretical interest. Efforts to carry out this kind of study in wind tunnels and liquid flumes meet with difficulty in establishing and maintaining vertical density gradients in

continuous flow. In this respect, the model towing tank has a distinct advantage since the density gradient (stable or unstable) can be established in a fluid at rest. Then, after the passage of the mesh has set up turbulent eddies, successive streak photographs taken by a camera, in a fixed position relative to the tank, but farther and farther downstream relative to the moving mesh, will record the turbulent decay (or growth) while the vertical eddy exchange proceeds to wipe out the density gradient.

Acknowledgements

The assistance of Prof. Gordon H. Strom in the initial design of the tank and of Donald F. Uecker in the design of the driving mechanism is gratefully acknowledged. The tank was largely constructed by Eugene K. Gutowski, Robert C. Sablotni, and Lawrence O. Nippa of the Physics Shop, while Alois J. Bragagnolo and Richard F. Selman of the Radiological Physics Staff Shop assembled the transmission and carriage.

Soil Temperature Studies

H. Moses

Information on soil temperature is of interest to the Meteorology Group, since the interchange of energy between the soil and the adjacent air affects atmospheric diffusion. These studies have been given added emphasis during the last six months by the collaboration of Professor James E. Carson of Iowa State College. During the summer of 1956, he spent one month in residence with the Meteorology Group at Argonne under a research agreement.

Professor Carson has concerned himself with such problems as the relations between soil temperature and weather conditions, the study of heat flow in soils, and checking theoretical models proposed by other investigators on the mechanisms involving heat flow in soils. He chose our data for use in his investigations since the soil temperature measurements we have, which were made simultaneously with the battery of meteorological information, provide one of the best sources of data of this type available. His studies based on our data are confined to the period of measurements from November, 1952 through December, 1955. A number of tabulations of data, which would be of interest both to Professor Carson and to the Meteorology Group, have been made by IBM techniques.

One type of investigation involves the determination of thermal diffusivity values in the soil. Thermal diffusivity is defined as the ratio of molecular conductivity to the product of soil density and specific heat.

101

Thermal diffusivity may be calculated in two ways from plots of soil temperature versus time for different levels below ground. One method involves the phase lag between the soil temperature waves and the other depends on the attenuation of the soil temperature amplitudes with depth. To carry out these measurements the soil temperature data were divided into groups of 10 days each; these were called decade groups. Decade averages of soil temperature values for each depth and average hourly values within each decade for the three-year period under investigation have been calculated by IBM techniques.

Another type of study involved the determination of average hourly differences in temperature between the air and the soil at various levels. A sample of the latter type of study is shown in Fig. 39.

Fig. 39. Average hourly temperature difference between air at 5.5 ft. above ground and soil at 1 cm below ground.

Curves showing the average hourly differences in air and the soil temperature at 1 cm are presented for a winter month, January; a summer month, July; a spring month, April; and an autumn month, October. It is of interest to note that in general the soil is warmer than the air. Also, in the diurnal variation of this temperature difference, a double peak occurs in three of the months, April, July, and October. The reasons for this double peak are not immediately obvious.

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103

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105

STAFF ACTIVITIES

Gordon Research Conference on Chemistry, Physiology and Structure of Bones and Teeth, Kimball Union Academy, Meriden, New Hampshire, July 16-20; R. E. Rowland, J. H. Marshall and J. Jowsey, Participants.

American Chemical Society Meeting. Atlantic City, New Jersey, September 17-21: "Determination of Gaseous Organic Halide Tracers in Air by Positive Ion Emission Techniques," by H. A. Schultz; "Measurement and Interpretation of Total Alpha and Beta Activity in Water," by J. Sedlet and A. F. Stehney.

American Meteorological Society Meeting, Chicago, Illinois, September 18: "Project Prairie Grass and the Great Plains Turbulence Field Program," by H. Moses.

Central Society of Nuclear Medicine Meeting, Hines Hospital, Hines, Illinois, September 23: "A Case of Accidental Skin Puncture Contaminated with Th^{227} ," by P. F. Gustafson, L. D. Marinelli and E. A. Hathaway.

American Chemical Society, Chicago Section Meeting, Chicago, Illinois, October 19: Bio Chemical and Special Activities Group Meeting - "The Spectroscopy of Gamma Rays Emitted by the Human Body," by L. D. Marinelli; A. F. Stehney, discussion leader.

Radioiodine Conference (AEC sponsored), Argonne Cancer Research Hospital, Chicago, Illinois, November 5-6: L. D. Marinelli, Chairman of Radiobiological Session.

Armour Research Foundation Seminar, Chicago, Illinois, November 16: "The Spectroscopy of Gamma Rays Emitted by the Human Body," by L. D. Marinelli.

American Physical Society Meeting, Chicago, Illinois, November 23-24: "Drift Velocity and Average Energy of Electrons in Noble Gases," by J. C. Bowe.

Radiological Society of North America, Chicago, Illinois, December 2-7: "The Values of W for Gases of Dosimetric Interest," by L. D. Marinelli; "Analysis of the Cs^{137} Content in Humans Due to Radioactive Fallout," by C. E. Miller, L. D. Marinelli and O. J. Steingraber; L. D. Marinelli also participated in a meeting of the Commission of Radiologic Units, Standards and Protection of the American College of Radiology.

106

Subcommittee meetings on Internal Dose of the National Committee on Radiological Protection and the International Commission on Radiological Protection, National Bureau of Standards Building, Washington, D. C., December 10-11: L. D. Marinelli and J. E. Rose participated.

Air Pollution Research Planning Seminar, Robert A. Taft Sanitary Engineers Center, Public Health Service, Cincinnati, Ohio, December 18-20: H. Moses participated.