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Errata

/ Page 10, paragraph 4: for HF read HL

/ Page 17, Table 6: 3rd and 4th columns should read $\text{cm} \pm E$

Page 19, formula for R_t : $t \gg 1$

/ Page 23, paragraph 3: Curve II, $R_t = .4t^{-.6}$

/ Page 54, item 1 under Radiation flux: for circular chart read strip chart

9/13/56

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THE ABSORBANCE SHIFT OF IRRADIATED ORGANIC SOLVENTS. PART II*

The Effect of Massive Doses of Co^{60} Radiation

I. B. Berlman

Distilled xylene samples in glass containers were exposed to radiation from a Co^{60} source. The doses were measured by means of a Victoreen "r" meter, and the absorbance measurements were made relative to an unirradiated xylene sample by means of a Beckman DK-2 automatic recording spectrophotometer.

Before irradiation the oxygen content of the samples was brought up to equilibrium with that of the atmosphere by bubbling air through them, since it was noted from the very beginning of this study that the magnitude of the absorbance shift was oxygen dependent. In Figure 1 a plot is shown of absorbance versus wavelength for various doses of Co^{60} radiation on pure xylene. Since the irradiated samples were compared to the unirradiated sample, each curve is a plot of the absorbance change caused by the radiation. It is to be noted that the absorbance at all wavelengths increases almost linearly with dose over the range from 10^5 to 7×10^6 r. Within the experimental limits of accuracy no detectable difference was noted in the absorbance measurements by varying the dose rate over the range from 150 r/min to 1500 r/min.

Figure 2 shows the relative absorbance of benzene, toluene, and xylene when exposed to a dose of 10^6 r from a Co^{60} source. In each case the irradiated solvent was compared to the unirradiated one.

Scintillation solutions were then made of xylene and 2 g/liter of diphenyloxazole (PPO) and given massive doses of Co^{60} radiation. The luminescent efficiency of the scintillating solution to gamma rays, i.e., Co^{60} , was measured before and after irradiation by means of a Dumont 6292 photomultiplier tube, associated electronic equipment, and a 20-channel pulse height analyzer.

The criterion of luminescence efficiency used is based on the Compton distribution of electron recoils produced by gamma rays in organic scintillators of low "Z" material. The scintillations produced by these electrons were sorted as to energy distribution by the pulse height analyzer. In this spectrum, the pulse height along the leading edge of the Compton distribution, measured in mv, where the Compton distribution falls to one-half of its

*Part I appears in Report on Biological, Medical, and Biophysics Programs, Argonne National Laboratory. ANL-5518, p. 8 (1956).

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Figure 1. Absorbance change in pure xylene due to various doses of Co^{60} radiation.

Figure 2. Absorbance change in benzene, toluene, and xylene after exposure to 10^6 r from a Co^{60} source.

Figure 3. A plot of the light output, as expressed by the one-half maximum of the Compton luminescent response curve, versus dosage. The curve is composed of two sets of data normalized at the 10^6 r point.

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maximum value was chosen as the measure of luminescence. Thus, pulse height distributions were obtained for each irradiated sample and from an unirradiated sample, and the one-half maximum points were determined from the curves.

Plots of the one-half maximum points versus dose approximate a straight line on semilog paper as shown in Figure 3. The data for Figure 3 were obtained from two 15 cc solutions approximately a week after irradiation and are normalized at the 10^6 r point. It is felt, nevertheless, that these preliminary results need further confirmation since recent work indicates that the effect of radiation on the light output of scintillating solutions is dose-rate dependent.

In plastics, the absorbance induced by ionizing radiation decreased with time.⁽¹⁾ Similarly, in xylene the induced absorbance "bleaches" with time. Further measurements on this recovery effect are in progress and will be reported later. It is, however, interesting enough to note at this point that whereas the absorbance of xylene alone decreased with time after irradiation, the light output of a scintillating solution does not recover. This fact leads one to believe that the decrease in the light output of the scintillating solution is not only due to the absorption of the fluorescent light in transit through the liquid but also is due to the quenching and/or destruction of the scintillator.

Another related experiment was to expose a scintillating solution to a Co^{60} gamma-ray dose of 5×10^6 r. Samples of the solvent, xylene, and the scintillator, PPO, were irradiated separately (5×10^6 r) and a scintillating solution was prepared afterwards from the irradiated samples. The light outputs of these two scintillating solutions were compared, and within experimental limits, no reproducible difference was found. Thus if there is any protective effect of the solution by the scintillator because the scintillator channels some of the energy away in the form of visible radiation, the effect is masked by the large quenching and/or destruction of the scintillator material. When xylene alone was given a dose of 5×10^6 r and a scintillating solution made afterwards by adding to it unirradiated PPO, the resulting luminescent efficiency was 20% greater than that of a similarly irradiated solution of xylene plus PPO. This result seems to indicate that the radiation either destroyed a significant portion of the PPO or released from it significant amounts of quenching products, or actually reduced the luminescence by both mechanisms.

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RADON CONTAMINATION IN THE MEASUREMENT OF LOW LEVELS OF EXPIRED RADON. PART II

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

Part I of this study of contamination in the radon method of measuring natural amounts of body radium⁽¹⁾ showed that radium impurities in the materials of the apparatus contributed 0.02 μc Rn during collection periods of 30 min duration.⁽²⁾ This is a small and sufficiently constant amount for which corrections may be made readily. The present report describes experiments performed in order to measure contamination from sources which have their origin in, and are of a magnitude proportional to, the radon content of the atmosphere, namely: (1) diffusion of room-air radon through semipermeable materials during the analyses, and (2) the slow release into the breath of radon previously absorbed from the atmosphere by the bodies of the subjects.

In order to avoid the difficulties of measuring small amounts of radon, much of this work was done in air containing approximately 100 μc Rn/liter of air, i.e., about 500 times greater than normal. The source of excess radon was made by drawing air over crushed radon capsules into a partially evacuated air cylinder, and then adding air from cylinders of compressed air until the "radon tank" had been filled to a pressure of several hundred lbs/in². A period of localized heating served to mix the compressed air with the radon. This provided a convenient reservoir of radon which could be dispensed at a measured rate of flow into a room of controlled ventilation to increase the radon concentration of the room air to the desired level.

Diffusion of Radon through Semipermeable Materials. The components of the analysis apparatus⁽¹⁾ may be considered to consist of the following three groups: (1) impermeable materials for supplying radon-free air to the subjects and collecting radon from expired air (in addition to the metal and glass components, this group includes the polyethylene bag which is enclosed in a box filled with radon-free air); (2) flexible tubing for air supply and exhaust and a rubber helmet which encloses the head of the subject; and (3) the subject. For blank runs, the neck of the helmet was sealed, and the helmet alternately squeezed and released so as to force air from the helmet into the collection line and draw air from the supply system in a manner similar to the respiration of a subject.

Blank values are given in Table 1 for two ordinary rubber helmets (A and C) and a rubber helmet which was covered with an outer layer of polyethylene (Helmet B). For the ordinary helmets, 9 to 10 μc Rn were collected for runs of 15 min duration, and approximately twice this amount for 30 min runs made in air of the same high concentration of radon. On

the other hand, less than half these amounts was obtained for similar blank runs with the polyethylene-covered helmet. Since less than $0.1 \mu\mu\text{c}$ Rn was collected for 30-min blank runs with Helmets A and C in air of the normal concentration of $0.2 \mu\mu\text{c}$ Rn/liter, it is evident that radon diffuses through the rubber of the helmets and that polyethylene retards the diffusion.

TABLE 1

The effect of atmospheric radon on blank runs

Rn content of room air, $\mu\mu\text{c/liter}$	Helmet tested	Duration of Run, min	Volume collected, liters	Total Rn in blank, $\mu\mu\text{c}$	EVRA,* liters	EVRA*, liters/30 min
121	A	15	114	9.8	0.081	0.16
121	A	15	111	8.8	0.073	0.15
121	A	30	206	16.7	0.138	0.14
121	B	15	113	4.5	0.037	0.07
121	B	15	118	4.7	0.039	0.08
121	B	30	214	7.8	0.064	0.06
134	C	15	110	10.0	0.075	0.15
134	C	30	192	21.2	0.158	0.16

*The volume of room air containing the amount of radon obtained in the blank is defined as the volume of room air equivalent to the blank (EVRA).

In order to relate the amount of radon collected in the blank to the concentration of radon in the air, the results of Table 1 are also expressed as "equivalent volume of room air" (EVRA) which is the volume of room air containing the amount of radon collected. For example, $9.8 \mu\mu\text{c}$ Rn were collected in the first run listed in Table 1. This amount of radon is contained in 0.081 liter of air having a concentration of 121 $\mu\mu\text{c}$ Rn/liter. The average EVRA per 30 min obtained for Helmets A and C was 0.15 liter, while 0.07 liter/30 min was obtained for the polyethylene-covered helmet.

When blank runs at similar concentrations of radon in room air were attempted with a human subject, considerably higher values of the EVRA were obtained. As seen in Table 2A, the excess for 30-min runs amounted to 0.13 liter of room air containing 123 $\mu\mu\text{c}$ Rn/liter or approximately 16 $\mu\mu\text{c}$ Rn. Corrections for expiration of previously

absorbed radon amounted to less than $1 \mu\mu\text{c}$ Rn because the subject breathed the high level of radon for only a few seconds, and the analyses were made over a period of 1 to 5 hours after the start of breathing radon-free air. Thus, the source of a new leak was sought and ascribed to diffusion of radon through the skin of the subject into the blood stream and finally into the breath. Such diffusion was tested by placing the subject in a polyethylene bag sealed at the neck and making blank runs with the room air at low radon levels while the air inside the bag contained concentrations of radon considerably in excess of normal. Before radon was added to the bag and during the analyses, the subject breathed radon-free air at a pressure of 4 mm Hg in the respiratory helmet, so that direct entry (by a leak) into the helmet of air from the bag or the room was prevented. Table 2B shows that large amounts of radon were collected from the breath of the subject, whereas only a small amount of radon from the room air could have diffused into the helmet. The EVRA values for the two subjects of 0.13 liter and 0.09 liter in 30 min agree well with the excess previously noted in Table 2A when both the helmet and the body of a subject were exposed to high concentrations of radon.

TABLE 2

The effect of atmospheric radon on blank runs made with subjects

A. Helmet and torso of subject exposed to air of high radon content

Subject	Rn content of room air, $\mu\mu\text{c}/\text{liter}$	No. of Analyses	EVRA liters/30 min		EVRA for helmet, liters/30 min	Excess EVRA, liters/30 min
			Mean	Std dev		
AS	123	7	0.28	0.04	0.15	0.13
AS	124	7	0.27	0.02	0.15	0.12

B. Only torso of subject exposed to air of high radon content

Subject	Rn content of air, $\mu\mu\text{c}/\text{liter}$		No. of analyses	Radon in blank ($\mu\mu\text{c}/30 \text{ min}$)			Excess EVRA, liters/30 min
	Room	Bag		Total*	Helmet alone	Excess	
FI	3.3	325.	4	41.5 ± 1.9	0.5	41.	0.13
AS	0.34	95.	5	8.4 ± 1.2	0.05	8.4	0.09

*Mean value $\pm$ standard deviation of the mean.

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In measurements on the diffusion of gases through the skin, Behnke and Willmon⁽³⁾ found rates for helium corresponding to only one-half of the excess noted in Table 2 for radon. However, they also reported a strong temperature dependence for this effect, and no measurements of the temperature inside the bag were made in the present work.

Residual Atmospheric Radon in the Breath. In the radon method of measuring natural levels of body radium,⁽¹⁾ subjects are required to breathe radon-free air for periods in excess of 12 hours prior to collection of breath samples. As shown below, even this period of decontamination leaves a sizeable correction of the order of $0.1 \mu\mu\text{c Rn}$ to be applied to 30-min samples for contamination from residual atmospheric radon in the body. Thus, it was necessary to obtain data on the elimination of radon as a function of time after saturation uptake of atmospheric radon and to assess the range of variation for human individuals.

Harley, et al,⁽⁴⁾ made a detailed study of the rate of elimination of radon in the breath of a subject as a function of time after exposure for 8.5 hours to air containing $700 \mu\mu\text{c Rn/liter}$. They found that the data could be expressed as the sum of five exponential terms with half-lives of 0.4, 4.5, 41, 202, and 1100 min, respectively. A plot of their data in the region of 6 to 20 hours after the end of exposure is given in Figure 4, together with their data extrapolated to saturation uptake using the procedure of Tobias and coworkers.⁽⁵⁾

Figure 4. Concentration of radon in breath relative to the concentration of radon in air during uptake vs. time after the end of breathing a high concentration of radon.

○ Subject HL, 72-hr uptake □ Subject AS, 72-hr uptake
 — Data of Harley, et al., 8.5 hr uptake
 ---- Data of Harley, et al., extrapolated to saturation uptake

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In order to obtain a more direct measure of the elimination of radon after saturation uptake, two subjects were exposed to air containing approximately $100 \mu\mu\text{c Rn/liter}$ for a period of 72 hours at this laboratory. Immediately afterwards the room was cleared of excess radon, and the usual procedure for measurement of radon in breath was carried out. A plot of the radon content of breath samples relative to the original room level is given in Figure 4 as a function of time after the end of uptake. The data for subject HL agreed quite well with the extrapolated data of Harley, et al., whereas breath samples from subject AS at 16 hours contained only one-half the radon of those from HL. Furthermore, the radon content of breath samples from AS decreased at a faster rate than that in samples from HL.

Since it was not feasible to test many subjects for radon elimination after long periods of uptake, it was necessary to modify the experiments in order to evaluate the normal range of individual variation. Subjects were exposed to high levels of radon for a continuous period of 8 hours, allowed to leave for the day, and tested the following morning over a period of about 16 to 20 hours after the end of exposure. During the analysis period, the subjects breathed radon-free air in a helmet, and the room air was maintained at normal levels of radon.

The results are expressed on a per cent basis in Table 3 as the average concentration of radon in the breath of each subject relative to the concentration of radon in the air breathed during uptake. As indicated in the table, the subjects could be divided into four groups with a spread of about 0.05% between each group. All subjects were male with the exception of KG, who fell into the highest group. The most slowly deemanated radon from the body is believed to be that stored in the fat, and there was a fair correlation in Table 3 between apparent excess fat and the level of radon elimination. For example, EW and CB were in appearance the most overweight and the thinnest, respectively. It might be added that CB had a previous record as a diver in the U.S. Navy, a profession requiring rapid elimination of absorbed nitrogen.

For ^{HL}HF, the elimination rate 16 to 18 hours after the 72-hour period of uptake was 2.7 times as great as that for a similar period after the 8-hour uptake. The ratio for AS was 2.0. The difference in the two values is not unexpected for transfer of a gas from a flowing medium to a reservoir since it may be shown that the larger the reservoir, the longer half-saturation time; i.e., for HF the 8-hour uptake period fell farther short of saturation than for AS. Thus, for the highest group of Table 3, a factor of 3 might be required for extrapolation of the 8-hour data to saturation, whereas a factor of only 1.5 might apply to CB. This implies a spread of 0.5% to 0.035% relative to the original room level for residual radon released into breath samples collected 16 hours after saturation uptake.

TABLE 3

Residual atmospheric radon in the breath 16 to 20 hours
after an uptake period of 8 hours

Subject	No. of analyses	Per cent of uptake level*	
		Mean	Mean dev.
EW	3	0.17	0.02
JS	3	0.17	0.01
KG	3	0.17	0.02
LB	3	0.13	0.01
HL	4	0.11	0.01
HM	3	0.07	0.01
AS	5	0.07	0.01
CB	3	0.023	0.006

$$\text{*Per cent of uptake level} = \frac{\mu\mu\text{c Rn/liter in breath}}{\mu\mu\text{c Rn/liter in air during uptake}} \times 100.$$

For the usual concentration of 0.2 $\mu\mu\text{c Rn/liter}$ in room air, the correction for residual atmospheric radon might then be as high as 0.2 $\mu\mu\text{c Rn}$ (for a sample of 200 liters collected over a period of 30 min, 16 hours after the start of breathing radon-free air) or as low as 0.01 $\mu\mu\text{c}$. Further uncertainties in this correction would be caused by variations in the radon content of the atmosphere.

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ISOLATION OF Th^{228} FROM BONE WITH DIBUTYL PHOSPHATE

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

A recapitulation of the problem at hand is given in Table 4. Bone specimens from humans having body burdens of the order of $1 \mu\text{g Ra}^{226}$ are to be analyzed in order to determine whether appreciable amounts of Ra^{228} (MsTh_1) were also present when the radium dose was received 20 to 30 years ago. Assuming that it would be desirable to detect as little as 1% of Ra^{228} in the original radium dose, measurement of $0.1 \mu\mu\text{c Ra}^{228}/\text{g}$ ashed bone in the presence of $200 \mu\mu\text{g Ra}^{226}/\text{g}$ is required at present. In addition, the presence of approximately $36 \mu\mu\text{c}$ of Pb^{210} (RaD) and its daughters $/\text{g}$ of bone may be calculated from radioactive growth after correction for 70% loss of Em^{222} . The analytical procedure consists of isolating Th^{228} (RdTh) as a measure of the amount of its parent Ra^{228} . The last column on the right of Table 4 simply states that 0.005% of $200 \mu\mu\text{c Ra}^{226}/\text{g}$ equals $0.01 \mu\mu\text{c Ra}^{226}/\text{g}$ and is meant to indicate the degree of contamination permissible in the isolation of Th^{228} . Likewise, 0.03% of the RaDEF carried through the analytical procedure would result in $0.01 \mu\mu\text{c Po}^{210}/\text{g}$ or an amount equal to 10% of the $0.1 \mu\mu\text{c Th}^{228}$ expected from 1 g of ashed bone.

Extraction of Tracer Elements by DBP. The procedure previously described for the extraction of Th from solutions of bone⁽¹⁾ was further tested for recovery of Th and for removal of contaminating radionuclides. Unsatisfactory decontamination was observed for a number of test samples and was associated with the presence of a small amount of insoluble matter which remained after the ashed bone was dissolved in HNO_3 . As seen in Table 5, performance was greatly improved by filtering the solutions prior to extraction with DBP. In particular, the percentages of Ra^{226} , Po^{210} , and Bi^{210} carried through the extractions were below the limits indicated in Table 4. Recoveries of Th were poorer than desired, but seem adequate for the present problem. In the most recent group of tests, four unfiltered samples resulted in an average recovery of 92% with an average deviation of 2%, whereas four filtered samples provided an average recovery of 89% with an average deviation of 3%.

Counting of Alpha Rays from Th^{228} . Test samples with Th^{228} showed that the calculated growth of activity for the radioactive descendants of Th^{228} was not attained for the thin residues remaining after the DBP was ashed. Employment of an alpha-ray pulse chamber⁽²⁾ would be feasible but subject to the following limitations:

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- 1) Counting of the 8.78 Mev alpha rays of Po^{212} would be desirable in order to eliminate interference from alpha rays of other energies, but the variations in the retention of Em^{220} introduce considerable uncertainty in quantitative measurements;

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TABLE 4

Relative amounts of radioactive nuclides in ashed bone

Assumptions:

1. Body content of $1 \mu\text{g Ra}^{226}$ from dose received 27 years ago.
2. Bone weight when ashed = 500 g (70 kg man).
3. Ra^{228} : Ra^{226} ratio = 0.01 at time of dose.

Nuclide	$\mu\mu\text{c/g}$ at t_1^*	Fractional change		$\mu\mu\text{c/g}$ at $t_1 + 27 \text{ y}$	% of nuclide = $0.01 \mu\mu\text{c/g}$
		Decay or growth	Retention in bone		
Ra^{226}	200	1.0	~ 1.0	200	0.005
RaDEF^{**}	0	0.6	0.3	36	0.03
Ra^{228}	2	0.06	~ 1.0	0.1	-

* t_1 may be some time after receiving the dose, i.e., when Ra "becomes fixed."

**RaDEF denotes Pb^{210} in radioactive equilibrium with Bi^{210} and Po^{210} .

TABLE 5

Extraction of tracer elements from HNO_3 solutions of ashed bone by DBP

Tracer	Untreated solutions		Filtered solutions	
	No. of runs	% recovered	No. of runs	% recovered
Th*	12	79 - 98	8	76 - 94
Ra	5	0.03 - 0.001	2	0.004 - 0.0002
Po	8	0.2 - 0.01	2	0.006 - 0.004
Bi	8	0.01 - 0.001	2	<0.001
Pb	8	<0.001	2	<0.001

* Th^{230} (Io) tracer was used in order to eliminate complications of radioactive growth or decay.

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- 2) Alpha rays from Th^{228} or Ra^{224} possess energies similar to those of Po^{210} and Em^{222} , so that decontamination is essential; and
- 3) Some evidence of loss of Ra^{224} by recoil from DBP residues implies a further uncertainty in the measurement

Precipitation of Ra^{224} (ThX) with BaSO_4 . A satisfactory technique for measuring Th^{228} after extraction into DBP consisted of milking 3.64d Ra^{224} from the DBP- Th^{228} solution with 2 M HNO_3 . After addition of 2 mg Ba^{++} carrier, the acid solution was evaporated to dryness and the salts dissolved in 0.01 M HNO_3 . A lead sulfide precipitation was then performed to aid in decontamination, and the solution was transferred to a platinum planchet. Finally, H_2SO_4 was added in order to precipitate a smooth layer of BaSO_4 before evaporation to dryness under a heat lamp. Alpha rays from the sample could then be counted in an ordinary alpha counter because Em is quantitatively retained in BaSO_4 (3) and the characteristic growth and decay of Ra^{224} and its radioactive descendants provided a positive identification of the nuclide.

The dibutyl phosphate- BaSO_4 procedure is now being applied to a number of bone samples provided by Dr. R. J. Hasterlik of the Argonne Cancer Research Hospital.

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RADON SCINTILLATION COUNTER

H. F. Lucas, Jr.

The difficulties due to the effect of traces of water and ethylene dichloride on the counting efficiency of the radon scintillation counter described previously⁽¹⁾ have been resolved. It was found that the impurities remaining in newly constructed counters (probably ethylene dichloride) after pumping with a Cenco Hi-Vac vacuum pump were sufficient to increase the counting efficiency as much as 30%, while those remaining in one counter after eight months of use caused an increase of 6%. These impurities could be removed by evacuating the counter on a high vacuum system for 3 hours at less than 0.1 micron.

The addition of water or ethylene dichloride to the counter filling was observed to modify the distribution of the radon daughters when their decay was analyzed after removal of radon. To control the distribution of the radon daughters, a metal counter was modified by replacing the quartz window with one having a transparent conductive tin coating⁽²⁾ connected electrically to the metal shell.* An electrode, insulated from the shell, inserted at the top of the counter permitted, with appropriate potentials, the collection of the radon daughters on either the walls or the electrode. The counter was then filled with an aliquot of radon from a NBS radium solution, and the influence of various collecting potentials and of added water on the counting efficiency was determined. It is shown in Figure 5 (neglecting the slight radon loss during addition of the water) that the counting efficiency during collection of the daughters on the walls was independent of the collecting potential or the presence of the added water vapor. Conversely, collection of the daughters on the electrode was dependent on the collecting potential and the presence of the added water vapor. However, the geometrical counting efficiency for the radon daughters on the electrode is approximately the same as for radon daughters on the quartz window of an unmodified counter. Therefore, a potential induced on the window, by its proximity to the photomultiplier, influenced the distribution of the radon daughters within the counter, which was in turn made variable by changing water and ethylene dichloride content.

By observing the counting rates of several radon counters in a modified counting system and in a standard counting system, it was established: (1) that the negative voltage on the photocathode induced the potential on the counter window, and (2) that this induced potential influenced the counting efficiency for as long as 72 hours. Since cleaning the window with tissue would also induce a charge on the quartz, it was necessary to modify each radon counter.

*Less than 2% decrease in counting rate at a fixed bias setting was observed for a coating having a resistance of 2.5×10^3 ohms.

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Figure 5. Influence of water and potential on the collection of the radon daughters.

Several radon counters, with conductive windows, were tested by comparing the counting rates per standard sample with and without added ethylene dichloride vapor. The counting rates observed (Table 6) were corrected for radon decay, and the deviations observed include errors in sample collection, transfer, and counting. It is seen that the counting efficiency is not influenced (with the exception of counter #21) by the added ethylene dichloride vapor and that the reproducibility of counting rate for any one counter is $\pm 2\%$. Since the small differences in counting rate between counters has been qualitatively correlated with the amount of ZnS(Ag) scintillator area covered with adhesive, all the counters do not have the same counting efficiency. Improved construction techniques now being tested should eliminate the variations in ZnS(Ag) scintillator area as well as the change in counting efficiency with added ethylene dichloride observed in counter #21.

The counting efficiency of the radon counting system, summarized in Table 7, was determined by analyzing aliquots of radon from three radium solutions.* Since sources #1 and #2 are aliquots of the same NBS radium solution, while source #3 is an aliquot of a radium solution which was calibrated by alpha and gamma counting techniques, it is obvious that source #2 has lost some of its radium.

*converted from $\mu\mu\text{g}$ to $\mu\mu\text{c}$ by the relationship $1.000 \mu\mu\text{c} = 0.9774 \pm 0.0080 \mu\mu\text{g}$.

TABLE 6

Insensitivity of modified radon counter to
added ethylene dichloride vapor

Counter number	Ethylene dichloride added			
	None		3.2 cm Hg*	
	No. of samples	cpm $\pm$ E**	No. of samples	cpm $\pm$ E**
14	2	971 $\pm$ 7	4	981 $\pm$ 3
17	3	992 $\pm$ 7	2	994 $\pm$ 1
19	1	1000 $\pm$ 9	2	981 $\pm$ 18
21	4	939 $\pm$ 17	2	980 $\pm$ 5
22	5	1005 $\pm$ 14	3	992 $\pm$ 6
12			2	972 $\pm$ 4
18			2	1003 $\pm$ 3

*Partial pressure

**Standard deviation from the mean

(Vapor pressure of ethylene dichloride at 20° C = 8.5 cm Hg)

TABLE 7

Calibration of radon counting procedure as
determined by aliquoting of radon from
various radium²²⁶ solutions

Source No.	Number of samples	$\frac{\text{cpm}}{\mu\mu\text{c}}$
1	5	5.41 $\pm$ 0.06*
2	2	4.79 $\pm$ 0.06
3	4	5.22 $\pm$ 0.16

*Standard deviation from the mean

TABLE 8

Radon and counting losses

Location	% loss
Sample collection	0.10 $\pm$ 0.02
Sample transfer	0.89 $\pm$ 0.20
Pulse height discrimination	2.5 $\pm$ 0.8
Insensitive volume of counter	0.19 $\pm$ 0.01
Insensitive scintillation area	2.0 $\pm$ 1.0
Total	6.0 $\pm$ 2.0

The comparison of the observed and theoretical counting efficiencies requires correction of the observed efficiency for all radon and counting losses (summarized in Table 8), while calculation of the theoretical efficiency requires knowledge of the distribution of radon and its daughters. When it was determined that the radon was not adsorbed on the ZnS(Ag) scintillator (less than 1%), the theoretical counting efficiency for radon in the gas with various distribution of the daughters (Table 9) was calculated from constants obtained by the computer group. Previous investigation has shown (Figure 5) that the radon daughters do not remain in the gas or concentrate on the counter window, but are attracted to the walls. The theoretical counting efficiency for uniform distribution of radon in the gas and daughters on the wall (Case I, Table 9) is 5.68 ± 0.03 cpm/ $\mu\mu\text{c}$, which, when compared to the corrected observed 5.77 ± 0.11 cpm/ $\mu\mu\text{c}$, is within statistical deviation. The possibility of nonuniform distribution of the daughters on the walls has not yet been evaluated, and an increase of the theoretical counting efficiency of 5% is possible (Case IV).

TABLE 9

Theoretical counting efficiency for various distributions
of radon daughters based on uniform distribution
over the volume or areas involved

Case	Location of		Efficiency
	Radon	Daughters	cpm/ $\mu\mu\text{c} \pm E^*$
I	Gas	Walls	5.68 ± 0.03
II	Gas	Gas	5.78 ± 0.03
III	Gas	Window	4.16 ± 0.02
IV	Gas	ZnS(Ag)	5.97 ± 0.03

*0.9 confidence level

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// CALCULATIONS PERTAINING TO RADIUM PRODUCED IN VIVO FROM A THORIUM PARENT

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

In a report on Th²²⁷ Accident⁽¹⁾ experimental values of the Th²²⁷ and Ra²²³ body content and daily excretion of these elements were presented. In order to determine the validity of these results an attempt has been made to calculate the Ra/Th activity ratio as a function of time and the radium excretion expressed as the per cent of radium body burden at any given time.

Two methods of calculation were employed: 1) use of the power function representation of radium retention; and 2) radium retention expressed as a sum of four exponential functions the sum of which equal the power function within the proper time interval. The Ra/Th activity ratio is then obtained at any given time by dividing by the amount of thorium present. The assumption is made that the loss of thorium in vivo follows the physical decay of the isotope very closely, excretion being negligible. This seems justified in light of excretion analysis in this case, as well as from past observations on the high retention of injected thorium compounds.

Both the Ra/Th activity ratio and the radium excretion are measurable in this experiment, and both are intimately related to the metabolism of radium. Therefore a consistent theory must account for both simultaneously.

The power function representation of radium retention as a function of time is of the form

$$R_t = R_0 A t^b$$

where A and b are assumed to be constants, R₀ is the injected dose in suitable units and t is the time in days elapsed since injection.

Several modifications must be introduced when radium is produced in vivo from a thorium parent. Retention now becomes the integral of the individual daily radium increments due to thorium decay multiplied by the power function for the time elapsed between time of radium production and time of measurement. This integral must also include factors to account for the radioactive decay of both thorium and radium.

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The modified retention function at time t is of the form

$$R_t = \int_0^{t-1} R(T) A(t-\tau)^b e^{-\lambda_R(t-\tau)} d\tau$$

where $R(T)$, the radium produced per unit time, is a function of the thorium present. This is given by

$$R(T) = \lambda_T T_0 e^{-\lambda_T T}$$

where λ_R and λ_T are the decay constants of radium and thorium respectively, λ_T the effective decay of thorium in vivo and T_0 the amount of thorium introduced in the body. Since τ is the variable of integration, the expression for R_t may be written as

$$R_t = \lambda_T T_0 A e^{-\lambda_R t} \int_0^{t-1} e^{(\lambda_R - \lambda_T)\tau} (t-\tau)^b d\tau.$$

Making the substitution,

$$w = t - \tau, \tau = t - w, d\tau = -dw$$

the limits become $w = t$ when $\tau = 0$

$$w = 1 \text{ when } \tau = t - 1.$$

So that

$$R_t = \lambda_T T_0 A e^{-\lambda_R t} \int_1^t e^{(\lambda_R - \lambda_T)(t-w)} w^b dw$$

Since w is now the variable of integration, we obtain

$$R_t = \lambda_T T_0 A e^{-\lambda_T t} \int_1^t e^{-(\lambda_R - \lambda_T)w} w^b dw.$$

By making a further substitution,

$$v = (\lambda_R - \lambda_T)w, \text{ where } w = \frac{v}{(\lambda_R - \lambda_T)}$$

$$\text{where } dw = \frac{dv}{(\lambda_R - \lambda_T)}$$

having the limits

$$\begin{aligned} v &= (\lambda_R - \lambda_T)t & \text{when } w &= t \\ v &= (\lambda_R - \lambda_T) & \text{when } w &= 1 \end{aligned}$$

(21)

The retention equation can be put into the form

$$R_t = \frac{\lambda_T T_0 A e^{-\lambda_T^1 t}}{(\lambda_R - \lambda_T^1)^{b+1}} \int_{(\lambda_R - \lambda_T^1)}^{(\lambda_R - \lambda_T^1)t} e^{-v v b} dv$$

Since we are actually concerned with the Ra/Th activity ratio as a function of time, we are interested in the expression

$$\frac{\lambda_R R_t}{\lambda_T T_t} \text{ where } T_t = T_0 e^{-\lambda_T^1 t}$$

The expression to be calculated becomes

$$\frac{\lambda_R R_t}{\lambda_T T_t} = \frac{\lambda_R A}{(\lambda_R - \lambda_T^1)^{b+1}} \int_{(\lambda_R - \lambda_T^1)}^{(\lambda_R - \lambda_T^1)t} e^{-v v b} dv \quad (A)$$

The integral now has the form of the Incomplete Γ - Function, and therefore tables of this function may be used for its evaluation. The tables available were those by Karl Pearson, (2) in which the Incomplete Γ - Function is given as the ratio of the Complete Γ - Function for the same value of b . The tables are in terms of:

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

which in normalized form is

$$I(x, b) = \frac{\Gamma_x(b+1)}{\Gamma(b+1)} = \frac{\int_0^x e^{-x} x^b dx}{\Gamma(b+1)} \quad (B)$$

A transformation $u = \frac{x}{(b+1)^{1/2}}$ which gives

$$I(u, b) = \frac{\int_0^{u(b+1)^{1/2}} e^{-u(b+1)^{1/2}} [u(b+1)^{1/2}]^b (b+1)^{1/2} du}{\Gamma(b+1)}$$

and second transformation $v = u(b+1)^{1/2}$ brings B into

$$I(u, b) = \frac{\int_0^v e^{-v} v^b dv}{\Gamma(b+1)}$$

The integral of interest to us is

$$\int_{(\lambda_R - \lambda_T)t}^{(\lambda_R - \lambda_T)t} e^{-v} v^b dv \quad \text{which is equal to}$$

$$\int_0^{(\lambda_R - \lambda_T)t} e^{-v} v^b dv - \int_0^{(\lambda_R - \lambda_T)t} e^{-v} v^b dv$$

Using the retention equation $R_t = .54t^{-.5}$ as found by Norris, et al.,⁽³⁾ the quantity $v = (1 - .5)^{1/2} u = .707 u$ results from the tables; whereas using the derived equation $v = (\lambda_R - \lambda_T)t$ where for Ra^{223} , $\lambda_R = .0619 \text{ day}^{-1}$ and for Th^{227} $\lambda_T = .0408 \text{ day}^{-1}$, $v = .0211 t$.

For a given value of b , the quantity $I(u, b)$ is tabulated for different values of u so that we may obtain the value of $I(u, b)$ as a function of time by the relationship:

$$v = .707 u = .0211 t, \quad t = .707 u / .0211 = 33.5 u.$$

Furthermore the value of $\Gamma(1 - 1/2) = \sqrt{\pi}$; hence the values in the table must be multiplied by this quantity in order to obtain the value of the integral.

For example, the activity ratio at 10 days is calculated as follows

$$\begin{aligned} \frac{\lambda_R R_{(10)}}{\lambda_T T_{(10)}} &= .408 \int_0^{.211} e^{-v} v^{-1/2} dv - \int_0^{.0211} e^{-v} v^{-1/2} dv \\ &= 4.08 (.4852 - .1200) = .149 \end{aligned}$$

The Ra/Th activity ratio was also computed by breaking the power function $R_t = .54t^{-.5}$ down into a sum of four exponentials. In this method retention is unity at time zero and .54 at one day. The sum of coefficients must be equal to one and the decay constants for this case are as follows

$$R_t = .082e^{-.004t} + .107e^{-.0465t} + .175e^{-.193t} + .636e^{-1.0t}$$

where t is the post injection time in days.

This expression leads to the integral for radium retention

$$R_t = \lambda_T T_0 \int_0^t e^{-\lambda_T \tau} \left[.082 e^{-.004(t-\tau)} + .107 e^{-.0465(t-\tau)} + .175 e^{-.193(t-\tau)} + .636 e^{-1.0(t-\tau)} \right] e^{-\lambda_R(t-\tau)} d\tau$$

Integrating and performing the same operations as shown above, the activity ratio $\frac{\lambda_R R_t}{\lambda_T T_t}$ is obtained in the form

$$\frac{\lambda_R R_t}{\lambda_T T_t} = .202 [1 - e^{-.2051t}] + 0.98 [1 - e^{-.0676t}] + .051 [1 - e^{-.2141t}] + .039 [1 - e^{-1.0211t}]$$

For the sake of comparison, the value of the ratio at 10 days computed by this method is .176 which is about 15% larger than that obtained by using the Incomplete Γ -Function. The Incomplete Γ -Function value depends very sensitively upon the lower limit in the integral. If instead of $t = 1$, we had chosen $t = .8$, the computed value would have been .175 which agrees very well with the sum of exponentials method.

Figure 6 shows the Ra/Th activity ratio obtained experimentally at various times. The computed values, using the sums of exponentials, are also indicated for three sets of values of the constants A and b in the power function. These are Curve I, $R_t = .54t^{-.5}$, Curve II, $R_t = .4t^{-.6}$ and Curve III, $R_t = .3t^{-.7}$. The assumption made in the preceding calculations was that radium-free thorium was introduced into the body. Since the thorium actually introduced through the wound had a two-day growth of radium associated with it, correction has been made for this amount in the curves shown in Figure 6. The experimental points tend to lie between Curves II and III indicating radium retention follows a power function with somewhat different values for A and b than those found for injected radium.

The time derivative of the integral of a sum of exponentials will give the excretion rate; dividing it by the body burden at that time will give excretion expressed as per cent of body burden. Rate of radium excretion does not vary appreciably within the range of values of A and b considered here. The computed and experimental values for radium excretion are shown in Table 10. The agreement seems satisfactory except on the 36th day and this only by a factor 2.

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Figure 6. Ra^{223}/Th^{227} activity ratio as a function of time after the accident in days.

In the case of physical equilibrium, the Ra/Th ratio is 2.52 for Ra^{223}/Th^{227} , 1.005 for Ra^{224}/Th^{228} , 1.000 for Ra^{228}/Th^{232} , and 1.000 for Ra^{225}/Th^{229} .

Figure 7. Radium/thorium activity ratio in vivo.

With a given form of the retention equation, $R_t = .54t^{-.5}$, for example, the Ra/Th activity ratio is strongly influenced by the half life of the radium involved, (see Equation A). The longer lived the radium, the greater the probability of its being excreted. This is illustrated in Figure 7, where values of this ratio as a function of time have been calculated for $\text{Th}^{228}\text{-Ra}^{224}$, $\text{Th}^{227}\text{-Ra}^{223}$, $\text{Th}^{232}\text{-Ra}^{228}$, and $\text{Th}^{229}\text{-Ra}^{225}$.

TABLE 10

Total Ra^{223} excretion as per cent of body burden*

Day	Experimental value, %	Calculated value, %	
		$R_t = .54t^{-.5}$	$R_t = .3t^{-.7}$
36	7.6	3.1	3.6
48	4.0	3.4	3.8
70	3.9	3.7	3.9

*Excretion computed on the basis of 1250 cc of urine and 25 g dry feces.

It is to be realized that attainment of physical equilibrium between radium and thorium in some instances must be measured in units of geological time rather than in terms of human life-span. However the fact remains that in all instances the equilibrium existing in vivo is less than that anticipated on the basis of radioactive growth. From the standpoint of biological damage to be expected from a given burden of thorium this has some intriguing aspects. In the case of Th^{227} , the approximate energy release, excluding gammas, during the decay of a single Th^{227} nucleus to Pb^{207} is some 33 Mev. However, with only one chance in five of its remaining in the body once it has become Ra^{223} , the total energy release becomes 11 Mev; namely, the dose received is only 1/3 that which would apply if thorium were in physical equilibrium in vivo.

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MEASUREMENTS OF GAMMA RAYS IN VIVO

C. E. Miller

An 8" x 4" NaI(Tl) Crystal Placed in Operation. The 8" diameter x 4" thick NaI(Tl) blank which was received from Harshaw Chemical Company in January has been ground to size, roughened, and vacuum canned in MgO. A variation of the vacuum canning technique described by Swank and Moenich⁽¹⁾ was used with the substitution; however, in order to achieve a lower background, of #304 stainless steel components and quartz for aluminum and pyrex. The stainless steel can, which has a 10-mil thick diaphragm top, was fabricated with a tapered pumping hole so the crystal could be completely assembled and freed partially of air as the last step.

This crystal, although 8" in diameter, was mounted on a single 5" photomultiplier which has an effective photo cathode diameter of only $4\frac{1}{4}$ ". A teflon reflector cemented to the quartz window with DC-200 silicon grease covers that area of the crystal window not covered by the photo cathode surface. A collimated Cs¹³⁷ source placed parallel to the axis of the tube and crystal but $3\frac{1}{2}$ " from the center of the crystal yields the same pulse height spectrum as that of a collimated source placed on the axis of the crystal. This test demonstrated that the same light collection efficiency existed for events at the edges as for those at the center. It was felt that this arrangement, while yielding only about 9% resolution at the K⁴⁰ photopeak, was preferable to an arrangement utilizing several photomultipliers in that the problem of calibrating, as well as balancing the drifts of several tubes, was circumvented.

The gamma-ray background of the crystal within the 8" Fe room, as well as its response to an enriched K⁴⁰ source, is shown in Figure 8. No attempt has been made to reduce the background further by adding shielding since the shield would be extremely bulky, heavy, and expensive, due to the large overall size of the crystal mount.

The advantage of this crystal over the 4" x $1\frac{1}{2}$ " crystal is emphatically demonstrated by Figure 9, where the normal gamma-ray spectra of subject O.J.S. as obtained with both crystals are shown. The photoelectric peak is enhanced with respect to the Compton region by a factor greater than 2 because of the increased thickness of the crystal. This crystal thus has a higher sensitivity than the smaller one since a larger percentage of the detected gammas fall in the photopeak and thus stand out as a peak and not just a slight change of slope. While the signal to background ratio at the K photopeak is the same for the two crystals, the 8" x 4" crystal yields a net counting rate 8 times that of the 4" x $1\frac{1}{2}$ " crystal. As a consequence, the counting time can be reduced to 1/8 of the former with no loss in precision.

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Reproducibility of Data. The data previously reported⁽²⁾ has been taken with the subjects seated in a standard deck-type canvas chair and with the crystal detector located 8 cm from the 12th thoracic vertebra. This arrangement has proved to be unsatisfactory for several reasons. One difficulty was that as the subject fatigued, he had a tendency to shift position and thus change his effective geometrical distance from the crystal. As a consequence his absolute value varied from run to run. Another and more serious difficulty was the fact that the counting rate/ μ c of body burden depended upon the subject's height and weight distribution. For example, the counting rate/ μ c of body burden would be 40% higher for a 5'2", 120-lb subject than for a 5'10", 190-lb subject. This change in c/m/ μ c is due partly to the greater absorption and scattering in the larger person. The main contributing factor was the greater distance between the detector and the feet and head of the taller person.

In an attempt to eliminate those two difficulties, a new tilted chair was designed and built and the detector positioned as shown by Figure 10. The subject lies in the chair and does not have a tendency to move since he must do work in order to raise any part of his body. The detector is suspended over the subject in either of the positions labeled #1 and #2. As a result, all areas of the body are essentially at the same distance from the counter and the effect of height is minimized.

The distance from the center of the crystal to both the back and seat of the chair is kept fixed at a particular value. The front or nearest surface of a thick, heavy-set person is thus closer to the crystal than that of a thin subject. Apparently, the increased counting rate due to the reduction in distance between the subject and detector counterbalances the greater absorption in the larger person, and thus a fortuitous arrangement has been realized which is fairly independent of height and weight.

To evaluate the above arrangement fully, data were taken on a group of 12 subjects with the detector located at the back of the subjects as well as at positions #1 and #2. In addition, three of these subjects were checked on the one-meter arc position. These latter three subjects plus one other of the above twelve were given a calibrated K^{42} tracer and their K burdens calculated for each position. All four arrangements led to the same value of body potassium demonstrating that the material, K^{42} , was in equilibrium with the body's own K and that the value obtained was correct. As an example, the values of body K calculated for subject O.J.S. are:

1-meter arc	132 g.
Crystal at back	130
Crystal at #1 position	132.2
Crystal at #2 position	133.6

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Subsequently, the gamma-ray spectra of seven subjects, one being one of the four mentioned above, were obtained in position #1 after which time they were given a calibrated K^{42} tracer. The K^{42} data for the ten subjects are tabulated in Table 11. The net counts per minute from K^{42} in vivo have been divided by the amount of K^{42} in μc present at the time of the measurement which was some 14 hours or more after ingestion. The data listed in the table are thus in terms of counts per minute per microcurie of beta activity, so that the values shown for any position of any subject can be compared directly with those values for any other subject in any position.

The K^{42} spectrum has been broken into two energy bands. The first covers those gamma rays from 0.88 Mev to 1.39 Mev and are hereafter referred to as the Compton region. The gamma rays falling below 0.88 Mev were not considered because the Cs^{137} body burden precludes any calculations of K^{40} levels within scintillations below this energy. The second band covers those gammas falling within the photoelectric peak.

The advantages of positions #1 and #2 are observed upon examining Table 11. The $c/m/\mu c$ in the photopeak for position #1 is essentially independent of the height and weight of the subject. The 36% spread in the counting rate from the smallest to the largest subject when the crystal was placed at the small of the back has been reduced to a 5% spread for position #1. It is thus possible to estimate the subject's body K to within 3% without administering any radioactive tracer if the chair position has been previously calibrated with a range of subjects.

Cesium in Humans. It was reported in the last semiannual report⁽²⁾ that most human beings contain a slight body burden of Cs^{137} , a fission product. This burden has increased from 50 to 100% in the four subjects on whom consecutive runs have been made since July 1955. Figure 11 shows the gamma-ray spectrum of the author on October 10, 1955, and on February 13, 1956. The increase in this subject exactly paralleled the increase in subject H.A.M.

The absolute Cs body burden varies from one subject to the next, and no correlation exists between the potassium level and the amount of Cs present. The amount of Cs present seems to depend upon the diet of the individual as can be seen from the gamma-ray spectrum of four subjects of the same age group (50 yrs) shown in Figure 12. All four subjects had the same amount of K and thus only one spectral curve is shown above 750 kev. Three curves are shown at energies below the Cs photopeak. The lower thin line is the shape expected at the lower energies if the subject is free of Cs. The dashed curve is that of an individual having a body burden of about 3 $m\mu c$ of Cs and demonstrates the Cs contribution. The top curve is that of a subject possessing 9 $m\mu c$ of Cs. The spectra of the other two subjects fell between these two curves and have been omitted

TABLE 11

Counting efficiency with different geometrical arrangements

Subject	Subject on 1-m arc				Subject seated in tilted chair											
	Crystal at center of 1-m circle				Crystal at small of back				Crystal at position #1				Crystal at position #2			
	Compton region		Photopeak		Compton region		Photopeak		Compton region		Photopeak		Compton region		Photopeak	
	cpm	Dev.	cpm	Dev.	cpm	Dev.	cpm	Dev.	cpm	Dev.	cpm	Dev.	cpm	Dev.	cpm	Dev.
H.A.M. #1 5'10½" - 135# 1.915 #/in									932	14.6	1718.6	37.6				
K.G. #2 5'2" - 120# 1.935 #/in	204	8.8	391	23	1201	173	2040	315.7	913	33.6	1696	15.2	594	8.2	1089	9
E.A. #3 5'8" - 143# 2.1 #/in									943	3.6	1690	9.0				
L.S. #4 5'6" - 142# 2.15 #/in									898.2	51.6	1633	48				
J.S. #5 5'11" - 154# 2.17 #/in									937	9.6	1707	26.0				
O.J.S. #6 5'7" - 152# 2.27 #/in	200	4.8	379	11	1029	1	1737	12.5	949.4	2.8	1690	9.0	585	17.2	1101	21
P.H. #7 5'11½" - 179# 2.503 #/in									960	13.4	1689	8.0				
C.E.M. #8 5'9½" - 175# 2.52 #/in									988	41.4	1646	35.0				
C.T.E. #9 5'10" - 190# 2.724 #/in	181.6	13.6	334.5	33.5	968	60	1622	102.3	917.3 (927.5)*	29.3	1638 (1677)*	43.0 (4.0)*	575	27.5	1042	38
M.H.S. #10 5'5" - 190# 2.923 #/in					915	113	1498	226.3	1028	81.4	1703	22	655	52.8	1088	8
Average	195.2		368		1028				946.6		1681		602.2		1080	
Average deviation		9.07		22.5		86.8		164.2		28.13		25.28		26.3		19
Average error		4.65%		6.1%		8.44%		9.5%		3%		1.5%		4.37%		1.76%
Maximum error		7%		9.1%		16.8%		18.3%		8.6%		2.57%		8.8%		3.5%

*Second run; subject moved excessively during first one.

Figure 8

Figure 9

Figure 10

Figure 11

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to avoid confusion in the graph. These curves serve to demonstrate the threefold difference in Cs burdens from one subject to another due to their difference in diet. The main difference in the diet of the four subjects is the quantity of milk each consumes, i.e., the higher the milk consumption, the greater the Cs burden.

The two subjects mentioned in the last semiannual who had ingested a small quantity of Cs¹³⁴ have been excreting the isotope with an approximate biological half-life of 100 days for the past 60 days. This value is considerably greater than the 17-day half-life reported in Handbook 52⁽³⁾ and is in agreement with the biological half-life found for the two subjects recently given Cs¹³⁴ by Dr. E. C. Anderson of Los Alamos. Very little additional work is contemplated along these lines except to follow the two Cs¹³⁴ subjects and to follow the Cs¹³⁷ level of a few subjects.

Cesium in the Urine. In order to prove definitely that the foreign gamma-ray photopeak in vivo was due to Cs¹³⁷, attempts were made to identify Cs in the urine. Urine samples were collected from various subjects over a 3-day period and the gamma-ray spectrum obtained from the whole sample. The gamma-ray spectrum was also obtained from a pure enriched K⁴⁰ source. These two spectra plus the difference spectrum are shown in Figure 13. Although the difference spectrum fits the expected gamma-ray spectrum of Cs¹³⁷, it was felt that all doubt must be removed concerning the possibility that it might be due to some other radioactive isotope emitting a gamma ray of the same energy.

Consequently, Dr. Jacob Sedlet, Bio-assay Group, Industrial Hygiene and Safety Division, performed a chemical separation designed to extract Cs or Rb present. The cesium carrier used was first checked for radioactivity, and the procedure was performed on distilled water blanks to prove that Cs¹³⁷ was not being introduced by any other route. At the conclusion of the chemical separation, the gamma-ray spectra of the various residues were determined, and the following spectrum was obtained from the separated Cs fraction. (See Figure 14.) Rubidium isotopes can be ruled out on the basis of their physical half-life as well as by the energy of the gamma rays being emitted. The remaining salts, while containing K, do not contain the foreign photopeak. These chemical procedures provide definite proof that the contaminant is an internal one and that it is Cs¹³⁷.

Foods. The gamma-ray spectra of numerous foods have been obtained. Cesium has been found in all samples of domestic milk, most samples of cheese and some samples of meat. A typical milk spectrum is shown by Figure 15. No fission product activity has been found in any vegetable. A #1 sample of tobacco yielded the pure K spectrum of Figure 15, but contained no cesium or other fission product.

Figure 12

Figure 13

Figure 14

Figure 15

Incident Victims. Several subjects who were suspected of having accumulated a body burden of some isotope have been brought to our laboratory. In one instance, a 2-year-old boy had eaten the letters off a radium dial clock. In a second case, a girl had cut her finger while wearing externally contaminated rubber gloves. In still another instance, several subjects had entered a room in which the air contained a high level of fission products. In all cases mentioned, a ten-minute run was sufficient to state that the subject in question did not have a total body burden in excess of 10^{-9} c of any mixture of isotopes other than the normal quantity of K and Cs¹³⁷.

Acknowledgment

The author is pleased to acknowledge his indebtedness to Mr. L. D. Marinelli for his valuable collaboration and guidance during the course of this work. Thanks also are due to Mr. O. J. Steingraber for his capable technical assistance.

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PROGRESS REPORT: ANALYSIS OF RADIUM DAMAGE TO BONE BY
MEANS OF MICRORADIOGRAPHY AND AUTORADIOGRAPHY

R. E. Rowland

Sections of human femur available from those radium cases in which total body activity measurements have been made are being microradiographed and autoradiographed. The sections are cut from plastic-embedded bone in thicknesses of 100 microns, separated by 150 microns. Each section, after it has been microradiographed, is covered with stripping film for autoradiography. From these sections, the following information is being sought:

1. The fraction of the haversian systems which contain radium.
2. The fraction of the haversian systems which show abnormalities.
3. The average length of radium activity in haversian systems.
4. The average activity per system and the average activity per unit area of those systems containing radium.
5. The total dose to each system from its radium burden.
6. Comparison of (5) with average dose to skeleton.

MICRORADIOGRAPHY: THE DETERMINATION OF MASS PER UNIT AREA OF BONE SLICES

Robert E. Rowland

The use of microradiographs to determine the density of specific elements in suitable sections has been recognized for some time.⁽¹⁾

In studies of radium damage to bone, as well as in studies of accretion and resorption rates of calcium in haversian systems, we can now obtain the density of the apatite crystal structure per cubic micron of bone. In other words, we may obtain a numerical value for the degree of calcification, or mineralization of the bone.

The calculation of mass per unit area, from which we can derive density if the thickness of the section is accurately known, is based upon the similarity in the shape of the absorption curves for Ca, P, and Al, as is shown in Figure 16.

Figure 16. A semilog plot of the mass absorption coefficients (μ) for aluminum, phosphorus and calcium as a function of energy. Superimposed are the energy distributions of the photons emitted from the x-ray tube when operating at 10 kev, with a total filtration of 1 mm of beryllium. (The characteristic x-rays from the tungsten anode have been ignored.)

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Aluminum is employed as a step wedge, and from the degree of darkening of the photographic plate under the various thicknesses of aluminum, we obtain a calibration curve which can then be applied to the bone sections.

On Figure 16 is shown the theoretical shape of the energy distribution of the X rays incident on the sample. This has been calculated by assuming that the distribution follows the expression given by Kramer as described by Lindell⁽²⁾ and has been checked by measuring the distributions with a proportional counter and a single channel pulse height analyzer. Note that sufficient filtration has been employed to insure that most of the incident X rays have an energy greater than the value of the K-absorption edge of calcium. Over the range of photon energies involved, we obtain the following relations from Figure 16.

$$\begin{aligned}\mu_{Ca} &= 3.2 \times \mu_{Al} \\ \mu_P &= 1.45 \times \mu_{Al}\end{aligned}\tag{1}$$

where μ is the mass absorption coefficient of the element denoted by the subscript, in units of cm^2/g .

The absorption of X rays is given by

$$I = I_0 e^{-\mu\rho d}\tag{2}$$

where

I_0 = original intensity of the beam, a measure of the number of X rays;

I = the intensity after passing through a thickness d of a material;

μ = the mass absorption coefficient of the material, in cm^2/g ;

ρ = the density of the material in g/cm^3 ;

d = the thickness of the material, in cm;

The mass absorption coefficient, μ , is a function of X-ray energy, so that the equation as written above is actually energy dependent.

Fortunately, the energy dependence is the same for the three elements under consideration as is shown by the fact that constant ratios exist between their mass absorption coefficients over the employed range of photon energies (Equation 1).

The absorption of these X rays by a thin bone section is given by

$$I = I_0 e^{-[\mu\rho d]_{Ca}} e^{-[\mu\rho d]_P}\tag{3}$$

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Here we have assumed that the X ray absorption due to O, H, and the trace elements present is insignificant.

Now from the step wedge of aluminum which was radiographed with the bone we can determine that thickness of Al which produced the same absorption of the X rays as any given area of the bone. For this thickness we write

$$I = I_0 e^{[-\mu \rho d]_{Al}} \quad (4)$$

Now we have defined Equation 4 as being equal to Equation 3, so that under this condition we have

$$[\mu \rho d]_{Al} = [\mu \rho d]_{Ca} + [\mu \rho d]_P \quad (5)$$

Eliminate the μ 's from this equation with the aid of (1)

$$[\mu \rho d]_{Al} = 3.2 \mu_{Al} x [\rho d]_{Ca} + 1.45 \mu_{Al} x [\rho d]_P \quad (6)$$

or

$$[\rho d]_{Al} = 3.2 [\rho d]_{Ca} + 1.45 [\rho d]_P \quad (7)$$

Here we have eliminated the energy dependence and find that the numerical value of the left hand side is a measured value. If we assume that all of the calcium present in the bone is in the form of the hydroxyapatite crystal,

then we can show that

$$[\rho d]_{Ca} = 2.15 [\rho d]_P \quad (8)$$

With this relationship, we obtain from Equation 7 by substitution,

$$[\rho d]_{Ca} = 0.258 [\rho d]_{Al} \quad (9)$$

or

$$[\rho d]_{\text{hydroxyapatite crystal}} = 0.645 [\rho d]_{Al} \quad (10)$$

Hence, we have shown that a microradiograph of a thin bone section and suitable aluminum step wedge can be employed to obtain the mass per unit area of hydroxyapatite crystal in any given area of the bone.

In practice, we employ a densitometer connected to the microscope, which reads the density of a $10\text{-}\mu$ diameter spot on the microradiographic image.

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The appearance of a typical radiograph is such that the differences in the degree of calcification among haversian systems are exaggerated. That is, while wide contrast in appearance is visible between different systems, the actual difference in the degree of calcification is small. If we call the most highly calcified system 100% calcified, then no bone appears whose density is less than 75% of this value. This has previously been reported by Amprino.⁽³⁾

This leads to the question, are there regions of calcifying bone where the degree of calcification is less than 75% of the maximum value? In a typical radiograph there are no such regions, but this is the result of the nonlinear response of the microradiographic plate. One radiographic exposure will cover satisfactorily only a certain limited range of mineral densities, which is dependent upon the response of the photographic emulsion, as well as the usable range of the densitometer. To demonstrate the existence of areas of lower calcification, softer X rays must be employed, which will produce microradiographs of those areas of bone in which the density of the mineral is less than 75% of the maximum value.

Such areas appear as a narrow ring of low density around some of the larger haversian systems. These rings are semicalcified osteoid borders, which can only be demonstrated in haversian systems where the growth is rapid and the relatively uncalcified border consequently wide. The areas of tissue with less than 75% calcification are very narrow suggesting that calcification of the osteoid up to this value is a rapid process.

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HALOGEN SENSITIVE POSITIVE ION EMITTER: CHLORINE-INDUCED AND THERMAL TRANSIENTS

H. A. Schultz

Introduction

The halogen sensitive positive ion emitter is a potential tool for the study of gaseous diffusion problems of interest to this Laboratory. The latest semiannual report⁽¹⁾ summarized the background (without halogen) performance of a unit built for the further study of the mechanisms involved.

The Effect of Chlorine

The equipment was designed for a closed-vessel type of experiment in place of the continuous flow experiments undertaken heretofore. Traces of chlorine were added to the dried air used to fill the vessel. When the equipment was started under a selected set of promising conditions, it was found that the expected enhancement of the ion current occurred but that the effect was transient. The ion current approached its background value approximately exponentially, indicating that the chlorine was taken out of the reaction. The half-life was less than five minutes.

In order to study such a transient effect, it is essential to separate the chlorine-induced effect from any similar transients due to other causes. Among other possible causes are (a) some constituent of the filling gas, (b) thermal transients, and (c) something released by components of the apparatus. All three possibilities have been attacked.

The experiment has been simplified by replacing air with dried argon. The temperature control equipment has been improved. The more volatile components of the apparatus have been eliminated or replaced. The transient effects that remain are smaller and more reproducible than formerly, but their cause has not yet been established.

Thermal Problems

On the basis of previous work with other equipment,⁽²⁾ the region selected as the most promising to explore provides an ion current which is practically independent of filament temperature. The temperature of the boat (source of ionizable material) is, on the other hand, very critical since the ion current increased by about 4% per °C.

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The power supplies already in use had proved valuable for controlling the temperature during continuous operation.⁽¹⁾ There was a question, however, about their reliability during warmup, particularly in the case of the boat. The boat power supply was therefore modified as shown in Figure 17B to reduce boat temperature variation during halogen-induced transients.

The heated element is used as one arm of a phase-shifting network. A change in temperature causes a change in resistance which changes the phase of the signal applied to the thyatron to counteract the change in resistance.

When the boat, with a hot resistance of about 0.026 ohm, was used in the original circuit, variation in the resistance of the leads had a significant effect upon the operation of the network. This difficulty was removed by the use of two transformers, as shown in Figure 17B. Stability of ion current for hours at a time indicates that the degree of regulation of boat temperature is probably better than the voltmeter-ammeter method of temperature determination.

Since ion current varies exponentially with boat temperature, it is the maximum temperature at the center of the boat that is most important. This temperature is too low (about 400°C) for the optical pyrometer, and it is not practical to attach a thermocouple because of the cooling effect of the thermocouple leads and the high collecting voltage. The present calibration curve was constructed by using optical pyrometer data obtained at temperatures above the operating range plus thermocouple data for one end of the boat from room temperature up. The resistance vs central temperature curve is almost linear. Straightforward empirical interpolation gave essentially the same results as a more elaborate scheme in which

Figure 17. A. Power supply for control of filament temperature.
B. Modifications in power supply for control of boat temperature.

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reasonable empirical heat loss vs temperature relations were fed into the theoretical equation of Langmuir, et al.⁽³⁾ for the temperature distribution along a short filament.

If the unit is stopped, the gas changed, and the unit started again, the ion current starts about 20% higher than normal. This excess ion current decays to half in about 10 to 15 min., but the decay does not appear to be exponential. As a control, the unit is stopped for the same length of time, but the gas is not changed. In this case, the original ion current returns with no appreciable transient. Possible causes of this residual effect are (a) inadequate cleaning of the gas, (b) different initial temperature of the gas, and (c) physical or chemical changes within the vessel during the exhaustion of the original gas. It should be possible to eliminate the first two. The third might possibly be bypassed by injecting the halogen into the vessel without removing the other gas. This would be feasible only if the halogen does not enter into a reaction at room temperature during the time of mixing.

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SEPARATION OF CESIUM FROM URINE

J. Sedlet*

Cesium activity was separated from urine by a procedure similar to that given by Glendenin and Nelson.⁽¹⁾ Inactive cesium carrier was added to the urine and the urine ashed and dissolved in hydrochloric acid. The cesium was separated from the bulk of the salts present in urine by two precipitations of cesium silicowolframate. Preliminary experiments indicated that this procedure provided adequate separation of cesium from amounts of potassium, sodium, calcium, and magnesium in excess of those found in urine. Additional purification of the cesium followed the procedure cited. The final precipitation was made as cesium perchlorate.

The chemical yields, as determined by weighing, were 50 to 60%. To check the purity of the final cesium perchlorate precipitate, one precipitate was analyzed for cesium and potassium by flame photometry. This sample contained 28 mg of cesium (50 mg was originally added to the urine) and 2 mg of potassium. The beta activity in this sample corresponded to 77 disintegrations per minute of Cs^{137} , while 2 mg of potassium contributes only about 3.4 dpm. Blank samples were analyzed together with the urine samples to detect any activity added during the analyses. The beta activity in blank samples was less than 5% of the activity in the urine samples.

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PARTICLE ACCELERATOR--INSTRUMENTATION

H. A. May

Pulsed operation of the 250 kev Cockroft-Walton accelerator is desired for a number of experiments being planned. For investigation of the gamma activity produced by neutron activation of liquid scintillator impurities, a slow repetition rate and variable duty cycle seems indicated. A simple motor-driven cam timer controlling the ion source r.f. oscillator and/or extractor voltage should suffice. For producing millimicrosecond bursts of neutrons, beam deflection techniques⁽¹⁾ are required.

A study of our ion optics, preliminary to alterations required for pulsed operation, led to several changes. Marked improvement in the focusing of the deuteron beam at the target has been achieved by redesign of the focus electrode and changes in potential distribution. It is now possible to obtain a 70 μ amp beam distributed quite uniformly over a 7 mm spot at the target while the diffuse current collected on collimator with 10-mm diameter aperture located 50 cm in front of the target is less than 7 μ amp. With higher beam currents, the loss to the collimator becomes proportionately greater. The figures quoted are for an accelerating voltage of 150 kev.

It has been found possible to use a standardized design of ion source bottle used also by two groups in the Physics Division. Since fixtures for fabrication to close tolerances are on hand in the Glass Shop, an improvement in availability and cost results. Also, the smaller bottle diameter results in a more intense discharge and lower r.f. power for a given target current. Although no measurements have been made in verification, this seems to indicate a greater relative number of singly charged ions present, i.e., lowered recombination rate. Studies are continuing to improve and optimize the geometry of the extractor tip canal.

Design of a beam-deflection system, which can be installed at the high voltage end of the accelerating tube with minimum changes in our present electrode configuration, remains as the immediate objective. Neutron pulse width will then be measured, and klystron bunching tube techniques⁽²⁾ employed, if necessary.

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USE OF GRIDDED ION CHAMBER FOR Ra AND RdTh ANALYSIS

H. A. May

The feasibility of performing quantitative analysis of bone-extracted samples still remains to be demonstrated, due to difficulties in source preparation. In the meantime, further improvement of the detecting system has proceeded with three interrelated goals: (1) improved resolution, (2) greater stability, and (3) reduced background.

Resolution. The energy resolution of alpha-particle pulses is determined by a number of factors. It has been shown⁽¹⁾ that by proper chamber design (grid parameters, spacing to other electrodes, operating potentials, etc.) the contribution of the chamber to total energy spread of the peaks may be made negligible. Spread of pulse-height distribution caused by noise in the amplifier and by source thickness and straggling remain as the most important factors.

The first may be evaluated by introducing artificial pulses of constant amplitude at the chamber grid. With pulses equivalent to those produced by a 5-Mev alpha emitter, and an analyzer channel width equivalent to 25 kev, we obtain a distribution very nearly Gaussian, having a standard deviation of 0.65 channel widths or 16 kev. The corresponding width at half maximum is 37.6 kev.

The pulse-height distribution of any uncollimated (i.e., 50% geometry) alpha source will show an unsymmetrical low energy tail dependent upon source thickness, and hence it cannot be described entirely by a single number such as the full width at half maximum. One of our best standards, 5.5-Mev alpha from Pu^{238} , gives for this figure of merit a value of 72 kev (i.e., 1.3% resolution), but sources from standard radium solutions (with small amounts of BaCl_2 carrier) give widths of 250 to 300 kev. It is possible to reduce the half width by use of collimators, if the attendant reduction in counting rate can be tolerated, but for low activity bone-extracted samples, this is undesirable. It is apparent that further effort toward improving resolution must be concerned with improved sources.

Stability. The stability requirement, like that of resolution, is important in this application only as it affects our ability to determine total number of counts in two energy peaks a few hundred kilovolts apart with an accuracy no better than 10%. For a source with two peaks 500 kev apart, having a counting rate in each peak equal to that of the background of our chamber at present, a counting period of 100 hours is required. A slow decrease in amplifier gain has been observed, amounting to 0.1% in a few days. The rate is not constant and is believed caused by aging of the radio-grade selenium rectifiers which supply the direct current to the preamplifier

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heaters. A variable resistor now provides for manual compensation; we plan to install shortly the more reliable silicon junction power rectifiers. A degenerative feedback voltage regulated supply would be preferable, but considerations of space available and cost indicate the simpler approach.

Gain stability may also be affected by voltage fluctuations in the B-plus supply. Gas-filled VR tubes frequently develop a slow flicker, the glow discharge moving erratically back and forth on the anode surface. All VR tubes were recently replaced by the Hytron premium quality equivalents, types 6626 and 6627. None of these has shown similar failure to date.

Background. The background counting rate of the chamber has been determined frequently, and large fluctuations caused by contamination from samples being counted noted. It is believed that all thin evaporated Ra or RdTh samples lose significant amounts of radon or thoron. Two samples were prepared by evaporation and flaming, every effort being made to have them similar. The growth of activity of the daughters was followed for several days and compared with theoretical growth (growth in a sealed source). Average retention factors of 64% and 76% were found. Radon leaking from the sample may decay within the chamber before being swept out with the chamber gas. Counts from the daughter elements distributed upon inner chamber surfaces cannot be readily distinguished from true chamber background by energy analysis but can be identified by their decay time characteristics. Contamination by RdTh sources is of much greater consequence since the 54.5-sec half-life of thoron is short relative to the time for complete exchange of counter gas.

The true background from surfaces within the chamber appears to be about 9 c/hr over the energy region 5 to 9 Mev. We believe that this can be reduced considerably if the activity level of samples made available requires it.

Overnight background counts have tended to be lower than counts made during the day, and this was interpreted as evidence for occasional counting of AC line disturbances. In order to evaluate the noise rejection capabilities of line filters, power supply filters, and regulators, etc., a simple device was constructed for producing line transients controlled in amplitude and repetition rate. It consisted of an unsymmetrical multivibrator, running at about 3 pps, which was directly coupled to a relay drive stage. This relay, with a contact rating of 15 amp, was used to place a momentary heavy load upon the same line supplying the counting equipment. Short burst of r.f. may be obtained by controlling a small Tesla coil of the type used for leak-testing glass vacuum systems. If this is allowed to spark back to its own line cable, particularly strong line disturbances are available. By this testing means, the equipment has been rendered completely immune to extraneous noise sources.

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DRIFT VELOCITY OF ELECTRONS IN GASES

J. C. Bowe

The drift velocity of electrons can be determined by measuring the time duration of the current pulse induced in an external circuit by a group of electrons moving in a gas filled chamber under the influence of a uniform electric field. The time measurement is made from oscillograms obtained by photographing single trace transients of the current pulse as displayed on a Model 517 Tektronix oscilloscope. The oscillograms are accurately and relatively easily read with a front view film reader which was recently designed and constructed in our shops. Previously, the oscillograms were projected and hand traced onto graph paper using a photographic enlarger.

A typical oscillogram obtained with neon is shown in Figure 18a. The distance between the midpoints of the rising and trailing edges of the pulse is taken as a measure of the electron transit time. An examination of the trailing edge of the pulse reveals that it is much longer than the rising portion. This is presumably due to diffusion--a spreading out of the electron cloud. There is a simple relation between diffusion coefficient and mobility, which, combined with the Einstein diffusion equation, permits evaluation of the Townsend energy factor, k . This factor is defined as the ratio of the mean energy of agitation of an electron to the mean energy of thermal agitation of a gas molecule. If the electrons are in a field free space they soon come into thermal equilibrium with the gas molecules, in which case, k has the value unity. However, if an external field is present, the average equilibrium energy of the electrons is increased above the thermal energy of the surrounding gas molecules, and k may increase tenfold or a hundredfold, depending upon the particular gas and the field strength. The only values of k presently available are those determined by J. S. Townsend's method or some modification of it. In the noble gases neon, helium, and argon, for example, the values were determined over 30 years ago. It would be of considerable interest if an independent measurement of k were available.

Townsend's method consists in measuring the lateral spread of a diffusing beam of electrons. It involves the measurement of currents of the order of 10^{-12} amp. For more details, the reader is directed to Reference 1. In the present experiments, the diffusion of the electrons parallel to the electric field is evidenced by the time dispersion of the trailing edge of the current pulse.

According to Einstein's diffusion equation, a group of particles which are localized in a small volume at time $t = 0$, will at any later time, t , have a space distribution which is Gaussian. The root mean square

Figure 18. Oscillogram 18a is a current pulse in neon taken with a chamber voltage of 40 and an E/p_0 value of 0.13. The transit time was determined as $(5.4 \pm 0.1) \times 10^{-6}$ sec. Oscillogram 18b is an electronic differentiation of 18a. The dispersion time, T , was determined to be $(1.43 \pm 0.14) \times 10^{-6}$ sec. With these values the Townsend energy factor k is computed to be 15 ± 3 .

diffusion distance in a particular direction is given by

$$\sqrt{\overline{X^2}} = \sqrt{2Dt} \quad (1)$$

where D is the diffusion coefficient and t is the time of diffusion. The relation between the diffusion coefficient and the drift velocity, v , is given by

$$D = 0.0235 k \cdot v/E \quad (2)$$

where k is the Townsend energy factor and E is the electric field strength. Combining Equations 1 and 2

$$k = \left(\frac{1}{0.0235} \right) \left(\frac{\overline{X^2}}{2t} \right) \left(\frac{E}{v} \right) \quad (3)$$

Except for the mean square distance of diffusion, every quantity on the right hand side can be measured. However, the diffusion distance can be related to the observed time dispersion, T , by

$$4 \overline{X^2} = v^2 T^2 \quad (4)$$

af h

Substituting this into Equation 3 yields,

$$k = \left(\frac{V}{0.188} \right) \left(\frac{T^2}{t^2} \right) \quad (5)$$

where V is the voltage applied to the chamber.

The dispersion time, T, is obtained by measuring the difference in the width of the leading and trailing edges of the current pulse. In order to determine this quantity more accurately, the current pulse was differentiated by means of a shorted delay line circuit. The resulting waveform is shown in b of Figure 18. Calculations of k were made for neon over the range of $E/p_0 = 0.05$ to 0.9 volt/cm/mm Hg, but the resulting values were so scattered as to prohibit the drawing of a smooth curve.

The calculation for the sample oscillogram shown in Figure 18 leads to a value of $k = 15 \pm 3$. To get an idea of what order of magnitude might be expected, this can be compared with the value of 47 which was determined by V. A. Bailey, using the Townsend method.⁽²⁾ The comparison, however, is not otherwise significant inasmuch as we were unable to obtain a consistent set of values over a range of E/p_0 .

A study is now in progress to determine whether or not it is possible to read the oscillograms with sufficient accuracy and consistency as to lead to a meaningful set of k values. Simultaneously, all drift velocity data are being re-evaluated with the new film reader.

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METEOROLOGY

H. Moses

Multiple Exposure Smoke Pictures. As described in a previous report, (1, 2) a technique was developed for measuring atmospheric diffusion by taking multiple exposures of a smoke plume using two cameras. One camera took a side view of the plume and the other took a bottom view. A total of 100 usable picture pairs were obtained. All of the necessary measurements on the film have been completed. Computations to determine the smoke plume's width, height, space position, etc. on about 50 of the film pairs have been made by means of a desk calculator. The remaining computations are to be performed on the Avidac. Programming for this task has been completed.

In analyzing these measurements, values of the different parameters, n , C_z , and C_y , found in Sutton's diffusion equations were calculated. It was assumed that the boundary of the smoke plume represents a constant fraction of the concentration at the center line. As indicated earlier, there is some doubt concerning the validity of this assumption. Instead, it is assumed that the boundary of the plume represents a constant concentration surface.

Work is now in progress by the Argonne Computer Group to fit Sutton's equations to these smoke plumes using the latter assumption. The result of this work will yield the parameters n , C_z , and C_y which give the best fit. An additional parameter will also be obtained, namely the distance from the stack of the virtual origin.

It is necessary to assume a virtual origin since the expansion of the plume is very rapid within the first 5 ft. of the stack due to aerodynamic effects of the stack and the jet effect of the effluent. Since Sutton assumes a point source, a better fit of the data may be obtained by assuming a virtual origin some distance upwind of the stack.

Analysis of Pictures Taken by Type K24 Aerial Cameras The digitizing equipment allowing the reading of the space coordinates of selected points directly resulted in a tremendous saving in time. At present, a series of analyses are in progress to determine the following characteristics of the smoke plumes:

(1) The stack-induced turbulence. It is found that the area of the smoke plume increases by a factor of over 50 within the first 5 ft. after the smoke is emitted from the stack. This is very likely due to both the turbulence induced by the stack itself and the turbulence of the jet coming out of the stack.

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(2) The meander of the smoke plume. There is some question as to what is meant by the meander of the smoke plume. When one looks at a plane or side view of the smoke plume, one sees that the smoke has sinusoidal characteristics or sometimes a section of a smoke plume may even form a U-shaped configuration. These deviations are due to larger drafts in the atmosphere which will take one section of the smoke plume and deform it. In a true meander every particle within the plume would have to travel through the deviate course; however, in the case of the smoke plume only a section of the plume undergoes the main deviation, and the succeeding particles do not follow the same path. In this sense it is not a true meander. An attempt is being made to study in detail the meander mechanism.

(3) The growth rate of the plume. Measurements are made of the plume's diameter at various distances from the stack. The quantities $\frac{1}{A} \cdot \frac{dA}{dt}$ and $\frac{1}{A} \cdot \frac{dA}{dx}$ are calculated. A is the area of the plume, x is the distance along the plume from the stack, and t is time.

These calculations are quite laborious since we have many pictures and many measurements have to be made on each picture. It is planned to place the film measurements on IBM cards and have the computations made by the IBM computers.

(4) The height of rise of the plume. At any given time the height of rise is determined by the ratio of stack velocity to wind velocity, atmospheric stability, wind speed, surface roughness, and stack effluent temperature. All of these parameters are measured with the exception of surface roughness. However, this parameter is a function of wind direction at any given location. It should therefore be possible to obtain a relationship between the height of rise of the smoke plume and these parameters.

An examination of the K24 photographs, however, indicates that in any one series the height of rise varies between consecutive pictures taken only seconds apart. This is true when unstable conditions are present. Under these conditions averages have to be taken.

Automatic Data Processing

The problem of punching IBM cards directly from the meteorological sensing elements was investigated in detail during the past six-month period. Such an automatic system would greatly expedite the processing of data since it would eliminate the expenditure of time for reading off the data from the recorder charts and for key punching them to prepare IBM cards. With a system of this kind, not only would the over-all accuracy be increased since the human element is eliminated, but also more data would be analyzed than is now possible with the manpower at our disposal.

The measurements to be included in this system are those which can be recorded electrically. These are measurements of wind speed, wind direction, ambient temperature, solar radiation flux, soil temperature, and atmospheric stability.

Tables 12A through 12D show the types of measurements that are being made and the form in which the punched data is desired. The processing of the meteorological data has become economical only through the availability of commercially produced components which have appeared on the market during the past few years. A general scheme for the processing of the data is shown in the block diagram, Figure 19.

There are several features necessary to this system which increase the initial cost. These are:

(1) Necessity for averaging most of the meteorology data. Since the meteorological variables such as wind and temperature are rapidly varying quantities, it is necessary to average these measurements over a time period. In the past, use has been made of 10-min. averaging periods beginning at 5 min. before the hour and ending at 5 min. after the hour. The variables of solar radiation and radiation flux values are averaged for the entire hour beginning on the hour. In order to study the relations among variables such as wind profiles, that is, the wind measurements at different levels, it is necessary that the averaging for each of the measurements be made simultaneously. This requires separate averaging units for each instrument.

(2) Thermocouple scale linearization. Difficulty arises from the fact that the e.m.f. output from copper-constantan thermocouples is not proportional with temperature. Compensation for nonlinearity is obtained by resistive padding of retransmitting slide wires. Retransmitting slide wires are essentially voltage dividing resistors or potentiometers upon which a controlled DC voltage is impressed. This slide wire is mechanically coupled to the existing electronic potentiometer recorder but is isolated from it electrically. The DC voltage output of the retransmitting slide wire is proportional to the value of the measured variable.

An important component of the system which has to be specially designed for this installation is the programmer. The essential parts of the programmer consist of a synchronous motor which drives cams to activate microswitches or stepping switches. These connect each of the variables in turn into the system so that its value may be recorded on an IBM card.

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TABLE 12A

Meteorological data characteristics to be considered in
design of automatic data processing system

	Wind speed	Wind direction
1 Type of recorder	Esterline-Angus	Esterline-Angus
2 Number of recorder	6	6
3 Sensitive element	Propeller operating DC generator	Wind vane operating Mark I Synchro
4 Character of chart trace	Fluctuating--fluctuations exceeding 10% of full scale possible. Frequency about 1 cycle/sec	Fluctuating--fluctuations exceeding 10% of full scale possible. Frequency about 1 cycle/sec
5 Linearity of output	Linear	Linear
6 Form of data	10-min average once per hour	10-min average once per hour
7 Time interval of average	From 5 min before hour to 5 min after hour	From 5 min before hour to 5 min after hour
8 Number of points for average	A minimum of 100--one every 6 sec	A minimum of 100--one every 6 sec
9 Output of sensing element	0-11 volts	0-360°--angular position of a shaft
10 Accuracy of data	Two-figure accuracy	Two-figure accuracy
11 Final form of data	0-99 mph punched on IBM cards	Tens of directional degrees: Nos. 1-36 punched on IBM cards

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TABLE 12B

Meteorological data characteristics to be considered in
design of automatic data processing system

	Ambient temperature	Temperature difference between two levels
1 Type of recorder	Brown Elektronik Potentiometer (strip chart)	Brown Elektronik Potentiometer (strip chart)
2 Number of recorders	1	1 - 4 separate layers measured on a single point recorder--stepping switch
3 Sensitive element	Thermopile of 5 copper-constantan thermocouples	Single copper-constantan thermocouple
4 Character of chart trace	Fluctuating--fluctuations exceeding 10% of full scale possible. Frequency about 1 cycle/sec	Fluctuating--fluctuations exceeding 10% of full scale possible. Frequency about 1 cycle/sec
5 Linearity of output	Nonlinear--copper-constantan thermocouple compensation necessary	Nonlinear--copper-constantan thermocouple compensation necessary
6 Form of data	10-min average once per hour	4 2-min averages once per hour
7 Time interval of average	From 5 min before hour to 5 min after hour	1--from 5 min before hour to 3 min before hour 2--from $2\frac{1}{2}$ min before hour to $\frac{1}{2}$ min before hour 3--from the hour to 2 min past hour 4--from $2\frac{1}{2}$ min past hour to $4\frac{1}{2}$ min past hour
8 Number of points for average	A minimum of 100--one every 6 sec	A minimum of 20 points for each 2-min average
9 Output of sensing element	Approximately 8 mv range	-.5 to +0.2 mv
10 Accuracy of data	Three-figure accuracy	Two-figure accuracy
11 Final form of data	-20 to +20° C--Range 1 0 to 40° C--Range 2	From +9.9 to -5.0 punched on IBM cards

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TABLE 12C

Meteorological data characteristics to be considered in
design of automatic data processing system

	Temperature radiation flux plate	Radiation flux
1 Type of recorder	Brown Electronik Potentiometer (circular chart)	Brown Electronik Potentiometer (circular chart)
2 Number of recorders	1	1
3 Sensitive element	Single copper-constantan thermocouple	Beckman and Whitley Radiometer (bank of copper-constantan thermocouples)
4 Character of chart trace	Nearly smooth	Either smooth or fluctuating with large amplitude
5 Linearity of output	Nonlinear--copper-constantan thermocouple compensation necessary	Linear
6 Form of data	Instantaneous reading once per hour	1-hr average recorded once per hour
7 Time interval of average		1-hr average punched out on the hour
8 Number of points for average		A minimum of 600 points--one every 6 sec
9 Output of sensing element	Approximately 2 mv range	-5 to +15 mv
10 Accuracy of data	Two-figure accuracy	Three-figure accuracy
11 Final form of data	0 to 99° F punched on IBM cards	-0.50 to +1.20 langley's/min

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TABLE 12D

Meteorological data characteristics to be considered in
design of automatic data processing system

	Solar radiation	Soil temperature
1 Type of recorder	Leeds and Northrup Micromax Potentiometer (strip chart)	Leeds and Northrup Micromax Self-Balancing Resistance Bridge (strip chart)
2 Number of recorders	1	1-8 levels and reference are recorded in sequence which is programmed by a stepping switch
3 Sensitive element	Eppley Pyrheliometer (50-junction thermopile)	100-ohm copper thermohms
4 Character of chart trace	Either smooth or fluctuating with large amplitude	Smooth
5 Linearity of output	Linear	Linear
6 Form of data	1-hr average recorded once per hour	Instantaneous readings in the following sequence: 1--1 min past hour 2--3 min past hour 3--5 min past hour 4--7 min past hour 5--9 min past hour 6--11 min past hour 7--13 min past hour 8--15 min past hour
7 Time interval of average	1-hr average punched out on the hour	
8 Number of points for average	A minimum of 600 points--one every 6 sec	
9 Output of sensing element	0 to 15 mv	0° to 60° C, 90.15 to 113.79 ohms; and -30° to +30° C, +78.33 to 101.97 ohms
10 Accuracy of data	Three-figure accuracy	Three-figure accuracy
11 Final form of data	0.00 to 2.00 langleys/min	0.00 to 60.0° C and -30.0 to +30.0° C; punched on IBM cards

Figure 19. Block diagram of automatic data processing system
 Meteorology Group Argonne National Laboratory

The output from the selector switches is fed into a high impedance amplifier. This output is adjusted for scale when necessary, and fed into a self-balancing potentiometer. The angular rotation of a shaft on this potentiometer corresponds to the value of the meteorological parameter to be measured. An analog digital converter is coupled to this shaft, the output of which goes to a diode matrix. This matrix translates the output of the digital converter to impulses necessary to punch the meteorological information on IBM cards.

Perhaps the most difficult part of the entire system is the averaging function. This process is the one process which has not been standardized among the manufacturers of data processing equipment. For example, the Brown Instrument Company uses an RC network to charge a condenser for a certain period of time. The condenser output is then related to the meteorological parameter average. IBM and the Metrotype Corporation uses voltage pulses to build up the voltage in a series of counters. When the sum of the voltages from the pulses equals the measured voltage, a null balance detector recognizes this condition and sends the voltage into an accumulator. This process is repeated once every several seconds or more frequently over the measuring period. The total voltage in the accumulator is then directly proportional to the average of the meteorological measurement.

Other companies such as Fischer and Porter Company or the Panellit Corporation use a cam driven by a synchronous motor with a cam follower whose position is related to the value of the parameter measured. The cam makes one revolution every 15 or 30 sec. The cam follower makes contact with the cam for a fraction of the period depending upon the value of the meteorology parameter. As long as contact is present between cam and cam follower, a contactor will move on a slide wire. An amount proportional to the value of the measured variable is the distance moved. After a predetermined number of cam rotations, the contactor has moved the same number of increments on the slide wire. The total distance moved is proportional to the value of the meteorological parameter measured.

A fourth method, suggested by the General Precision Laboratory, was to use a velocity servo whose speed is proportional to the value of the measured variable. The total number of rotations of the servo-motor shaft is then proportional to the average.

At the present time the condenser method appears to be the most practical though the accumulator method is also a possibility.

There are also a number of analog to digital convertors in the field. Such units are manufactured by the Giannini Corporation and the Coleman Engineering Company; IBM also makes an analog to digital convertor.

Although the entire system is basically simple, there is a considerable amount of detail and planning involved in order to provide satisfactory data. For example, provision has to be made for handling negative quantities. Also, provision has to be made for properly punching the cards when the sensing element is inoperative. A clock mechanism must also be provided for punching the date and time group together with special codes to identify the meteorological data being punched.

Some thought has been given to the form in which the data should be punched. Paper tape, magnetic tape, and IBM cards were considered. In view of the fact that we have over 6 years of data on IBM punch cards, it appears desirable that we should continue punching data on cards rather than on tape.

The Five-Year Climatological Report

A substantial amount of time during this six-month period was spent on the preparation of the five-year climatological report. The main purpose of this report is to provide data based on the meteorological measurements made at Argonne for use in planning and operations by engineering and scientific groups of the Laboratory and for climatological research. This report covers the measurements made of wind speed and wind direction at two levels, temperature, atmospheric stability, precipitation, pressure, relative humidity, solar radiation, and soil temperature.

The report is divided into three parts: In the first part are presented data concerning single variables. The tabulations and material presented are similar to those presented in other climatological reports both at Argonne and elsewhere. Tables are shown of daily or hourly averages, frequency distributions of such variables as maximum and minimum temperatures, and also tabulations of the number of days on which a given value of some meteorological variable was exceeded, such as the number of days with precipitation greater than one inch or the number of days with temperatures exceeding 90°F.

In the second part a series of tabulations not customarily found is presented. In these the interrelationship between meteorological variables, such as solar radiation and wind, or atmospheric stability, precipitation, and wind are shown. The use of the IBM tabulating and computing equipment has made such tabulations possible where heretofore the amount of labor would have been prohibitive.

In the third part of this report the data obtained at the Chicago Midway Airport are compared with similar data obtained at Argonne. In general, the difference between the variables was obtained for each hour of the day, and frequency distributions were tabulated for these differences either by the hour, by month, or as a function of some other

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variable. For example, a table is shown giving the frequency distribution of the difference in temperature between Argonne and the Chicago Midway Airport as a function of the temperature at Argonne. Other tables are presented in which individual variables are presented in the form of frequency distributions. For example, there is a series of tables, one for each month, and a summary table giving the joint frequency distribution of the wind direction to 16 points at the Chicago Midway Airport and the Argonne National Laboratory. These tables show for any one direction, at one location, the frequency distribution of directions recorded at the other location.

There have been a number of special tables included at the request of operating groups in the Laboratory. For example, there are tables showing the occurrence frequency of wet bulb temperatures exceeding specified amounts such as 70°F, 75°F, etc. Also, there is a set of tables showing the joint frequency distribution of temperature and dew point. Special calculations were necessary to obtain these tables since the Meteorology Group measures only relative humidity. The dew point for each hour had to be calculated from the dry bulb temperature and relative humidity measurement.

At the present time all of the tables have been completed, and a few errors are being checked. Overlays and titles have been prepared. This report, ANL-5592, will be ready for submission to the Technical Information Division and Graphic Arts in the near future.

Temperature Variations As A Function of Horizontal Distance

In order to obtain a better understanding of the motion and structure of the atmosphere, an attempt was made to determine the character of the thermal eddies in the atmosphere. This was done by placing two thermocouples made of #36 copper and constantan wire at varying horizontal distances apart, i.e., one-half inch, one inch, two inches, etc., up to 100 feet. The temperature difference between the two thermocouples was recorded on the Brown Electronik Potentiometer currently used for recording the temperature difference in the vertical direction. The recorder was well suited to measuring temperature differences of the order observed in the horizontal direction. Figure 20 shows a record obtained on January 9, 1956 with the thermocouples mounted one meter above the ground. The thermocouples were mounted crosswind. During this time the temperature ranged from 32-34°F and the wind at 79 inches was N-NW, 7 miles per hour.

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Figure 20. Temperature difference between two points separated horizontally and located one meter above ground.

It is rather surprising to note that over distances as small as three inches, temperature differences of the order of over 1°F or $1/2^{\circ}\text{C}$ occurred. At one foot separation, temperature differences of 2°F were not uncommon. In this run the maximum was reached at about 40 feet although measurements were made out to 100 feet. This decrease with distance was due to the fact that the sun was going down and the turbulence in the atmosphere was decreasing. The run was made on a sunny day when considerable convection was present. A similar run made on a cloudy day indicates that the atmosphere does not contain such sharp gradients.

Instrumentation

The following is a summary of the additions and improvements to our meteorological instrumentation used both in connection with the investigations of atmospheric diffusion and the climatology program.

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Improved Synchronization of Wind Recorders. At the present time, wind is measured at six different locations at the Meteorology Laboratory. Four of these locations are on the tower. The fifth instrument is mounted at 79 inches above the ground southwest of the Tower, and the sixth is on a 19-foot pole northwest of the Meteorology Tower. Each instrument requires two recorders, one for direction and one for speed. There are, therefore, a total of 12 recorders providing wind information. To improve the synchronization of the recorded information, synchronous motor drives have replaced the spring-driven chart drives. One synchronous motor is used to drive the charts of four recorders. In each group the motor is located in one of the recorders and the other three are mechanically coupled to it.

To improve synchronization further, a chronograph marking pen was installed on the six wind speed recorders. The six wind direction recorders were equipped with these previously. Each side marking pen is connected to a control timing mechanism. This timer is programmed so that a ten-minute pip begins at five minutes of the hour and ends at five minutes past the hour in each left hand margin of the recorder. On the half hour a ten-second pip is made. This scheme greatly facilitates matching the charts from the various recorders.

Solar Radiation Flux. The measurements of solar radiation flux have been transferred from the adjustable span, adjustable zero Brown recorder to a conventional type of Brown recorder which we have modified to have a range from -5 to +15 millivolts. Special chart paper and a special scale range have been purchased so that the net radiation flux in langleys per minute can be read directly from the charts. The chart range is from -0.5 to +1.2 langleys per minute.

The Photogrammetric Plotter. In order to facilitate the analysis of the smoke pictures from the K24 aerial cameras, a system for digitizing the data was devised.

In the photogrammetric technique using the K24 cameras, a smoke plume is photographed from opposite sides by the cameras. These cameras are mounted on a 400-foot base-line and are elevated 30° with respect to the horizontal. To analyze the data, the film is reprojected on opposite sides of semitransparent screen. The images coincide only if the smoke plume lay in the vertical plane bisecting the base-line. As is usually the case, the smoke plume meanders or deviates from this plane. Sections of the images from the two films can then be brought into coincidence by moving the screen toward one of the projectors. A coordinate system is used in which x is along the plume, z is vertical, and y is crosswind to the plume. The origin is at the stack top. The x and z values of any point on the plume can be determined by measuring distances on the screen; cursors are used for this purpose. The y coordinate is obtained by measuring the amount of displacement of the screen from the half-way position between the projectors.

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Digitization was accomplished by connecting synchro-generators to each of the motors driving the screen and the cursors. Each synchro-generator was coupled to a separate synchro-receiver which was in turn coupled to a Veeder-Root counter. In this manner the x, y, and z coordinates of any point on the film where the smoke plume images coincided could be read directly in digital form. The speed of analyzing these data has been markedly improved since this system has been installed.

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