ANL-5919
Biology and Medicine
(TID-4500, 14th Ed.)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
P. O. Box 299
Lemont, Illinois

RADIOLOGICAL PHYSICS DIVISION
SEMIANNUAL REPORT

January through June, 1958

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

September, 1958

Preceding Report:

ANL-5829 - July through December, 1957

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
United States Atomic Energy Commission

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

	<u>Page</u>
The absorbance change of irradiated organic solvents. Part VI	
I. B. Berlman	5
Drift velocity of electrons	
J. C. Bowe	7
Progress report on flux and spectrum measurements of the cosmic ray neutron background	
R. Grismore, L. D. Marinelli and B. G. Oltman	11
Variable energy neutron source	
R. Grismore, B. G. Oltman and L. D. Marinelli	14
Radium toxicity studies	
R. E. Rowland, J. H. Marshall and J. Jowsey.	19
The effect of parathyroidectomy on haversian remodeling of bone	
Jenifer Jowsey, R. E. Rowland, J. H. Marshall and F. C. McLean	27
Calculation of the possible effects of calcium diffusion in bone crystallites	
J. H. Marshall and R. E. Rowland	35
Positive ion emission	
H. A. Schultz	47
Determination of Sr^{90} in biological materials	
D. P. Krause	53
Determination of Ac^{228}	
D. P. Krause and A. F. Stehney.	54
On the stratospheric Sr^{90} fallout	
P. K. Kuroda	55
Illinois municipal water supplies having unusually high concentration of Ra^{226}	
H. F. Lucas, Jr.	58
Gamma-ray emitting radioactivity in soil	
P. F. Gustafson	61
The human spectrometer. Introduction	
C. E. Miller	64

TABLE OF CONTENTS

	<u>Page</u>
I. The gamma-ray background of Iron Room I (airborne radioactivity) C. E. Miller and J. B. Corcoran	64
II. Ra^{226} and Th^{232} activity in humans C. E. Miller	66
III. Techniques which may yield more accurate Ra^{226} measurements <u>in vivo</u> C. E. Miller and O. J. Steingraber.	67
IV. Follow-up of U^{235} inhalation incident C. E. Miller	71
V. The elimination pattern of Co^{60} acquired by inhalation C. E. Miller and H. A. May	72
VI. <u>In vivo</u> measurement of Cs^{137} C. E. Miller and L. D. Marinelli	74
Some investigations on the source of background counts in NaI(Tl) crystals P. F. Gustafson, L. D. Marinelli and S. S. Brar	77
On the nature of the 3.25-Mev peak in large crystals H. A. May	85
Background of Iron Room II H. A. May	89
Evaluation of RCA 5-inch tube, Type 7046 H. A. May	91
Optimum performance from large NaI(Tl) crystals H. A. May	92
[#] The effect of meteorological variables upon the vertical and temporal distributions of atmospheric radon H. Moses, A. F. Stehney and H. F. Lucas, Jr.	102
Argonne Meteorological Model Towing Tank P. Frenzen	113

TABLE OF CONTENTS

	<u>Page</u>
Atmospheric diffusion studies	
Peter E. Wasko and Harry Moses	125
Staff activities	139
Publications	140

THE ABSORBANCE CHANGE OF IRRADIATED ORGANIC SOLVENTS PART VI.*

I. B. Berlman

In previous reports (see, i.e., Fig. 16 of Part IV) it was noted that the dose-effect relationship of absorbance in γ -ray irradiated xylene depended on whether the sample was aerated or evacuated during irradiation. Noteworthy was the fact that irradiation in vacuo led to the presence of structure in the absorbance curve in the vicinity of 3400 Å and that variability in results of an evacuated system (see Part V) was as high as 30%. This report deals with the elucidation of these phenomena.

As reported in Part V, E. J. Hart suggested that ozone formation in the aerated samples could have been responsible for our results. The conditions of irradiation, in fact, favored the hypothesis because the samples were being irradiated in uncapped vials to let air diffuse into the solution during irradiation in an attempt to maintain aeration. Obviously, if ozone was being produced in the vicinity of the xylene during irradiation, it too could diffuse into the solution. The following experiment was performed to test the effect of ozone on unirradiated and irradiated xylene. Ozone from an ozonator was bubbled through freshly distilled xylene. The result was that the absorbance increased in the region of 3000 Å. This is the same type of absorbance increase observed when oxygen is bubbled through xylene. However, when ozone was bubbled for a short time through vacuum-irradiated xylene the radiation induced absorbance peaks in the 3400 Å region were drastically reduced. This result points to one of the external causes for a difference in the absorbance change produced in aerated xylene and evacuated xylene.

Another source of trouble has been found in the nitrogen-oxygen compounds such as nitric oxide, etc., which are formed in the air during irradiation. To test their effect on freshly distilled xylene and on irradiated xylene the following procedure was followed. Some nitric oxide was momentarily bubbled through freshly distilled xylene. The xylene assumed a yellowish cast. This was the same yellowish appearance that aerated xylene assumes after a dose of about 10^{10} ergs/cm³. Vacuum irradiated xylene does not assume this yellowish cast after a dose of 10^{10} ergs/cm³. Also when nitric oxide was released in the immediate vicinity of a vacuum-irradiated solution, the absorption peaks of the irradiated xylene in the 3400 Å region were also markedly reduced. Thus nitric oxide gas is a second possible source of interference in absorption studies.

*Part I. ANL-5518, p. 8 (1956) Part IV. ANL-5755, p. 44 (1957)
 Part II. ANL-5596, p. 3 (1956) Part V. ANL-5829, p. 5 (1957)
 Part III. ANL-5679, p. 5 (1957)

To avoid the effects of ozone, nitric oxide and other possible harmful gases, the doubly distilled xylene solutions were bubbled with nitrogen and placed in glass-stoppered vials before irradiation. These precautions resulted in absorption spectra identical to those produced under vacuum conditions. The reproducibility was further improved by passing the pre-purified nitrogen gas over hot copper to remove any residual oxygen,* through a cold charcoal trap to remove remaining gases and vapors, and then by bubbling it through a trap containing doubly distilled xylene.

With these improvements, the reproducibility of the absorption data is approximately 10%.

The glass-stoppered vials provided a secondary advantage in that the bleaching of the absorption after irradiation was reduced. By keeping the irradiated xylene in the stoppered vials and in the dark, the absorbance decreased less rapidly than when the xylene was open to air after γ -ray exposure.

The author would like to express his thanks to Mr. M. Zarr for his aid in making the absorbance measurements, and to Miss P. Walsh for aid in exposing the samples to the gamma-ray source in the Chemistry Building.

*Dr. S. Gordon has just indicated that hot copper sometimes produces NO and that a better procedure would be to follow that outlined on pp. 166-167 of "Experimental Inorganic Chemistry," by R. E. Dodd and P. L. Robinson.

DRIFT VELOCITY OF ELECTRONS

J. C. Bowe

Progress Report

A critical examination of the results previously published has been undertaken for the purposes of possible refinement and clarification.

A. Theoretical

The computed values of electron drift velocity and energy distribution function presented in ANL-5829 were obtained under the assumption that the gas molecules were at rest prior to collision with the electron. This assumption is valid if the mean energy of the electrons is large compared with the thermal energy of the gas molecules. However, at very low values of E/p_0 , it is possible that this condition is not satisfied. In order to check the sensitivity of this condition, hand calculations of $\bar{W}$ were carried out at the lowest E/p_0 values for nitrogen and argon, taking the motion of gas molecules into account. A smaller value of $\bar{W}$ was obtained for both gases. The previous value of 0.055×10^6 cm/sec for nitrogen at $E/p_0 = 0.01$ became 0.046×10^6 cm/sec, and in argon, at $E/p_0 = 0.005$, the value of 0.097×10^6 cm/sec was changed to 0.060×10^6 cm/sec. Further calculations over a range of E/p_0 values for each of the gases are being performed by the Applied Mathematics Division.

When the motion of gas molecules is taken into account, the distribution function⁽¹⁾ is

$$f_0(\epsilon) = A \exp. \left[\frac{-6m}{M(eE)^2} \int_0^\epsilon \frac{\sigma_t^2(\epsilon) \epsilon d\epsilon}{\frac{6m \sigma_t^2(\epsilon) \epsilon kT}{M(eE)^2} + 1} \right]$$

where k is the Boltzmann constant, T the gas temperature, m and M are the electron and atomic masses, respectively, ϵ and e the electron energy and charge, respectively, E the electric field strength and $\sigma_t(\epsilon)$ the transport cross section. Using this function, the expression for the average velocity of drift due to the electric field is found to be

$$\bar{W} = \left(\frac{16\pi}{mM eE} \right) \int_0^\infty \frac{\sigma_t(\epsilon) \epsilon^2 f_0(\epsilon) d\epsilon}{\frac{6m \sigma_t^2(\epsilon) \epsilon kT}{M(eE)^2} + 1}$$

If $kT \ll \frac{M(eE)^2}{6m \sigma_t^2(\epsilon) \epsilon}$, these formulae become identical to those used in the previous calculations.

B. Experimental

It seems desirable to establish the justification for equating the electron drift time to the time duration of the current induced in the external circuit. Accordingly, the following topics are considered:

- a) the ideal current pulse and its distortion by the input circuit,
- b) transient energy distribution, and c) the effects of diffusion.

a) Distortion of the Signal by the Input Circuit

The current induced in an external conductor due to a group of electrons drifting with constant speed W across the gap of a parallel plate condenser is given by

$$i(t) = \frac{N(t) e W}{d}$$

where d is the plate separation and $N(t)$ is the total number of electrons in the group at time t . It should be noticed that the magnitude of the current is independent of the spatial distribution of electrons. In the ideal case where N electrons are instantaneously liberated from the cathode with an energy distribution characteristic of the impressed electric field, the current rises as a step function and remains constant until the electrons cross the gap. Upon reaching the anode, the current instantaneously returns to zero if no diffusion took place. The width of this square current pulse clearly represents the electron drift time. If diffusion is taken into account, the trailing edge of the pulse is no longer square. The drift time is now determined exactly by the interval between the leading edge and the midpoint of the trailing edge because of the symmetric diffusion of the electron cloud.

A situation which more nearly approximates the actual experimental conditions may be described as follows. Let N electrons be liberated from the cathode at a uniform rate for a time t_1 . The ideal current pulse is then of the form of a trapezoid with the leading and trailing edges having the same time duration t_1 if the effect of diffusion is neglected. The drift time is now given by the interval between corresponding points of the two edges, and for convenience, the midpoints may be selected. If diffusion is taken into account, the time duration t_2 of the trailing edge will be greater than t_1 , and in this case the drift time is given exactly only by the distance between the midpoints of the two edges. This time interval can be interpreted as the drift time of a median plane perpendicular to the electric field and dividing the electron cloud in half. The time coordinates of the midpoints determine when half of the electrons have been released from the cathode or have reached the anode. Using this method of time measurement, no correction need be made for diffusion if the initial spatial distribution of electrons is uniform.

Assuming that the ideal current pulse has a trapezoidal shape, the distortion introduced by the RC input circuit can be investigated. If $RC \ll t_1$, the leading and trailing edges will be delayed by a time RC and the drift time is determined exactly by the time coordinates of the mid-points. If $RC \gg t_2$, but less than the drift time, the edges of the distorted pulse are approximately exponential. In this case, some correction to the measured pulse width may need to be made, especially if t_2 is much different from t_1 .

In the present experiments, RC is about one-half of t_1 , and the dependence of pulse width on RC was therefore investigated. Since the same results, within the experimental error, were obtained when RC was decreased by a factor of ten, no corrections to the drift time were made.

b) The Equilibrium Distribution

If the distance of advance from the cathode required for the photoelectrons to come to the equilibrium energy distribution characteristic of the impressed field is a significant fraction of the total drift distance, a correction to the measured time should be applied.

In the present experiment, the energy distribution of the photoelectrons is not known. Energies as high as 2.5 ev may be present. The photoelectrons are emitted in the forward direction symmetrically distributed about the normal to the surface.⁽²⁾ After a few mean free paths, the motion of the photoelectrons can be considered random, but with an average energy different from that which is characteristic of the impressed field. At the pressures employed in these experiments, this random motion is achieved in less than a few hundredths of a millimeter from the cathode. The random energy then changes with time until the proper equilibrium value is attained.

Since a computation of the time required to reach equilibrium embodies many assumptions and simplifications, the results obtained can be misleading. Therefore, an empirical approach toward the evaluation of this factor was considered more appropriate. Such an approach is possible because the equilibrium distance from the cathode is pressure dependent. However, the results obtained at the various pressures employed in these measurements were in agreement within the experimental error indicating that the correction was negligible. No measurements were made at the very low pressures where the correction might become significant.

c) Diffusion Effects

Diffusion causes no difficulty in the determination of drift time provided that the initial spatial distribution of electrons is uniform. If this

is the case, diffusion does not cause the median plane to be displaced. Should the initial spatial distribution be non-uniform, the median plane becomes displaced because of the diffusion current, and a correction to the drift time must be considered.

In these measurements, the initial spatial distribution of electrons is not known. However, since the diffusion distance of the median plane is pressure dependent, the effect, if it exists, can be detected. No pressure dependence in the measured drift velocities was observed, indicating that the effect is small, and therefore no corrections for it were made.

Acknowledgement is due to Dr. F. F. Rieke of Midway Laboratories for an interesting discussion which helped to clarify and organize much of the material presented here.

References

1. S. Chapman and T. G. Cowling, The Mathematical Theory of Non-Uniform Gases, p. 350, Equation 13 (altered notation). Cambridge: Cambridge University Press, 1939.
2. A. L. Hughes and L. A. DuBridge, Photoelectric Phenomenon. New York: McGraw-Hill Book Co., Inc., 1932.

PROGRESS REPORT ON FLUX AND SPECTRUM MEASUREMENTS OF THE COSMIC-RAY NEUTRON BACKGROUND

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

A recent report⁽¹⁾ on measurements of the cosmic-ray-produced neutron flux as a function of altitude has aroused speculation that the neutron dose being received by contemporary man per year may be larger and a more important fraction of the total natural dose than had been previously supposed. The resolution of this question is, of course, of great importance in connection with studies designed to determine the effects of low-level radiation on man through attempting to observe variations in the incidence rate of possible effects as a function of variations in the natural radiation background.

The "twin" liquid scintillation fast neutron spectrometer^(2,3) already in use in this laboratory should be ideally suited to comparing the natural neutron and gamma-ray dose rates at intermediate energies because of (1) its high efficiency, (2) its ability to separate the neutron and gamma-ray spectra, and (3) the resemblance of its xylene detecting fluids to tissue. In connection with other studies,⁽³⁾ overnight background runs were made with this instrument in a room on the first floor of Building D-203 about a year ago. The pair of matched runs of the ordinary xylene (hydrogen-rich) and fluorinated xylene (hydrogen-deficient) scintillation cells taken in one such measurement is shown in Fig. 1 below. Both cells were operated with one-quarter inch thick lead shields about the scintillators in these runs. The data from the 256-channel pulse height analyzer have been combined into eight-channel subtotals in the plot, and error bars indicating the statistical standard deviations have been attached to the points. The ratio of the operating times of the two cells is that required to yield a good match between their spectra when they are run at equal distances from a Cs^{137} gamma-ray source. Unfortunately, the two cells had to be operated on different nights in taking the background data of Fig. 1 since sufficient electronic equipment was not then available for simultaneous measurements. While the spectrometer has not yet been calibrated in terms of energy, it is believed that the gamma-ray energy range covered by these data is of the order of 0.1 to 1 Mev, and the neutron energy range of the order of 0.5 to 3 Mev. Thus, it is seen that the cosmic-ray-produced neutron background, represented by the difference between the spectra of the two scintillators, is appreciable in size in comparison with the gamma-ray background, some measure of which is afforded by the spectrum of the fluorinated xylene cell. In making such comparisons it should be remembered that the gamma-ray and neutron spectra must be displaced relative to one another to compare them energywise, and that the channel widths do not have the same meaning in terms of energy in the two cases.

Figure 1. Neutron and gamma ray background spectra measured with the "twin" scintillation fast neutron spectrometer cells in room B-110, building D-203; 1/4-in. lead shields about the scintillators.

In order to follow up the above interesting results, a program has been initiated to measure the natural neutron and gamma-ray backgrounds under various conditions with the "twin" scintillation spectrometer. Additional electronic equipment has been assembled to permit simultaneous operation of both the fluorinated and ordinary xylene cells. An Argonne-type 256-channel pulse height analyzer with a split memory feature is now available which will analyze the spectra from both cells at once. The electrical circuits for the photomultiplier tubes observing the scintillation fluids have been changed over for positive photomultiplier high voltage operation so as to reduce further the already low probability of chance noise coincidences. To date, preliminary runs have been made in the basement of Building D-203 both inside and outside of the second Iron Room facility^(4,5) located there. The Iron Room provides a convenient check on the results by decreasing the gamma-ray background relative to the cosmic-ray neutron background by a known amount. The runs which have been made so far in the basement seem to provide a qualitative check on the results of Fig. 1. However, the data are not being presented at this time because of certain anomalies apparently arising from the untried character of some of the new electronic equipment.

References

1. H. W. Patterson, W. N. Hess, B. J. Moyer and R. W. Wallace, The Flux and Spectrum of Cosmic-Ray-Produced Neutrons as a Function of Altitude, UCRL-8208 (University of California Radiation Laboratory, Berkeley, California, May 23, 1958).
2. I. B. Berlman and L. D. Marinelli, Rev. Sci. Instr. 27, 858 (1956).
3. R. Grismore, P. F. Gustafson, B. G. Oltman and L. D. Marinelli, Radiological Physics Division Semiannual Report, July through December, 1957, ANL-5829 (Argonne National Laboratory, Lemont, Ill., February, 1958) pp. 39-40.
4. C. E. Miller, L. D. Marinelli, R. E. Rowland and J. E. Rose, Nucleonics 14, No. 4, 40 (1956).
5. C. E. Miller, Radiological Physics Division Semiannual Report, July through December, 1957, ANL-5829 (Argonne National Laboratory, Lemont, Ill., February, 1958) pp. 144-149.

VARIABLE ENERGY NEUTRON SOURCE

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

The lack of a simple, relatively inexpensive source of monoenergetic fast neutrons with energy variable through the range of biological interest has been a factor retarding the forward progress of the field of neutron dosimetry. The requirements which such a source should meet have been listed in a previous report.⁽¹⁾ The proposed plan for developing such a source using a low-voltage charged particle accelerator and a set of charged particle-neutron reactions was also discussed therein.

In the period since the previous report⁽¹⁾ studies have been initiated to assess the usefulness for the source of the $\text{Si}^{28}(\text{d},\text{n})\text{P}^{29}$ and $\text{B}^{10,11}(\alpha,\text{n})\text{N}^{13,14}$ reactions with the 250-keV Cockroft-Walton linear accelerator. Bombarding both a natural silicon target (92% Si^{28}) and a carbon target (for a background reference) with 220-keV deuterons and using the Perlow recoil-type neutron spectrometer⁽²⁾ to compare the resulting spectra, possible evidence of $\text{Si}^{28}(\text{d},\text{n})\text{P}^{29}$ neutrons at low intensity was found. However, it quickly became apparent that the spectra of any such neutrons were being greatly overshadowed by scattered D-D neutrons resulting from a buildup of deuterium in the targets through bombardment. Using the improved "twin" liquid scintillation fast neutron detector⁽³⁾ described in the last report⁽¹⁾ large numbers of scattered D-D neutrons were also detected from a quartz target. Quartz can be used as a silicon target material with 250-keV deuterons because the Q-value of the $\text{O}^{16}(\text{d},\text{n})\text{F}^{17}$ reaction is -1.63 MeV (calculated from the masses), and was chosen because of its smooth, nonporous surface.

Comparison runs were next made between new targets of quartz, gold and zirconium which had not been loaded with deuterium previous to the series of runs. It was found that for targets which have been used for only two or three bombardments, comparable D-D neutron yield rates per microampere of deuterons are obtained from quartz and gold, and these rates are about twice that obtained from a zirconium target which has been hit with approximately the same number of microcoulombs. The quartz and gold rates observed were of the order of 5% of the rate which would have been obtained from a thoroughly-loaded zirconium target. It was also found that the D-D neutron flux rose to an appreciable figure very rapidly after the beginning of the bombardment of completely new targets. These observations were made with the "twin" scintillation fast neutron detector and with a Hanson and McKibben long counter.⁽⁴⁾

The above demonstration of the relatively high D-D neutron yields which can result from deuterium loading of a target during bombardment pointed up the need for a means of combatting this effect in the cases of the deuteron-neutron reactions being considered for the variable energy source.

At the suggestion of Dr. J. E. Rose, a hot air jet* designed for welding plastics was obtained. This jet is capable of directing a stream of air of controllable temperature variable up to the order of 800°F against a point one-eighth inch from its nozzle. It is hoped that it will be possible to prevent the buildup of deuterium in the targets by heating them, since it is known that hydrogen can be driven from an occluder by raising its temperature sufficiently. A special arrangement has been built for a natural silicon target in which the silicon slug is mounted in intimate contact with a silver piece which closes the end of the metal tube passing through the center of a Kovar insulator.** The Kovar is to be mounted in the beam port at the end of the target chamber of the Cockroft-Walton generator. The silicon target will thus be thermally insulated from its surroundings so as to take advantage of the heating effect of the beam, and it will be possible to direct a stream of "Hot Jet" air against the silver backing piece from outside the vacuum through the Kovar tube. With a thermocouple mounted in the silver backing slug as a monitor it should be possible to control the target temperature quite closely. The entire target assembly was built to withstand temperatures up to the order of 900°F. This target is to be tested soon.

As was mentioned previously in this report, a study of the $B^{10,11}(\alpha,n)N^{13,14}$ reactions has also been started. Up to the present time only natural boron targets (19% B^{10} , 81% B^{11}) have been available, but a supply of enriched B^{10} (to 92%) is on order. With a natural boron target under bombardment with 250-kev alpha particles two energy groups of neutrons would be expected, one at about $1\frac{1}{4}$ Mev from B^{10} , and one at about 400 kev from B^{11} . The spectra actually obtained were measured with a $Li^6I(Eu)$ crystal spectrometer consisting of a $1\frac{1}{2}$ inch diameter by 2 mm thick $Li^6I(Eu)$ crystal mounted on a Dumont type 6292 photomultiplier tube. The photomultiplier was connected for positive high voltage operation in order to reduce tube noise, and its output was fed to a Canadian Marconi, Ltd. multiple discriminator-type 24-channel pulse height analyzer.

In order to obtain some idea of the energy range covered by this spectrometer, the latter was used to measure the spectra of a D-D neutron source both without and with a paraffin "thermalizer" about the crystal. The results of the two runs are shown in Fig. 2 and Fig. 4, respectively, the normal counting background with the generator off having been subtracted from the data in each case. Error bars indicating the statistical standard deviations have been attached to the points where the magnitudes were sufficient to be seen on the plot. For both runs the photomultiplier was operated at 1161 volts, and the bombardments were carried out at 225 kev with 8 μ amp of deuterons to the deuterium-zirconium target. The crystal was located at a scattering

*"Hot Jet," manufactured by D. and R. Plastic Welders, Inc., Hazardville, Conn.

**Kovar clear glass insulator type No. 95.2013, manufactured by the Stupakoff Ceramic and Manufacturing Company, Latrobe, Pa.

angle of 90° from the deuteron beam direction in each measurement with the flat top surface facing the target. For the straight D-D run of Fig. 2 the crystal was 11.2 cm from the target, and for the "thermalized" run of Fig. 4 it was about three times as far away, the apparent greater intensity in the latter case being a result of the great increase in the $\text{Li}^6(n,\alpha)\text{H}^3$ reaction cross section with decreasing energy. The thermal neutron peaks are evident in both runs between about channels 4 and 9, and the high energy edge of the D-D spectrum is seen to appear between channels 19 and 20. The failure to resolve the D-D peak is doubtless due to the poor inherent resolution of the present uncooled $\text{Li}^6\text{I}(\text{Eu})$ crystal and to the significant numbers of neutrons scattered from the walls and floor of the accelerator room which are counted.

The spectrum measured with this spectrometer for a natural boron target under 225-keV alpha particle bombardment at $18\mu\text{amp}$ to the target is shown in Fig. 3. The crystal was again located in the same position relative to the target as for the D-D neutron run of Fig. 2, that is at a scattering angle of 90° and distance of 11.2 cm. Carbon was chosen as a suitable target material for a background run to be used with these data, the Q-value of the $\text{C}^{12}(\alpha,n)\text{O}^{15}$ reaction as calculated from the masses being -8.45 MeV. An alpha bombardment run with a carbon target carried out under the same conditions as were used in the boron target run showed no counts significantly above "generator-off" background except in channel 1. The points plotted in Fig. 3 show the net natural boron target data with the carbon target data subtracted as background. The error bars, as before, indicate the statistical standard deviations of the points.

It may be seen from Fig. 3 that neutrons from one or both of the $\text{B}^{10,11}(\alpha,n)\text{N}^{13,14}$ reactions are definitely observed although the yield rate is small. The fact that the spectrum goes all the way out as far as channel 15 seems to indicate that neutrons from the higher Q-value $\text{B}^{10}(\alpha,n)\text{N}^{13}$ reaction are being seen, in spite of the fact that B^{10} is the isotope of lesser abundance. The low energy portion of the spectrum of Fig. 3 below channel 5 is probably due both to neutrons which scatter elastically in the crystal rather than undergoing the $\text{Li}^6(n,\alpha)\text{H}^3$ reaction, and to gamma rays from the $\text{B}^{10}(\alpha,p)\text{C}^{13}$ reaction⁽⁵⁾ which has a Q-value of 4.07 MeV. The 0.21-MeV and 3.73-MeV energies of the C^{13} gamma rays are not large enough to produce pulses of amplitude comparable to those produced by thermal neutrons which have initiated the $\text{Li}^6(n,\alpha)\text{H}^3$ reaction, of course. The 4.78-MeV Q-value of the latter reaction⁽⁶⁾ gives too great an advantage to the neutrons in the matter of pulse height.

Considering only the neutrons in the portion of the spectrum of Fig. 3 above the location of the thermal peak, and using the neutron detection efficiency curve for a 2-mm Li^6I crystal given by Murray and Schenck,⁽⁶⁾ an order of magnitude calculation indicates that the yield of the thick natural boron target must be at least of the order of 10 neutrons per microcoulomb of 225-keV alpha particles. It is probably actually considerably

greater than this. More complete studies of the $B^{10,11}(\alpha,n)N^{13,14}$ reactions will be carried out as soon as delivery has been secured of the enriched B^{10} target material and of a new $1\frac{1}{2}$ inch diameter by $1/4$ inch thick $Li^6I(Eu)$ crystal, which has also been ordered. This new crystal is capable of being cooled to liquid nitrogen temperature for improved resolution.

One additional neutron-producing reaction with small Q-value has been located in the literature and is being added to the previously-given list⁽¹⁾ of reactions which are to be investigated for possible use in the variable energy neutron source. This is the $V^{50}(p,n)Cr^{50}$ reaction with a Q-value of +0.4 Mev.⁽⁷⁾ Unfortunately, the natural isotopic abundance of V^{50} is only 0.24%,⁽⁸⁾ but the fact that the other naturally occurring vanadium isotope, V^{51} , has a (p,n) reaction Q-value of -1.53 Mev⁽⁷⁾ makes feasible the use of vanadium enriched in V^{50} to any reasonable degree. The current catalog⁽⁸⁾ of the Isotopes Division of Oak Ridge National Laboratory lists enriched V^{50} as "temporarily out of stock."

References

1. R. Grismore, P. F. Gustafson, B. G. Oltman and L. D. Marinelli, Radiological Physics Division Semiannual Report, July through December, 1957. ANL-5829:36-45 (1958).
2. G. J. Perlow, Rev. Sci. Instr. 27:460 (1956).
3. I. B. Berlman and L. D. Marinelli, Rev. Sci. Instr. 27:858 (1956).
4. A. O. Hanson and J. L. McKibben, Phys. Rev. 72:673 (1947).
5. F. Ajzenberg and T. Lauritsen, Revs. Mod. Phys. 27:118 (1955).
6. R. B. Murray and J. Schenck, Scintillation Response of $Li^6I(Eu)$ Crystals to Monoenergetic Fast Neutrons, CF-56-5-76, Oak Ridge National Laboratory, p. 3 (May 11, 1956).
7. J. P. Schiffer and L. L. Lee, Jr., Phys. Rev. 109:2098 (1958).
8. Catalog and Price List of Stable Isotopes, Oak Ridge National Laboratory, p. 63 (April 1957).

RADIUM TOXICITY STUDIES

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

I. Quantitative Observations on Ra^{226} in Human Bone

The quantitative autoradiographic study of the deposition of Ra^{226} in the bones of radium patients, first reported in ANL-5829, has been continued. The number of patients studied has been increased to 12, all of whom have carried their burden for 20 or more years. These patients are listed in Table 1 with their terminal body burden, age at, and route of acquisition of the radium, and age at which death occurred.

Table 1
Radium Patients

Patient	Terminal radium burden, μc	Sex	Age acquired	Method of acquisition	Age at death	Years radium carried	Comments
118	10.5	F	48	Medically: Ra water?	84	36	Low Ca diet at time of Ra acquisition
103	6.8	F	36	Medically: I.V.?	64	28	Fibrosarcoma
302	3.8	F	15	Dial painter for 2 mo.	47	32	Osteogenic sarcoma
R	3.6	F	32	Medically: I.V. 41 weekly injections	56	24	10 μc Ra^{226} per injection
RB	2.7	F	20	Dial painter for 1 yr.	53	33	Carcinoma of right mastoid
313	1.3	F	18-47	Dial painter for 28 yr.	48	30	Tipped brush with lips for first 2 years Fibrosarcoma
IJ	1.2	F	18-21	Dial painter for $3\frac{1}{2}$ yr.		31	Sample obtained age 49. Resection of upper third of femur
Q	1.2	M	33	Medically: I.V. 45 weekly injections	59	26	10 μc Ra^{226} /injection
BM	1.2	M	21-23	Medically: I.V. injections	51	28-30	Sarcoma
112	1.2	F	25	Medically: oral	48	23	Fibrosarcoma
ML	~1	F	16-17	Dial painter for 18 months		39	Amputation, age 55
I	0.8	F	28	Medically: I.V. 18 weekly injections	53	26	10 μc Ra^{226} /injection

In Table 2 are listed the measured quantities from the autoradiographs; namely, the maximum hot spot intensity observed and the average value of the diffuse radium deposition both expressed in terms of activity per unit volume. The method of measurement has been described previously.⁽¹⁾

Table 2

Patient	Terminal body burden, $\text{Ra}^{226} \mu\text{c}$	Initial activity ratio $\text{Ra}^{228}/\text{Ra}^{226}$	Body weight, kg	Uniform label, $\mu\text{c}/\text{mm}^3$	Maximum hot spot, $\mu\text{c}/\text{mm}^3$	Average diffuse, $\mu\text{c}/\text{mm}^3$	Ratio: hot spot/diffuse	Ratio: diffuse/uniform label
118	20.5	0.0003	52	40.5×10^{-7}	8.6×10^{-5}	11.0×10^{-7}	78	0.27
103	0.8	0.0004	57.5	23.7	8.0	6.4	125	0.27
302	3.8	-	57	13.4	4.1	16	26	1.29
R	3.6	0.0002	55	13.1	3.5	4.8	73	0.37
RB	2.7	-	55?	9.8	3.1	8.1	38	0.83
313	1.3	0.0013	43.6	5.9	0.59	3.2	18	0.54
IJ	1.2	0.02	60	4.0	0.99	1.0	99	0.25
Q	1.2	0.0002*	70	3.4	2.5	1.7	147	0.50
BM	1.2	-	70	3.4	1.3	0.91	143	0.27
112	1.2	0.012	59	4.1	0.3	1.5	20	0.38
ML	1	-	60	3.3	1.3	2.1	62	0.64
I	0.8	0.0002*	55?	2.9	2.7	1.2	225	0.41
Average							88	0.50

Not measured, but assumed to be identical to R. These activity ratios were measured by Wallace and Steinley.⁽⁷⁾

Two criteria of acceptability have been added to improve the results. In counting hot spots a fairly wide spread in the values obtained had been noticed. The alpha tracks are counted from a NTA* plate, which is pressed into contact with the bone section for exposure, and removed for development. The bone sections were then covered with a stripping film which is finally developed and fixed while still on the bone section. When the NTA plate and the bone section with stripping film were superimposed under the microscope, it was observed that some of the clusters of alpha tracks visible on the NTA plates covered a larger area than the corresponding clusters observed in the stripping film. This implied poor contact between the section and the NTA plate. The activity per unit volume obtained from such areas was thus too low. The intensities reported in Table 2 are from hot spots in which the area covered by the cluster in the NTA plate has been checked and found to be the same as that indicated by the stripping film.

Poor contact, as described above, is not serious in evaluating the diffuse component, for a large area is surveyed. Since the area over which the counts are made must be representative, it has been found advisable to choose the diffuse areas to be counted by examination of autoradiographs made on No-Screen film. This film gives a visible picture of the hot spots and diffuse distribution; thus by this means representative diffuse areas are easily chosen and then evaluated by track counting of the identical area on NTA emulsion.

Several significant items are found in Table 2. First, the ratio of the hot-spot to the diffuse activity ranges from a low of 18 to a high of 225,

*Type NTA nuclear track plate, Eastman Kodak Co.

with an average value of 88. In all of these cases the radium was acquired over a considerable time [from 2 months (302) to $3\frac{1}{2}$ years (IJ)]. While a single injection would produce the highest ratio, it would fail to produce a hot spot with sufficient area for an accurate alpha track count. Our criterion has been that an entire haversian system be labeled to be included in the hot-spot analysis, and we feel that a period of 8 to 12 weeks is required in the human for the closing time of a secondary haversian system.

In Table 2 we have calculated a "uniform label," the activity per unit volume of bone that would exist in each patient if all the radium retained in the body were uniformly distributed throughout all the bone. It has been previously pointed out that the diffuse distribution was approximately half of this uniform label.⁽¹⁾ For these twelve cases we find that the average ratio is 0.50, with a low of 0.25 and a high of 1.29.

This last high value is quite remarkable in that it states that the diffuse distribution as measured is greater than a hypothetical uniform distribution, an apparent impossibility implying that either or both types of measurement are incorrect. However, from Table 1 we note that this patient acquired the radium during a two month period when she was only 15 years old. If we assume most of the activity was taken up in areas of growth, there is a good possibility that an abnormally large fraction of the originally retained radium has been removed by bone turn-over. Cortical bone, where the loss of activity is very low, might thus appear to retain a relatively large fraction of the terminal burden.

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

It was pointed out in the last semiannual report (ANL-5829) that the maximum hot-spot intensity found in two Elgin State Hospital patients was a factor of 10 lower than the value predicted by the average value of the blood specific activity. We now have three such patients in which the number of weekly $10\ \mu\text{c}\ \text{Ra}^{226}$ injections is known. These are Q, R, and I; from Table 2 it is seen that their maximum hot spots are very nearly equal; 2.5, 3.5, and $2.7 \times 10^{-5}\ \mu\text{c}/\text{mm}^3$, respectively. The predicted hot-spot intensity is $36 \times 10^{-5}\ \mu\text{c}/\text{mm}^3$ for all three of these patients.

Does a corresponding loss of activity take place in the diffuse component? A hint that it does is contained in the ratio of the diffuse to the uniform label, which is the order of $1/2$ for all these cases, for this same ratio is found in the dog one month after injection of radium. Another indication of a loss from the diffuse is obtained from the augmentation rate. Marshall (ANL-5829) has defined the uptake of calcium as indicated by the diffuse labeling of bone, as observed at times the order of a month, as augmentation. The rate of augmentation is given by

$$f = \frac{a}{\int_0^{T_s} Bdt}$$

where

f = the augmentation rate (g Ca/g Ca day)

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

T_s = time of sacrifice (days after injection of isotope)

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

and

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

For dogs, rates of the order of 5×10^{-4} g Ca/g Ca day for animals a year or so of age and 1.5×10^{-4} g Ca/g Ca day for an older dog were measured.

If we apply this equation to the diffuse level found in a human almost 30 years after injection, what apparent augmentation rate is obtained? The integral of the blood curve for the three Elgin cases may be approximated by multiplying the average weekly specific activity by the number of weeks over which the injections were continued. The "rates" obtained terminally are tabulated in Table 3.

Table 3

Patient	Terminal burden, $\mu\text{c Ra}^{226}$	No. Weekly Ra^{226} 10 μc injections	Total activity injected, μc	% of Injection retained terminally	Apparent augmentation rate, $\frac{\text{g Ca}}{\text{g Ca day}}$
Q	1.2	45	450	0.27%	1.72×10^{-6}
R	3.6	41	410	0.88	5.17×10^{-6}
I	0.8	18	180	0.44	3.18×10^{-6}

These apparent rates are thus of the order of $3 \times 10^{-6} \frac{\text{g Ca}}{\text{g Ca day}}$, a factor of 100 less than the values obtained for dogs. However, this does not necessarily imply that the diffuse component activity has been reduced to only 1/100 of its original value; we have reason to suspect that the augmentation rate in humans is considerably less than the rate in dogs.

We have one measurement of an augmentation rate in a terminal human. Using Ca^{45} as a tracer, we obtained a rate of the order of 5×10^{-5} g Ca/g Ca day, measured in a 51 year old man who died 23 days after injection. While a terminal case is always doubtful, since this value is a factor of 10

larger than the values obtained for the Elgin cases, at least 90% of the original diffuse activity appears to have been lost after 26 to 28 years.

Thus we have evidence for the loss of activity both from the hot spots and the diffuse in human bones over this period by the process we have called diminution. This loss is so great that it appears, at least in cortical bone, to be a major method of removal of the isotope.

III. Terminal Dose Distributions

Spiers⁽²⁾ has developed an equation for determining the dose to cavities of various sizes in bone from alpha particles originating from uniform concentrations of an isotope around the cavity. Recently Kononenko,⁽³⁾ by a rigorous mathematical analysis, has made minor changes in the analyses. Spiers' equation has the form:

$$\text{Dose Rate} = \frac{N\sigma R_T}{2\rho} \times \bar{F} \text{ ergs}/\mu^3/\text{day}$$

where:

- N = the number of alpha particles emitted per μ^3 per day
- σ = average energy loss of the alpha particles in soft tissue
(Spiers' value, 0.21×10^{-6} ergs/ μ)
- R_T = alpha particle range in soft tissue (Spiers' value, 44μ)
- ρ = ratio of alpha particle ranges in tissue and in bone (Spiers' value, 1.36; Range in bone = 32μ).
- $\bar{F}$ = A geometrical coefficient, dependent upon the cavity size.

Values of $\bar{F}$ from Spiers and Kononenko are tabulated below (Table 4) for various cavities. The values of Kononenko are used in subsequent calculations.

Table 4

Dimensions of cavity, μ	$\bar{F}$ - Average for a cylinder	
	Spiers	Kononenko
0	2.00	2.00
1 - Canaliculi	1.89	1.97
5 - Lacunae containing osteocytes	1.60	1.84
10 - Lacunae containing osteocytes	1.35	1.70
20 - Haversian canals	1.00	1.42
50 - Haversian canals	0.55	0.82

The major problem in dosimetry of bone is the choice of the proper location to measure the dose, not the actual mechanics of how to measure it. Fortunately, in the case of alpha emitters, the problem is simplified.

Since the range is so short, one can evaluate the dose delivered to an osteocyte or to a 100-micron thick layer of periosteum or endosteum and state that these are the maximum doses that can be delivered anywhere in the bone. Since the range of the alphas in soft tissue is only 44μ , and is 30μ in bone, only a limited volume of tissue receives an appreciable alpha-particle dose. Even though very large concentrations of activity exist in trabecular bone, since the specific activities (radium per gram calcium) are no greater than those found in haversian bone, no volume of tissue can receive a dose greater than that received by the osteocytes in their lacunae, although a much larger volume of tissue will receive a high dose in such regions.

The terminal dose delivered to a lacuna 10μ in diameter is calculated and listed in Table 5 for the 12 patients. The dose is given for a lacuna in a hot spot and for one in the diffuse distribution. The calculations are based on:

- a) 30% radon retention in vivo
- b) 40% retention during autoradiography
- c) $1/2$ of equilibrium value of Ra D has grown in
- d) Spiers' equation and values of σ and ρ .
- e) Kononenko's modifications of Spiers' geometrical factors
- f) Density of bone tissue is one gram/cm³.

Table 5

Patient	Terminal body burden, $\mu\text{C Ra}^{226}$	Terminal dose rate to a 10μ cylindrical cavity (lacuna), rad/day		Total accumulated dose to a lacuna, rads	
		Hot spot	Diffuse	Hot spot	Diffuse
118	10.5	33	0.42	860,000	11,000
103	6.8	30	0.24	620,000	5,000
302	3.8	15	0.61	360,000	14,000
R	3.6	13	0.18	220,000	3,200
RB	2.7	12	0.31	280,000	7,400
313	1.3	2.2	0.12	48,000	2,600
IJ	1.2	3.8	0.038	86,000	860
Q	1.2	9.5	0.064	180,000	1,200
BM	1.2	4.9	0.034	104,000	720
112	1.2	1.1	0.057	18,000	960
ML	~1	4.9	0.080	70,000	2,200
I	0.8	10	0.045	94,000	860

Also listed is the total (lifetime) dose to a lacuna in both the regions of high and low activity. This number is obtained by multiplying the observed terminal dose rate by the number of days the radium has been carried and then doubling this result. The total increase over the terminal dose rate is unknown, but doubling seems a safe approximation. Van Dilla⁽⁴⁾ indicated that one-half of the lifetime dose delivered to bone occurs over the first three-year period from the observation of the shape of the retention curve.

The average dose to a 10μ layer of periosteum or endosteum or lining of a haversian canal will be about one-half of the value calculated for a lacuna.

Hoecker⁽⁵⁾ calculated the dose rate to bone for two radium cases, and obtained values comparable to those given here. However, the reported body burdens for his two cases were quite large. In Table 6 below we have expressed Hoecker's data in terms of concentrations and have also listed the values we obtained for bone sections from the same individuals. It is apparent that the activities determined are in satisfactory agreement.

Table 6

Case	Terminal body burden, μc $\text{Ra}^{226} + \text{Ra}^{228}$	Hoecker: maximum concentration, $\mu\text{c}/\text{mm}^3$	This report		Estimated terminal Ra^{226} burden, μc
			Maximum hot spot, $\mu\text{c} \text{Ra}^{226}/\text{mm}^3$	Diffuse, $\mu\text{c} \text{Ra}^{226}/\text{mm}^3$	
E.E	50	3.67×10^{-5}	4.1×10^{-5}	23×10^{-7}	13.8
I. L	8	2.25×10^{-5}	2.0×10^{-5}	5.9×10^{-7}	3.5

The body burdens, as measured by Martland,⁽⁶⁾ were expressed in terms of micrograms of radium, but actually consisted of mixed Ra^{226} and Ra^{228} . The alpha-ray tracks counted by Hoecker, 19 and 23 years after death of the two patients, and in this report, 26 and 31 years after, were predominantly due to Ra^{226} . On the basis of the observation that the diffuse level is approximately one half of the uniform label, the Ra^{226} burden of these two patients is estimated in Table 6. It is of interest to note that Martland's values are 3.6 and 2.2 times higher: this discrepancy could be due to the differences in physiological activity of bone during ingestion and to the difference in the $\text{Ra}^{228}/\text{Ra}^{226}$ ratio in the material ingested by the two cases.

IV. Conclusions

It is apparent from this study that extremely high dose rates to bone cells exist, higher than had previously been considered. Dose rates of the order of 30 rads per day have been measured at terminal body burdens of

6.8 to 10.5 μc , and of the order of 10 rads per day at 1 μc levels. Thus, with these burdens lifetime doses in the range from 10^5 to 10^6 rads to bone tissue are to be expected.

The loss of radium from bone by a mechanism other than direct resorption, whether it be due to diffusion, long-term exchange, etc., is surprising in its magnitude. It appears to be the major method of removal of the isotope from compact cortical bone.

The measured ratio of hot-spot to diffuse activity is an order of magnitude larger than previously considered values. Perhaps more significant, however, is the quantity of radium associated with the diffuse component. Its magnitude, approximately one-half of the uniform label, seems to doom to failure any attempt to remove radium in large amounts from the skeleton without involving the entire skeleton.

References

1. Rowland, R. E., J. Jowsey and J. H. Marshall. Radiological Physics Division Semiannual Report, ANL-5829: 109-114 (December 1957).
2. Spiers, F. W. Alpha-ray Dosage in Bone Containing Radium. Brit. J. Radiol. 26:296-301 (1953).
3. Kononenko, A. M. Calculation of the Intensity of the Alpha-Radiation Dose Arising from a Radioactive Substance Distributed Inside the Organism. Biophysics 2(1): 98-117, (1957).
4. Van Dilla, M. A., B. J. Stover, R. L. Floyd, D. R. Atherton and D. H. Taysum. Radium (Ra^{226}) and Radon (Em^{222}) Metabolism in Dogs. Radiation Research 8:417-437 (1958).
5. Hoecker, F. E. and P. G. Roofe. Studies of Radium in Human Bone. Radiology 56:89-98 (1951).
6. Martland, H. S. The Occurrence of Malignancy in Radioactive Persons. Am. J. Cancer 40:2435-2516 (1931).
7. Wallace, D. E., and A. F. Stehney. Radiological Physics Division Semiannual Report, ANL-5829:115-120 (December 1957).

THE EFFECT OF PARATHYROIDECTOMY ON HAVERSIAN REMODELING OF BONE*

Jenifer Jowsey, R. E. Rowland, J. H. Marshall and F. C. McLean**

Abstract

The occurrence of haversian remodeling has been measured in control and parathyroidectomized adult dogs given a tracer dose of calcium-45. Autoradiographs, microradiographs and histological sections were made of samples of diaphyseal and metaphyseal bone and the numbers of resorption cavities and newly formed haversian systems compared for the control and experimental animals. The results indicate that haversian remodeling is reduced in the parathyroidectomized animals. Measurements of the actively calcifying lengths of the systems show that the reduction in the remodeling incidence in the experimental animals is not the result of a shorter length undergoing calcification but of a smaller number of systems being formed. Two of the dogs used in the experiment were females that had borne and weaned a litter six weeks previously. In both these animals there is increased bone remodeling compared with that in the male dogs; however, the parathyroidectomized female shows a reduced incidence of remodeling compared with her female control.

Introduction

That there is an interrelationship between the parathyroids, osteoclastic activity, and resorption of bone is well established, even though the details of the relationship are not all known. The parathyroid glands monitor the calcium ion concentration in the blood plasma and respond to a diminution in this concentration by increased activity, leading in turn to increased resorption of bone with consequent release into the blood of dissolved calcium. These are component parts of a feedback mechanism concerned in the homeostatic regulation of the calcium ion concentration in the fluids of the body.

The resorption of bone and the subsequent rebuilding, referred to as remodeling, is of two sorts: 1) the remodeling incident to growth; and 2) the internal reconstruction of compact bone, known as haversian

*Submitted to Endocrinology

**Professor Emeritus, Department of Physiology, University of Chicago

remodeling. The remodeling of growth is especially prominent at the junctions of the epiphyses of the long bones with their shafts where, in the metaphyseal regions, a reshaping of the bones continues throughout their growth in length. While it has been shown that parathyroidectomy leads to a partial failure of the normal resorption pattern in this region, there is not a complete cessation of osteoclastic activity.

The present paper is concerned with the internal, or haversian remodeling of compact bone, which occurs in two main stages: 1) a tunneling process, occurring apparently at random in compact bone, leading to the formation of resorption cavities; and 2) the filling in of these tunnels or cavities by the laying down of concentric layers or lamellae of bone, resulting in the formation of new haversian systems. It is this remodeling in adult compact bone that plays an important part in the maintenance of a constant calcium ion concentration in the blood, and thus performs an important physiological function. It occurs throughout the life of the animal and in man even appears to increase in old age.

Little is known about the control of haversian remodeling. The tunneling operation resembles resorption in other areas of bone and is accompanied by the presence of osteoclasts; when construction of a new haversian system begins, these are replaced by osteoblasts, and new bone formation proceeds in the usual fashion. It has seemed desirable to determine whether the resorption incident to the formation of cavities in compact bone, to be later filled in by new haversian systems, is wholly or partly under the control of parathyroid activity. To this end littermate adult dogs have been thyro-parathyroidectomized, and the effects upon the internal structure and remodeling of cortical bone have been observed and compared in control animals.

Methods

A litter of four 2-year old dogs was used, three of which were males, and one of which was a female that had borne a litter of four puppies weaned six weeks before the experiment began. As a control for this female a mongrel of the same age that had weaned four puppies six weeks previously was used. Two males and the one female of the litter were thyro-parathyroidectomized, four parathyroids being identified from each animal. The serum calcium and phosphorus of the experimental animals were determined before and after operation; there was a sharp drop in the calcium level and a rise in serum phosphorus after removal of the parathyroids. Both the control and experimental animals were given 5% calcium chloride intraperitoneally and fed on a high calcium diet for 12 days after the operation. The animals were kept for two months without further treatment to permit the bone tissue to be stabilized.

The two female dogs, one experimental and one control, were given a single injection of Ca^{45} and killed a month after injection; one male experimental dog was injected once and again a month later and killed a week after the second injection; two male dogs, one experimental and one control, were injected three times with Ca^{45} every two months and killed one week after the last injection. It was hoped that a comparison between animals given one injection and those given multiple injections would give an indication of the rate of bone building. However, in this respect, no differences were observable between the animals that had received single and multiple injections.

As the dogs were killed the femurs and tibia-fibulas were removed and fixed in 100% alcohol. A piece approximately one and one-half inches long was cut from each tibia shaft, the midpoint being the point of union of the tibia with the fibula; this piece served as the sample of tibial diaphysis (Table 7). Each femur shaft was transected at the midpoint, and the half-inch distal to this was preserved as the sample of femur diaphysis. Beyond this a quarter-inch length was decalcified and prepared for histologic sections. Distal to this another half-inch block served as the sample of femur metaphysis. The samples of tibial diaphysis, femur diaphysis and femur metaphysis were embedded in methyl methacrylate and cut into cross sections 100 μ thick and 500 μ apart.

Table 7

	No. of calcifying osteons/cm ² of section			Average length of calcifying osteons, mm
	Femur diaphysis	Femur metaphysis	Tibia diaphysis	
Male experimental	11	18	14	0.66
Male experimental	11	22	26	0.71
Male control	31	52	72	0.74
Female experimental	45	67	160	0.92
Female control	78	213	366	0.88

Microradiographs and low resolution autoradiographs (Kodak Spectroscopic Plate Autoradiographic Type A) were made of the sections. From the autoradiographs the number of actively calcifying osteons in each section was counted. An actively calcifying system was taken to be demonstrated by an increase in darkening of the film by a factor of three or more times that of the diffuse reaction. An estimation of the area of each section was made and the number of calcifying osteons per square centimeter for each of the three pieces of bone from each dog obtained.

The number of calcifying osteons/cm² appearing in each section could vary because the actual number of different osteons varied or because the length of the osteons varied. To find out if the variations in the number of osteons were the result of changes in length or of the appearance of new systems, the length of the calcifying osteons was measured in the diaphyseal blocks of the femurs and tibias of the dogs. Calcifying osteons of lengths less than 500 μ were taken to be 500 μ long; this would result in a slightly higher value for the average length if many osteons are much shorter than 500 μ , but since the results are comparable rather than absolute, no error is incurred. The values for the average number and length of the actively calcifying region of osteons in the diaphysis of the femurs and tibias, and the metaphysis of the femurs of the five dogs comprise the results of the experiment. (Table 7)

The disappearance of Ca⁴⁵ from the blood of the five dogs was measured after each injection and curves drawn which proved to be identical for the experimental and control animals.

Results and Discussion

A comparison of the values given for the three pieces of bone demonstrates the differences in turnover in the bones and in different regions of the same bone. The tibia diaphysis shows a consistently higher remodeling rate than the femur diaphysis, and the metaphysis of the femur has a higher rate than the diaphysis by a factor of two or more. However, these variations between the different bones and regions of the same bone are consistent in the five dogs, and the significant variations appear between the control and experimental animals.

The results show that the two female dogs, two months after bearing and weaning four puppies, are rebuilding their bone at a tremendous rate; however, the rate of rebuilding is slowed down to a half or less of its normal value in the parathyroidectomized female compared with her control. The male dogs show the same result, namely a rate of bone formation that is a factor of two or three less in a parathyroidectomized animal when compared with the controls. (Figure 5).

The changes in the lengths of the actively calcifying systems are not so dramatic. As values in themselves they mean little, but they are comparable within the experiment and they show that the large increases in numbers of calcifying osteons in sections from the male dogs cannot be explained merely by an increase in length. Both the control and experimental female dogs show an increase in the length of the calcifying systems which must contribute somewhat to the increase in number of calcifying osteons seen in each section.

The smaller number of calcifying systems resulting from parathyroidectomy indicates that the removal of the parathyroids reduced the rate of resorption. The alternative interpretation, that there was not a reduction in resorption but rather a failure of calcification, would lead one to expect a larger number of resorption cavities in the experimental animals, and this was not observed. Both the microradiographs (Fig. 6) and the histological sections show approximately the same number of resorption cavities -- perhaps fewer -- in the sections from the parathyroidectomized animals as in the sections from the controls.

Resorption is not completely prevented, however, and it is obvious from the results that factors other than the parathyroids influence the rate of resorption in adult cortical bone. In the two female animals there is the influence of pregnancy and lactation which apparently causes increased resorption which, though reduced in the parathyroidectomized animal, is still three or four times the incidence found in the male experimental animals. This increased resorption is reflected in the consequent increased rebuilding that occurs a month after weaning. It is perhaps worthwhile to point out here that the time at which there will be maximum uptake of an isotope in an adult animal is not during pregnancy but a week or more after the litter has been weaned, when repair of the bone is taking place.

We much appreciate the help of Dr. Smith Freeman of Northwestern University, who thyro-parathyroidectomized the animals, and Dr. Robert Flynn who cared for the animals during the experiment. We thank Charles Williams, now at Illinois Medical College, for his interest and conscientious technical assistance.

p

c

a

Fig. 5. Autoradiographs of mid-diaphyseal tibial sections from the experimental and control dogs.
Magnification x 7

a) Experimental male	b) Experimental male	c) Control male
d) Experimental female	e) Control female	

Fig. 6. Microradiographs of mid-diaphyseal femoral sections from an experimental and control animal. Magnification x 23
a) Experimental male b) Control male

CALCULATION OF THE POSSIBLE EFFECTS OF CALCIUM DIFFUSION IN BONE CRYSTALLITES

J. H. Marshall and R. E. Rowland

Introduction

The uptake of Ca^{45} by mature bone without a corresponding increase in its total calcium content has indicated the existence of a long-term exchange of calcium in bone.⁽¹⁾ By some mechanism, a typical calcium atom can transfer from blood to the mineral of bone, remain in bone for months or years, and then transfer back again to the blood. Since the Ca^{45} diffuse uptake, and hence the rate of calcium transfer between blood and a given microscopic volume element of mature bone, does not decrease rapidly with age,⁽¹⁾ such an exchange of calcium atoms will eventually result in a loss of radioactive calcium atoms from labelled bone by a mechanism other than resorption. What could this mechanism be?

Recrystallization ($n = 1$)

Several mechanisms have been suggested, of which we will consider three. First, recrystallization: If whole crystallites were occasionally dissolving and other crystallites forming so as to maintain mineral mass constant, radioactive atoms incorporated into the forming crystallites would not be released until some later time when these new crystallites in turn are dissolved. If this dissolution were random as to choice of crystallites, and if the fraction of the total number of crystallites which dissolved per unit time were constant, such recrystallization, improbable though it may be, could explain the magnitude of the observed diffuse uptake of Ca^{45} and provide an explanation for the long-term retention of atoms in an exchange process.* This recrystallization model can be represented for Ca^{45} tracer experiments by a single large mineral compartment coupled to the blood and extracellular fluid as shown in Fig. 7.

Fig. 7

One-compartment model representing recrystallization, barrier penetration, or diffusion limited to crystallite surfaces.

*We use "exchange" as defined on the microscopic level:⁽¹⁾ Any process involving equal and opposite rates of transfer of calcium atoms in a single microscopic volume of bone mineral (in which no resorption is taking place).

The analogy is based on the fact that, at a constant rate of crystallite formation and with random dissolution, the rate of transfer of Ca^{45} from bone to blood would be directly proportional to the quantity of Ca^{45} in bone, i.e., to the average specific activity of bone, and the rate of transfer of Ca^{45} from blood to bone would be directly proportional to the specific activity of the blood. Thus a single rate constant and a single bone compartment will describe the process even though there is not actual mixing within the compartment.

Barrier Penetration ($n = 1$)

The second mechanism to be considered is a barrier to calcium ions between blood and bone mineral. Such a barrier might occur on the crystallite level, due to a close juxtaposition of crystallites as discussed by Neuman,⁽²⁾ or due to diffusion of calcium through organic matrix, and result in a low probability of calcium transfer between extracellular fluid and crystallite. If the penetration probability of such a barrier remained relatively constant in time and were the rate-limiting step in the long-term transfer of calcium between blood and bone mineral, then this model, too, could be represented by a single large mineral compartment coupled to blood and extracellular fluid by a single rate constant. Both of these models, recrystallization and barrier penetration, can be considered mathematically as a special case of the following ($n = 1$).

Diffusion in Crystallites ($n = 5-10$)

The third mechanism we wish to consider is a diffusion of calcium within bone crystallites. Such diffusion might occur by way of lattice defects in bone mineral or through interstices between regular lattice points. Because the crystallites in bone are extremely small and the time constant for long-term exchange of calcium is long, the diffusion coefficient required by the model ($\sim 10^{-23} \text{ cm}^2/\text{sec}$) is not unreasonable for diffusion of atoms in solids. Such a model for calcium diffusion in bone crystallites is developed below.

A model for diffusion of calcium into bone mineral seems to present an immediate difficulty. If Ca^{45} is presented to a crystal surface by means of a single intravenous injection, one may picture Ca^{45} atoms beginning to diffuse into the crystal. Such diffusion would be a slow process, however, and the specific activity of the blood and extracellular fluid would quickly drop below the specific activity of the infinitesimal layer of crystal into which the Ca^{45} had diffused. Therefore, a large fraction of the Ca^{45} would diffuse back out again, because a net flow of Ca^{45} from crystal to fluid would start as soon as the specific activity of the fluid dropped below that of the surface layer of crystal. But is this surface layer infinitesimal? This is the crux of the problem. Electron micrographs show that bone

crystallites are so small, 30-50 Å in diameter, that a calcium ion (diameter 2 Å) diffusing from the surface to the center of a crystallite would have to make only the order of 5 to 10 discrete radial jumps between relatively stable locations within the lattice. The first jump for calcium below the surface of a crystallite would thus be far from infinitesimal, because the volume of this first below-surface location accounts for 20-30% of the total calcium content of the crystal. Therefore, diffusion of calcium within a crystallite cannot be handled directly by the continuous diffusion equation. Instead, we consider discrete jumps between a relatively small number of lattice locations, letting the probability of a jump in any direction be determined by a single diffusion coefficient. As shown below, the resulting time dependence of the Ca^{45} content of such a crystallite, either formed before or after Ca^{45} injection, is not inconsistent with present data on diffuse or hotspot specific activity, respectively.

Theory

Consider a particular microscopic volume of bone containing C grams of calcium in the form of cylindrical crystallites, and assume that the uptake and retention of Ca^{45} in the volume as a whole can be described by finding the behavior of Ca^{45} in a typical crystallite. Let this crystallite be long compared to its diameter and let its diameter be such that there are n discrete radial levels for calcium between its surface and its axis. Let the innermost level contain C_1 grams of calcium, the next C_2 and so on to the outermost level C_n , as shown in Fig. 8 for $n = 6$.

Fig. 8. Six-compartment model representing diffusion within a crystallite about 36 Å in diameter.

We determine the n values of C_i by letting their sum equal the total calcium content of the microscopic volume C and by letting C_i be proportional to the area between successive equally-spaced concentric circles:

$$C_i = \left(\frac{2i - 1}{n^2} \right) C \quad (1)$$

It follows from (1) that

$$\sum_{i=1}^n C_i = C \quad (2)$$

A transfer of calcium ions between the different levels (with no net flow) can then be described by a set of transfer rates M_i (g Ca/day). The n values of M_i must similarly vary with the radius because we may assume that the transfer of calcium ions across unit area in any direction is constant, whereas the surface areas of the cylinders separating the levels vary directly with radius:

$$M_i = \frac{M_n (2i - 1)}{2n - 1} \quad (3)$$

The quantity derivable from the diffuse uptake of Ca^{45} observable about one month after injection on a Ca^{45} autoradiogram⁽¹⁾ is the rate of long-term exchange

$$f = \frac{M_n}{C} \quad (4)$$

which was found for young adult dogs to be about 5×10^{-4} gCa/gCa day. Substituting (4) into (3):

$$M_i = \frac{fC (2i - 1)}{2n - 1} \quad (5)$$

Now if we let v_i be the specific activity ($m\text{Ca}^{45}/\text{gCa}$) of the i^{th} level in the crystallite, the gross rate of Ca^{45} transfer ($m\text{Ca}^{45}/\text{day}$) from level i to level $i + 1$ will be $M_i v_i$. The rate of change of the Ca^{45} content of level i is:

$$C_i \frac{d v_i}{dt} = M_i (v_{i+1} - v_i) + M_{i-1} (v_{i-1} - v_i) \quad (6)$$

Dividing through by C_i and substituting the values for C_i , M_i , and M_{i-1} from (1) and (5):

$$\frac{d v_i}{dt} = a(v_{i+1} - v_i) + b_i (v_{i-1} - v_i) \quad (7)$$

where

$$a = f \left(\frac{n^2}{2n-1} \right) \text{ and } b_i = a \left(\frac{2i-3}{2i-1} \right)$$

Diffusion Coefficient

If Δr is the radial separation between levels, the average radius of the i^{th} level is $(i - 1/2) \Delta r$. Using this expression, one can show that in the limit of large n , expression (7) reduces to the diffusion equation for cylindrical symmetry:

$$\frac{\partial v}{\partial t} = \left(\frac{\partial^2 v}{\partial r^2} + \frac{1}{r} \frac{\partial v}{\partial r} \right) \quad (8)$$

where $\frac{D}{(\Delta r)^2}$ corresponds to a above.

$$\text{Thus} \quad D = f \left(\frac{n^2 (\Delta r)^2}{2n-1} \right) \quad (9)$$

If we take the following reasonable values for the parameters in (9). for the magnitude of diffuse uptake of Ca^{45} , $f = 5 \times 10^{-4} \text{ gCa/gCa day}$,⁽¹⁾ for the single jump distance for calcium in the apatite lattice, $\Delta r = 3 \times 10^{-8} \text{ cm}$, and for the radius of bone crystallites, $n \Delta r = 18 \times 10^{-8} \text{ cm}$ ($n = 6$), we find $D = 1.7 \times 10^{-23} \text{ cm}^2/\text{sec}$.

Boundary Conditions

The set of n linear differential equations given by expression (7) has been solved on the IBM 650 digital computer under the following boundary conditions. There is no $i = 0$ level in our crystallite so $b_1 = 0$. The specific activity of the fluid at the surface of the crystallite, v_{n+1} , we assume to be equal to measured values of blood Ca^{45} specific activity as a function of time $B(t)$. When the short-term exchangeable compartments in bone have been better established, the model may be made more accurate (particularly at less than a day after injection) by inclusion of several compartments between the blood and the crystal surface. However, such compartments cannot contain much calcium in comparison to the crystallites so that their Ca^{45} content should soon reach a close transient equilibrium with the blood; thus their effect at times of a few weeks or longer should not be large. Data for the Ca^{45} specific activity in the blood of adult dogs for 100 days following a single injection⁽³⁾ are well represented by

$$B(t) = 1\,293 (t + 0.25 \text{ day})^{-1.15} \quad (10)$$

where t is measured from the time of injection in days.* The normalization constant outside the bracket has been chosen to make $B(1 \text{ day}) = 1.000$. (See Fig. 9).

Fig. 9. Specific activity of each layer and the average specific activity of a six-compartment crystallite under the two initial conditions. $B(t)$ represents the values of blood specific activity included in the calculation as a boundary condition.

Initial Conditions

Two different sets of initial conditions have been taken for the specific activities v_i of the different levels in the crystallite. The first set, representing DIFFUSE uptake, takes the n values of v_i to be zero at $t = 0$ in order to represent the behavior of a crystallite present at the time of injection. The second set of initial conditions, representing a crystallite in a HOTSPOT, takes the initial time as $t = 1$ day with all $v_i = B(1) = 1.000$, as if the crystallite were formed from blood calcium one day after injection. Because B drops rapidly and becomes negligible before v_n decreases

*We are indebted to S. A. Tyler of the Biological and Medical Research Division for pointing out the utility of the modified power function for describing blood specific activity data.

appreciably, the time at which a crystallite forms relative to injection determines only its initial specific activity and has no significant effect upon the relative retention of its Ca^{45} as a function of time.

Results

The average specific activity of a crystallite, $\bar{v}$, is the weighted average of the values v_i for the different levels:

$$\bar{v} = \frac{\sum_{i=1}^n C_i v_i}{\sum_{i=1}^n C_i} \quad (11)$$

or using (1) and (2).

$$\bar{v} = \sum_{i=1}^n \frac{2i-1}{n^2} v_i \quad (12)$$

Figure 9 shows typical results for $\bar{v}$ as a function of time together with the component values of v_i for the two different initial conditions. For a given model ($n = 1$ or $n = 5 - 10$), the theory has only one adjustable parameter, the rate of long-term exchange f . Figures 10 and 11 show results for $\bar{v}$ for different choices of the parameters. Figures 12-17 show interpolations of the same results as a function of f for three different times after injection.

Diffuse Component

For a crystallite existing at the time of injection, $\bar{v}$ increases in direct proportion to the time integral of B for many days after injection, just as though the activity were being incorporated into the bone by a process of accretion rather than of exchange; $\bar{v}$ reaches its maximum value when the specific activity of the surface level $\bar{v}_n$ crosses B . Thereafter $\bar{v}$ decreases approximately as $t^{-0.5}$ until the innermost level v_1 reaches its maximum. Then $\bar{v}$ decreases more rapidly and becomes exponential, eventually falling into transient equilibrium with B . (Transient equilibrium would eventually occur no matter what behavior B finally assumes.) If larger values of n are considered, such as $n = 30$, $\bar{v}$ begins falling earlier and maintains the approximate $t^{-0.5}$ behavior longer; thus for large n , the model approaches the behavior ordinarily expected from diffusion theory.

Fig. 10. Average specific activity $\bar{v}$ for an existing crystallite for different values of n and f .

Fig. 12

Fig. 11. Average specific activity $\bar{v}$ for a crystallite formed at 1 day after injection.

Fig. 13

Specific activity of crystallites in a hotspot or in the diffuse component relative to the value at 1 month.

Fig. 14

Fig. 16

Fig. 15

Fig. 17

Specific activity of crystallites in a hotspot or in the diffuse component relative to the value at 1 month

Hotspots

There is no comparable $t^{-0.5}$ behavior of $\bar{v}$ for new crystallites. Also unlike that for diffuse activity, the time dependence of $\bar{v}$ for the crystallites in a hotspot is independent of the detailed behavior of $B(t)$ for all times of practical interest. For large n the specific activity ratio of hotspots to diffuse activity would increase with time due to the more rapid loss of diffuse activity. See Fig. 18. For $n = 1$, the ratio decreases with time. For $n = 6$, the hotspot to diffuse ratio remains approximately constant (within 30-40%) between a week and 30 years after injection.

Fig. 18

Ratio of hotspot specific activity to diffuse specific activity relative to the value of the ratio at 1 month.

Effect of Less Than 100% Availability ($\alpha < 1$)

The tacit assumption has been made (expression 4) that all crystallites in the observed microscopic volume take part in the ion transfer. If this were not the case, if only a fraction α took part and the rest of the crystallites ($1 - \alpha$) were not available to the extracellular fluid, then the present calculations could be modified as shown below:

Observable quantity	Calculated quantity
$f^1 = \frac{\text{Diffuse specific activity at time } t}{\int_0^t B \, dt}$	αf
Diffuse specific activity	$\alpha \bar{v}$
Hotspot specific activity	$\alpha \bar{v} + (1 - \alpha) \bar{v}_{\text{initial}}$

Conclusions

Diffusion in bone crystallites provides a possible explanation of both the magnitude of the diffuse uptake and the long retention of calcium-like radioactivity in bone without recourse to a rapidly-decreasing availability of maturing bone as postulated by Neuman.⁽²⁾ Diffusion in crystallites larger than 100 Å in diameter ($n > 15$) would not describe both the observed diffuse uptake and the observed diffuse retention; however, diffusion in crystallites 30-50 Å in diameter ($n \sim 6$) fits the data quite well. In fact, one set of parameters, $f = 5 \times 10^{-4}$ /day, $\alpha = 1$, and $n = 6$, yields results consistent with the following observations:

- 1) the magnitude of diffuse uptake in young adult dogs,
- 2) the complete retention of diffuse activity over periods of a year or less,
- 3) the approximate constancy of the hotspot to diffuse ratio at different times,
- 4) a tenfold diminution in the specific activity of individual radium hotspots over a 30-year period.

These parameters would indicate 100% availability of crystallites in bone and a diffusion coefficient for calcium-like atoms in the solid phase of about 10^{-23} cm²/sec. Without additional data, however, it is not possible to distinguish definitely between diffusion and one of the mechanisms approximating $n = 1$ because in most tracer experiments (with times of observation of less than a year) most of the diffuse activity would still be in the surface compartment whether or not deeper penetration by diffusion were possible. (See Fig. 9).

The theory indicates that the long-term decrease in the specific activity of a hotspot should be approximately exponential no matter what mechanism is assumed ($n = 1$ or $n = 6$). (See Fig. 19.) An individual radium hotspot which had decreased in specific activity a factor of ten in 30 years would then have produced 3-4 times the accumulated dose one would calculate assuming no long-term exchange.

The major limitation of the present theory is that it considers the transfer rates to be constant with time, whereas a slow decrease in rate with age is to be expected. The effect of varying parameters can be estimated from Figs. 6-11. Another limitation is inherent in the assumption that the diffusion coefficient for the surface layer is equal to that for the deeper crystallite layers. However, if no diffusion took place between the first below-surface layer and the deeper layers, the solution for $n = 1$ with $\alpha = 0.2$ or 0.3 would apply (assuming for this purpose that all crystal

surfaces were available but crystal interiors were not). Finally, the effect upon these results of the short-term exchangeable components, diffusion in extracellular fluid and diffusion through the hydration shell, remains to be assessed.

Fig. 19

The time dependence of a tenfold loss of hotspot specific activity in 30 years for different values of n .

We wish to thank Merlin H. Dipert and the staff of the IBM 650 computer, Accounting Department, for programming and aid in the solution of this problem.

References

1. J. H. Marshall, R. E. Rowland, and J. Jowsey, Microscopic Metabolism of Calcium in Bone, Radiation Research (in press); and Radiological Physics Division Semiannual Report ANL-5829, pp. 79-108 (December 1957).
2. W. F. Neuman and M. W. Neuman, The Chemical Dynamics of Bone Mineral, University of Chicago Press (1958), p. 110.
3. J. Cohen, C. J. Maletskos, J. H. Marshall, and J. B. Williams, Radioactive Calcium Tracer Studies in Bone Grafts, J. Bone and Joint Surgery 39A, pp. 561-577 (1957).

POSITIVE ION EMISSION

H. A. Schultz

Introduction

Use of a tracer gas in the study of gaseous diffusion requires a detector which has a high reproducible sensitivity to the tracer, yet has a very low sensitivity to variations in the concentration of other gases involved in the experiment. The type of positive ion emitter described in previous reports has been found to be particularly well suited for atmospheric diffusion studies. Under selected operating conditions, the device is stable and is extremely sensitive to Freon-12 in trace quantities. Water vapor interferes somewhat with the measurement, but this interference can be eliminated by use of an appropriate cold trap to reduce the humidity to a low level. After the water has been removed, all samples of "background" air (without a tracer) give very nearly, but not exactly, the same response.

It is important to study the effect of various gases upon the ion current, in order to understand the operation of the device and to be able to use it most effectively. In making such a study, the general plan has been to use a chemically inert gas as a "control," then to substitute other gases, or to add definite concentrations to the control, singly, and in combinations. Combinations are especially important, as is evident from experiments with water vapor and hydrogen in air.

The gas surrounding the electrodes provides a medium through which the particles of ionizable material and the ions must move. In addition, the gas may enter into reactions which affect the ion current. It is, therefore, convenient to consider the effects of gases upon the ion current under two general headings; those due to the "transport properties" of the gases, and those due to the "reactive properties." The transport properties are the viscosity, thermal conductivity, and diffusion coefficients. These properties depend upon the mass of the particles, their internal degrees of freedom, and the details of their external force fields. The term "reactive properties" is here used to cover the ordinary chemical properties of the gases and the reactions between the gases and surfaces, including physical adsorption as well as chemisorption.

The transport properties are responsible for large ion current differences when a very light gas is compared with a considerably heavier gas. A comparison between the two inert gases, helium (molecular weight 4) and argon (molecular weight 40), was given in an earlier report.⁽¹⁾ However, many of the gases of interest, such as argon (molecular weight 40), air (avg 29), carbon dioxide (44), oxygen (32), and nitrogen (28) do not differ widely in transport properties; in comparing these the reactive properties may be more important than the transport properties.

Experimental Studies of Various Gases (Continued): Oxygen

Attempts to determine the effect, upon ion current, of adding various gases to argon have sometimes lead to nonreproducible results, indicating that all the pertinent variables have not been adequately controlled.⁽²⁾

What might the uncontrolled variables be? Careful control has been maintained over such physical factors as the collecting potential, the geometrical arrangement, and the resistance of the filament and of the "boat" (source of ionizable material). The results suggested that the nonreproducibility depended upon the reactive properties of the gases since the inert gas helium and the relatively stable gas nitrogen produced changes that were essentially immediate, reversible, and reproducible, while oxygen, carbon dioxide, and water vapor produced ion current effects that were sometimes inconsistent.

Oxygen was singled out as the first gas for further study since it is one of the major constituents of the atmosphere, since it combines readily with the alkali metals and since it is known to remain firmly attached to heated surfaces and to affect surface ionization strongly. It was natural to suspect that the variability of the oxygen effect might be due to trace impurities in the reaction vessel. Such impurities might enter from the filling system or the vacuum system, might leak or diffuse in from the atmosphere, or might be produced or released inside the vessel. Possible external sources have been attacked first. The filling system has been modified (several times), the vacuum system has been cleaned, and new methods have been devised for sealing the reaction vessel. A helium leak detector has been used in judging the effectiveness of the seals.

The present purification system is made entirely of glass, metal, and quartz, except that stopcocks and ball joints are sealed with low-vapor pressure silicone grease (Dow Corning High Vacuum Grease). The components, in order, are first, a dry-ice trap (for water vapor and condensable hydrocarbons), hot copper (for oxygen), hot copper oxide (for CO and H₂), Ascarite (a sodium hydroxide preparation, for CO₂), dry ice followed by magnesium perchlorate (for water passing the first dry-ice trap or formed in the copper oxide) and finally, hot magnesium in stainless steel (to take up nitrogen and residues of other gases). Beyond the last component there is a side arm used for introducing measured amounts of trace gases.

One outstanding, nonreproducible effect, consisting of a high ion current maximum which sometimes occurred during overnight runs,⁽²⁾ has not been observed with this system, nor with its predecessor, similar to it. No attempt has yet been made to produce the effect at will in order to determine the exact cause.

The only recent change in the exhaust system has been to clean the glassware. Two liquid-nitrogen traps in series have been in use for some time, and rigid start-up and shut-down schedules have been maintained, to minimize back diffusion of materials, such as water or hydrocarbons, from the pump.

The reaction vessel formerly had two wax seals, one between the bell jar and the metal base, the other in the stem through which the bell is filled. Both were designed so that only small areas of wax were in contact with the gas in the bell, and low vapor pressure Apiezon W* was used at both places. The stem seal has been replaced with a cold setting resin, Bonding Agent R-313.** Apiezon W is still used to seal the bell to the base, but the sealing method has been changed. The entire metal base was heated by a specially designed electric heater wrapped around the outer edges. This made it possible to produce a more reliable seal between the wax and the metal as judged by use of a helium leak detector.

With the external sources of impurity thus reduced, the effect of traces of oxygen in argon was re-examined. A series of tests with purified argon was followed by a series with argon plus 0.07% of added oxygen. The arrangement of electrodes was shown in an earlier report ⁽³⁾ Boat temperature, filament temperature, and collecting potential were varied, primarily within the region in which the effects of filament temperature and collecting potential are small. This is the region of potential usefulness. The added oxygen had very little effect upon the ion current, as will be discussed below, but it did affect the power requirement of the boat. With no oxygen added, the rate of heat dissipation of the boat maintained at constant temperature increased slowly with time. Oxygen decreased the rate and kept it constant. This resembles the effect of hydrocarbons, except that it was the filament power, not the boat power, that increased in the tests involving butane and other hydrocarbons. A tentative explanation of the increase in heat dissipation of the boat is that it is due to the products of thermal decomposition of vapor from the Apiezon W, or from the low vapor pressure fluorocarbon wax used to protect the metal base from chlorine. Possibly previous test arrangements allowed enough oxygen to leak or diffuse into the bell to prevent this effect. The effect of base temperature should be re-examined.

Two attempts have been made to increase the purity of the gas after it is inside the bell. An older one with silica gel seemed to have some promise. The most recent, with a heated zirconium ribbon, was unsuccessful, possibly because the electrodes and the enclosure were heated at the same time.

*Manufactured by Metropolitan-Vickers Chemical Co., Ltd., for Apiezon Products, Ltd., and sold in U.S.A. by James G. Biddle Co., 1316 Arch St., Philadelphia 7, Pennsylvania.

**Carl H. Biggs Co., Inc., 2255 Barry Ave., Los Angeles 64, California.

The study of the effect of oxygen, boat temperature, filament temperature, and collecting potential mentioned above is summarized in three graphs, Figs. 20, 21 and 22. The study was not quite finished when one of the 0.001 inch platinum control leads broke loose from the boat.

The relation between ion current and boat temperature at constant filament temperature and collecting potential is shown in Fig. 20. The familiar functional relation of the type $I = A e^{-b/T_B}$ applies, except at the very low temperature point "C," where the boat temperature is so low that the major part of the ion current is due to other sources of ionizable material, for instance, the relatively cool lower lead of the filament. No evidence was found for a "boat transition" in the region of 400° C to 600° C, as was reported previously.⁽²⁾ It is now thought that the transition was spurious. Visual examination of that particular boat indicated that its thermal expansion may have distorted it to produce a partial short circuit between the boat and one of its control leads. The slopes of the lines of Fig. 20 correspond to activation energies of 1.68, 1.93, and 1.97 electron volts. The differences between these values are small enough to be due to the exchange of heat between the filament and the boat. The control circuits maintain the filament and the boat at constant total resistance (therefore at constant average temperature), but the temperature distribution depends slightly upon the temperature of the other heated element.

The ion current depended somewhat upon the filament temperature throughout the entire region studied, including filament temperatures from 770° to 1230° C. This differs from the results reported previously for dry air, which showed a region of independence of filament temperature.⁽³⁾ It is not safe to compare these two sets of results directly since a number of changes was made between the two sets of tests, including a change in boat design to improve the degree of control over boat temperature.⁽¹⁾ The activation energy of the filament temperature effect was only 0.33 to 0.48 electron volts, comparable to the value found for one of the early units with a "nichrome" filament.⁽⁴⁾

The ion current varied with the collecting potential in the familiar pattern.⁽³⁾ Four curves are plotted in Fig. 22 representing four types of variation, three of them at one filament temperature, two at one boat temperature, all with argon plus 0.07% oxygen. Curve "B" shows the flat plateau typical of the central region in Figs 20 and 21. The presence of oxygen had no effect on this curve. Curve "A" shows a slight maximum, possibly due to field distortion near the lower guard ring due to direct emission from the heated boat. Curves "C" are typical of the very low boat temperature region in which auxiliary sources of ionizable material, such as the lower filament lead, are more effective than the boat. Curve "D" represents the low filament temperature, high boat temperature region.

Figure 20

Effect of boat temperature upon positive ion current at five different filament temperatures at a collecting potential of 300 volts in argon, with and without oxygen added. The background ion current (boat off) depended upon the recent history of operation and varied from 0.046 on the above scale after determining point "A," down to 0.01 and below at boat temperature of 464°C and 438°C and up to 0.030 after point "C."

Fig. 21 Effect of filament temperature upon positive ion current, under the conditions of Fig. 20.

Fig 22 Typical curves showing the effect of collecting potential upon positive ion current in argon plus 0.07% oxygen. Reference marks A, B, C and D correspond to points similarly designated in Figs. 20 and 21.

The lack of any definite oxygen effect upon ion current in the above experiments may be an indication that a certain amount of oxygen was present in all cases. Possibly the filament was supplied with oxygen obtained by the decomposition of K_2CO_3 ($K_2CO_3 \rightarrow 2 K + 1/2 O_2 + CO_2$). On the other hand, some of the oxygen so released would certainly be taken up by the potassium again, either on the collectors or elsewhere in the enclosure.

The results obtained with the tighter and cleaner gas handling system have shown very promising consistency. The boat which failed mechanically has been replaced with one of similar design but of more rugged construction, to be used in continuing the study of gases. Besides oxygen and chlorine, one of the most important gases to study is hydrogen since 3% hydrogen was found to have a stabilizing effect on the modified commercial unit used for field tests.⁽⁵⁾

References

1. H. A. Schultz, Radiological Physics Division Semiannual Report, ANL-5755, pp. 22-28 (1957).
2. H. A. Schultz, Radiological Physics Division Semiannual Report, ANL-5829, pp. 122-124 (1958).
3. H. A. Schultz, Report on Biological Medical and Biophysics Program, ANL-5518, pp. 35-44 (1956).
4. H. A. Schultz and J. G. Dodd, Jr., Quarterly Report, Division of Biological and Medical Research, ANL-4948, pp. 109-129 (1953).
5. H. A. Schultz, Anal. Chem. 29, pp. 1840-1842 (1957).

DETERMINATION OF Sr^{90} IN BIOLOGICAL MATERIALS

D. P. Krause

Peppard, et al.⁽¹⁾ have shown that tracer level Y^{90} may be quantitatively separated from its parent Sr^{90} by extraction from dilute mineral acid solution with the di(2-ethylhexyl) isomer (HDEHP) of dioctylorthophosphoric acid. Separation factors greater than 10^7 were obtained. This technique is being applied in this laboratory to biological materials, such as bone ash, using HDEHP obtained as a mixture of the mono- and diisomers from the Virginia-Carolina Chemical Corporation and purified by the procedure described by Peppard, et al.⁽²⁾

Tests showed that yttrium carrier was quantitatively extracted from 0.5 N HCl solution by 50% HDEHP in toluene. Further tests, using reagent grade $\text{Ca}_3(\text{PO}_4)_2$ as a "substitute" for bone ash, gave the same results. Samples of $\text{Ca}_3(\text{PO}_4)_2$ ranging in concentration from zero to 0.2 g per ml to which Sr^{90} - Y^{90} tracer had been added gave recoveries of 95-100% based on the weight of added carrier. Decay curves taken on the separated Y^{90} yielded a sixty-four hour half-life from 2000 cpm to 1 cpm after subtraction of a background counting rate of 1.1 cpm for the counter and 0.2 cpm for the sample blank.

The analytical procedure for Sr^{90} in bone ash based on the solvent extraction of its daughter Y^{90} avoids the somewhat cumbersome separation of Sr from large amounts of Ca and the necessity of correcting for recovery of added Sr carrier. One simply adds yttrium carrier and extracts directly from the sample solution. The behavior of possible interfering activities in this procedure is being studied. These are of two types: (1) long-lived activities which are not extracted but have shorter-lived extractable daughters, e.g., Ba^{140} - La^{140} and Ra^{228} - Ac^{228} , and (2) activities which may be only partially extracted, e.g., Th^{228} .

References

1. D. F. Peppard, G. W. Mason and S. W. Moline, J. Inorg. Nucl. Chem. 5:141-146 (1957).
2. D. F. Peppard, G. W. Mason, J. L. Maier, and W. J. Driscoll, J. Inorg. Nucl. Chem. 4:334-343 (1957).

DETERMINATION OF Ac^{228}

D. P. Krause and A. F. Stehney

The determination of Ac^{228} has been investigated as a method for the determination of its parent Ra^{226} in water samples and possibly in bone specimens. The Ra^{226} content of waters throughout Illinois has been measured by Lucas and Ilcewicz.⁽¹⁾ A determination of Ra^{228} in these waters is desirable in order to have a complete picture of the total radium content.

Actinium may be extracted into dibutyl phosphate (DBP) from a solution 0.1 N in HNO_3 ; under these conditions Th, Pb, Bi, and Po are also either completely or partially extracted. However, actinium may be separated from thorium by stripping the organic phase with 4 N HNO_3 . The actinium partitions to the aqueous phase while the Th remains in the organic phase. A PbS precipitation removes Pb, Bi, and Po. The supernatant liquid from the PbS precipitation then contains the purified Ac. Tests with an aged solution of Ra^{228} and its descendants showed that Ac^{228} was adequately separated from the other activities in this procedure.

References

1. H. F. Lucas and F. H. Ilcewicz, J. Am. Water Works Assoc. In press.

ON THE STRATOSPHERIC Sr^{90} FALLOUT

P. K. Kuroda

The fraction (α) of the stratospheric fallout in the total fallout (F) of a fission product (A) can be given by the following equation:

$$\alpha = \frac{y_B/y_A - T \cdot e^{(\lambda_B - \lambda_A)t}}{y_B/y_A - S'_0} \quad (1)$$

where y_B/y_A is the activity ratio of the fission products B and A freshly produced by a nuclear explosion at time $t = 0$, λ_A and λ_B are the decay constants of A and B, S'_0 is the B/A ratio in the stratosphere at $t = 0$ (prior to the nuclear explosion), T is the observed ratio of B/A in the troposphere or in rain water collected at a place far away from the test site at time t.

Equation (3) derived in the previous report⁽¹⁾ is a special case of the above equation.

T and S'_0 can be experimentally obtained, and hence α can be calculated from equation (1) if y_B/y_A is known.*

Assuming that U^{235} was used in all the nuclear explosions, the values of $F \cdot \alpha$ and $F(1 - \alpha)$ for Sr^{90} were calculated from the Ba^{140} and Sr^{90} contents of a complete series of rainfalls collected at Lemont, Illinois, during the period between October 15, 1957 and June 1, 1958, and the results are shown in Table 8. The change of the $\text{Ba}^{140}/\text{Sr}^{90}$ ratio in rain water is plotted in Fig. 23. The average rate of stratospheric Sr^{90} fallout observed at Lemont, Illinois, during this period was found to be $2.4 \text{ mc/km}^2/\text{year}$ and that of tropospheric Sr^{90} fallout $0.16 \text{ mc/km}^2/\text{year}$, i.e., 94 percent of the total Sr^{90} fallout was stratospheric.

The rate of stratospheric fallout decreased in the fall and winter and increased in the spring. These results seem to agree with the recent findings of Stewart *et al.*⁽²⁾ in England. This work is discussed in greater detail in a report now in press.⁽³⁾

*Radiochemical techniques used in this study are directly applicable to the problem of the detection of nuclear test explosions.

Table 8

Stratospheric and tropospheric Sr^{90} fallout at Lemont, Ill.
(October 15, 1957 through June 1, 1958)

Date of rainfall	Total Sr^{90} fallout, F , ($\mu\text{C}/\text{km}^2$)	Fraction in stratospheric fallout α	Stratospheric fallout, $F \cdot \alpha$, ($\mu\text{C}/\text{km}^2$)	Tropospheric fallout, $F \cdot (1 - \alpha)$ ($\mu\text{C}/\text{km}^2$)	Monthly rainfall R (mm)
10-15-57	3.7	0.65	2.4	1.3	74
10-15-57	9.6	0.28	2.7	6.9	
10-16-57	10.2	0.72	7.3	2.9	
10-22-57	16.9	0.97	16.4	0.5	
10-23-57	117	1.00	117	0.0	
11- 2-57	18.5	0.97	18.0	0.5	81
11- 7-57	8.0	0.88	7.8	0.2	
11-13-57	25.2	0.93	23.4	1.8	
11-18-57	18.4	0.99	18.2	0.2	
11-19-57	3.4	0.89	3.0	0.4	
11-30-57	1.1	0.93	1.0	0.1	
12-18-57	28.6	0.94	26.9	1.7	63
12-19-57	21.6	0.92	19.9	1.7	
12-25-57	15.0	0.97	14.5	0.5	
12-28-57	68.6	0.96	66.0	2.7	
12-31-57	35.0	0.97	34.0	1.0	
1-21-58	77.7	0.97	75.3	2.4	50
1-26-58	41.6	0.98	40.8	0.8	
2- 5-58	14.5	0.99	14.4	0.1	15
2-14-58	2.4	0.99	2.4	0.0	
2-14-58	2.4	0.99	2.4	0.0	
2-28-58	67.1	0.99	66.3	0.8	
3- 1-58	14.9	1.00	14.9	0.0	8
3- 5-58	38.1	0.99	37.7	0.4	
3-21-58	6.2	0.98	6.1	0.1	
3-28-58	1.8	0.79	1.4	0.4	
4- 4-58	36.9	0.76	28.0	8.9	36
4- 6-58	151	0.96	145	6	
4-22-58	126	0.97	122	4	
4-24-58	42.2	0.96	40.5	1.7	
4-28-58	25.9	0.88	22.8	3.1	
5- 3-58	134	0.96	129	5	31
5-14-58	97.4	0.96	93.5	3.9	
5-22-58	103	0.95	98	5	
5-30 to 6- 1-58	330	0.88	290	40	
Total	1714.0	----	1609.0	105.0	

FIGURE 23 VARIATION OF THE Ba^{140}/Sr^{90} RATIO IN RAIN WATER

Lemont, Illinois
October 15 through June 1
1957-1958

References

1. P. K. Kuroda, ANL-5829, Argonne National Laboratory, Radiological Physics Division, Semiannual Report, July through December 1957, page 167.
2. N. G. Stewart, R. G. D. Osmond, R. N. Crooks and E. M. Fisher, AERE, HP/R 2354, Atomic Energy Research Establishment, Harwell, Berkshire, 1957 (Unclassified).
3. P. K. Kuroda, ANL-5920, On the Stratospheric Sr^{90} Fallout, Argonne National Laboratory, October, 1958.

ILLINOIS MUNICIPAL WATER SUPPLIES HAVING UNUSUALLY HIGH CONCENTRATION OF Ra^{226}

H. F. Lucas, Jr.

From previous measurements of the concentrations of Ra^{226} in municipal water supplies in Illinois, (1,2) it appeared probable that unusually high levels would be found in the area lying between Moline, Ottawa, Macomb, and Bloomington (Fig. 24). Detailed sampling of private and municipal sources in this area has confirmed this expectation. The results reported in Tables 9 and 10, for limestone and sandstone wells, respectively, show the highest concentrations of Ra^{226} found in Illinois water sources. The highest value of $37 \mu\mu g Ra^{226}/l$ found in water from the St. Peters sandstone formation at Toulon is noteworthy in comparison with the maximum permissible concentration of $40 \mu\mu g/l$ recommended for Ra^{226} in drinking water. (3)

Fig. 24

Table 9

Concentration of Ra^{226} in limestone well water

Location			Ra^{226} $\mu\text{g}/\text{liter}^a$
Fig. 24	Municipal and private	Well No.	
D-4	Atkinson	1	0.81
D-3	Coal Valley	1	1.84 ^b
		1	1.86
	Private	-	7.84
E-4	New Windsor	1	1.21 ^b
	Industrial	-	0.80
E-4	Oneida	1	0.71
D-4	Ophiem	1	0.75
D-4	Orion		1.49 ^b
	Private	-	1.15
E-5	Toulon	2	5.96 ^{b,c}
		2	3.33
	Industrial	-	1.91
	Private	-	1.57

^aEstimated error 2% or 0.05 $\mu\text{g}/\text{liter}$, whichever is larger.

^bSample collected June 1957.

^cNearby deep sandstone well idle.

Table 10

Concentration of Ra^{226} in deep sandstone well water

Location			$\mu\text{g Ra}^{226}/\text{liter}^a$	Formation
Fig. 24	Municipality	Well No.		
E-3	Alexis	2	0.75 ^b	St. Peters
E-4	Alpha	2	6.40	St. Peters
F-3	Biggsville	2	15.9	St. Peters
D-5	Buda	2	11.1	St. Peters
F-3	Bushnell	2	18.7	St. Peters
		2	21.3	St. Peters
D-6	Depue	1	9.22	St. Peters
		2	9.58	St. Peters
		2	9.68 ^c	
E-4	Galesburg	Florence Ave.	8.30	Galesville
E-4	Galva	3	9.68	St. Peters
E-4	Kewanee	1	8.31	Galesville
		2	6.27	Galesville
		3	7.35	Galesville
F-4	Knoxville	1	10.3	St. Peters
		2	6.31	Galesville
D-6	Ladd	1	7.58	St. Peters
F-3	Monmouth	4	5.28	Galesville
		5	4.53	Galesville
		6	6.82	Galesville
E-5	Toulon	1	36.9	St. Peters
E-3	Viola	1	8.10	St. Peters
E-4	Woodhull	1	4.20	St. Peters

^aEstimated error 2% or 0.05 $\mu\text{g}/\text{liter}$, whichever is larger.

^bPump started for sample.

^cAfter aeration - filtration for iron removal.

In Illinois, there were 756,000 people being served by 150 municipal water systems which obtain all or part of their water from deep sandstone formations (1950 census). To date, the concentration of Ra^{226} has been determined in deep sandstone well water at 56 municipalities, which have a population of 482,000. Of these, 97,000 live in municipalities having Ra^{226} concentrations of less than $3 \mu\mu\text{g/l}$, 126,000 from 3 to $4 \mu\mu\text{g/l}$, and 259,000 greater than $4 \mu\mu\text{g/l}$. The Ra^{226} content of the deep sandstone well water from the remaining 94 sites with a combined population of 274,000 has yet to be determined. In addition, it should be noted that the entire area obtaining water from the geological formations now known to yield water of high Ra^{226} content in Illinois includes most of Wisconsin, Iowa, and Missouri, and parts of Minnesota, Oklahoma and Arkansas.

References

1. H. F. Lucas, Jr., and F. H. Ilcewicz. Radiological Physics Division Semiannual Report, ANL-5829:125-135 (1957).
2. H. F. Lucas, Jr., and F. H. Ilcewicz. J. Am. Water Works Assoc (in press)
3. National Bureau of Standards Handbook 52, U. S. Department of Commerce (1953).

GAMMA-RAY EMITTING RADIOACTIVITY IN SOIL

P. F. Gustafson

A. Improvement in Methodology

Foremost among the improvements made in the course of the soil radioactivity program has been a change in sample size and geometry used in the routine analysis of soil activity. Previously 1 kg of soil, contained in a 1-liter polyethylene bottle placed on top of a 5" x 4" NaI(Tl) crystal, was the standard sample and counting arrangement. Realizing that the geometry under these conditions was far from ideal, a re-entrant stainless steel cup was made which could be filled with soil and placed over the crystal. The crystal and phototube are mounted in a vertical position, the crystal being on top. The cup is inverted and slips down over the crystal. One-sixteenth-inch stainless steel was used in the fabrication of the containers and is sufficiently thick to stop 3-Mev betas. A determination of the optimum size or thickness of soil sample was carried out using equal thicknesses of soil around and over the crystal. The results of using 1-, 2- and 3-inch layers are indicated in Table 11 along with those obtained with 1 kg of soil in the 1-liter bottle. The net counting rate increases with sample size as expected, and the time required to attain a given net count is correspondingly reduced. The degree of attenuation or self-absorption also increases with sample size, particularly for lower energy photons, some of which are of interest in the soil analysis. A 1-inch layer (~2.5 kg) of soil was found to be most desirable from the standpoint of both sensitivity and resolution; the total net counting rate with this amount of soil is 5.4 times that obtained from a 1-kg sample of the same soil placed in a bottle on top of the counter, and the net counting rate per kg is 2.2 times that with the bottle. Whereas some of the weaker samples were previously run for 16 hours, similar net counts can be had in 3 to 4 hours, or a net count 5 times as large may be obtained in 16 hours' counting time. Approximately 2.5 kg of Na_3PO_4 is placed in one of the holders for background measurements, and a similar quantity of this compound spiked with appropriate amounts of the various radioisotopes serve as standards.

Table 11

Study of net total counting rate as a function of the size
of soil sample and of sample-counter geometry

<u>Sample geometry</u>	<u>Weight</u>	<u>Net counting rate</u>	<u>Net counting rate/kg</u>
1-liter bottle	1.00 kg	1.00	1.00
1" layer	2.41 kg	5.39	2.24
2" layer	6.65 kg	10.12	1.52
3" layer	13.00 kg	14.13	1.09

B. Determination of the Cs^{137}/Sr^{90} Activity in Soil

As an outgrowth of the rather simple and straightforward method of analysis of fission products in soil offered by gamma-ray spectroscopy, a test run is being made on the ratio of Cs^{137} activity relative to that due to Sr^{90} in the same soil sample. If this ratio proves to be fairly constant for the same soil type, or more ideally for all soil types, then it should be possible to assay Sr^{90} indirectly by means of the Cs^{137} content of a sample. This would do away with the necessity for laborious chemical analysis for Sr^{90} , thereby omitting both time and cost of running samples.

Arrangements have been made with the Division of Biology and Medicine in Washington, D. C., to run 10 soil samples, selected at random, which have already been analyzed for Sr^{90} . Thus far 5 samples have been received and the Cs^{137} values determined. The activity ratio varies from 1.69 to 2.33 with a mean value of 1.91. This series consists of four samples from Hawaii, two from a 0 to 2-inch depth, two from a 2- to 6-inch depth, and one sample from Ithaca, New York, a 0 to 2-inch depth. Although there is considerable difference in soil type between the New York sample and those from Hawaii, the ratio does not change appreciably. The anomalously high value of 2.33 was obtained on one of the Hawaiian samples (2- to 6-inch depth) which had a Cs^{137} concentration of less than 0.5×10^{-10} curie/kg. With this order of activity it is roughly estimated that the statistical error of counting can become high enough to account for the difference. The individual ratios are tabulated in Table 12.

Table 12

Cs^{137}/Sr^{90} activity values in soil

Beltsville No.	Site	Collection date	Depth	Activity ratio
561144	Ithaca, N.Y.	10-56	0 - 2"	1.91
57996	Kawailoa Girl's School, Hawaii	12-57	0 - 2"	1.77
57997	Kawailoa Girl's School, Hawaii	12-57	2 - 6"	1.83
57998	Leilehua Golf Course, Hawaii	12-57	0 - 2"	1.69
57999	Leilehua Golf Course, Hawaii	12-57	2 - 6"	2.33*

Average 1.91

*High value - see text

In peeling apart the spectrum of the New York soil sample, a line appeared at 0.84 Mev as shown in Fig. 25. The peeling technique shows a slight surplus of counts in the vicinity of 1.1 Mev in the case of the two 2-to 6-inch depth Hawaiian samples. The 0.84-Mev line is tentatively assigned to Mn^{54} on the basis of energy and of its having a sufficiently long half-life to be still present in significant amount (the New York sample was collected in late 1956). The 1.1-Mev line (assuming it is indeed a line, as there are not sufficient counts to observe a true peak) may be due to Zn^{65} . Both of these radioisotopes are activation products, with apparently high yield in thermonuclear explosions, and both have the requisite fairly long half-lives so that they are able to outlast the more abundant, but shorter lived, fission products.

Fig. 25

Gamma-ray spectrum 5" x 4" NaI with natural activities removed; 16-hour run BIM steel room 2.2 Kg soil sample No. 561144-1

THE HUMAN SPECTROMETER

Introduction

C. E. Miller

As in previous reports from this Division, under this heading are collected some of the research activities carried out in our low-level facility with scintillation gamma-ray spectrometer. With the availability of three iron-shielded rooms on our site, these activities have expanded to include measurement of radioactivities in soil and some much needed developmental studies concerning stability and magnitude of background pulse height scintillation spectra. To obviate the considerable editorial work necessary to the coordination of all phases of this work, we report under this section only activities that have taken place in the first iron shielded room, and these concern mainly experiments in vivo. The remainder of the work is reported under the authorships of P. F. Gustafson and H. A. May.

I. The Gamma-Ray Background of Iron Room I (Airborne Radioactivity)

C. E. Miller and J. B. Corcoran

The gamma-ray background within a heavily shielded enclosure has been discussed in previous semiannual reports.⁽¹⁻⁶⁾ Extensive data are given in the last report⁽⁶⁾ regarding the reduction of the gamma-ray background of an 8-inch diameter by 4-inch thick NaI(Tl) crystal as a function of thickness of Fe shielding. The data given herein are, in a sense, an extension of the past reports, for they concern the ultimate sensitivity attainable by our methods. The background of Iron Room I had been observed to fluctuate because of three different airborne radioactivities. At times the fluctuations were traced to the presence of A^{41} from the research reactor or of N^{13} from the cyclotron⁽⁴⁾ by the simple inspection of the photopeaks resulting therefrom. The presence of even a very small concentration of either of these products has seriously affected the accuracy of measurements and, at times, has forced a suspension of all runs.*

To overcome this difficulty a storage air system was installed so that air free of radioactivity could be fed to the iron rooms. The storage system consists of two 500-cubic foot tanks which can be filled to 10 atmospheres. Through appropriate valves, air can be pumped into, or used from, either tank independently of the other. Normally, the two tanks would be

*Similar inconveniences were experienced by Dr. P. F. Gustafson in the low-level facility installed in the Biological and Medical Research Division (vide infra).

filled when the wind is not blowing from the above sources. In the case of a prolonged southerly flow of several days, one tank can be filled and the air aged for 24 hours to allow the 1.83-hr A^{41} (or the shorter lived N^{13}) to decay while air is being used from the other tank. In other instances the difference between routine background spectra has been large enough to yield a true $Ra(B+C)$ spectrum, hence it must be presumed to be due to fluctuations in the amounts of radon daughters present within the room. The possibility that parts of these fluctuations are due to translocations of the active deposit within this room cannot be totally excluded although it seems unlikely because, to our knowledge, there is no substantial electric field present in the room, all electric cables being covered by grounded shields. In order to obtain some idea of the effect produced by the presence of radon in the room, air which was aged for ten days, was used to inflate two plastic weather balloons placed on either side of the crystal which was located in the center of the room. Under these conditions the background decreased from 915 to 860 cpm and climbed to 895 overnight as the balloons deflated partially and basement air diffused into the room. This experiment confirmed our suspicions on the detectable role of $Ra(B+C)$ in the background, but because of the long time taken in the readings, it did not yield any information on either the location of the active deposit or its magnitude as a function of Rn concentration in the room.

To obviate contamination with Rn spilled in room, the latter was sealed on the outside with polyethylene sheeting which was in turn taped to the floor. The room was kept sealed for six days during which the background climbed from 965 to 1212 c/m. The growth of the background, assessed at 0.75, 1.75, 2.75 and 5.75 days after sealing, bore to a typical radon growth curve arbitrary ratios of 4.65, 3.72, 4.45 and 4.26, strongly suggesting that the increase was due to radon growing from a virtually freely emanating Ra source. At the peak the Rn concentration was $3.25 \mu\mu c/\text{liter}$. Upon flushing the now unsealed room with aged tank air ($0.002 \mu\mu c/\text{l.}$) the concentration fell to $0.15 \mu\mu c/\text{l.}$ and the background counting rate dropped to 907 c/m. Left overnight, the Rn concentration climbed to $0.22 \mu\mu c/\text{l.}$ and the counting rate to 911 c/m. These data indicate that the background count between 25 kv and 1575 kv increased by approximately 100 c/m per each $\mu\mu c/\text{l.}$ in the room.

Under the conditions of the experiment either (or both) the walls of the room or the concrete floor may be responsible for the production of radon; from what is known about the radioactivity and porosity of building materials, the floor is by far the most probable. Further study of this problem will lead to improvement in stability of background.

One puzzling feature remains. After sealing the room, if the circulating air blower within the room is kept running, the background does not change appreciably ($\pm 2\%$), even if the seal is kept as long as five days.

Apparently circulation of air must contribute to a translocation of Ra (B+C) away from the crystal, for it is difficult to envision this ventilation preventing diffusion of radon from the floor into the room. This phenomenon may be of importance in methods aimed at minimizing and stabilizing that portion of the background count due to decay products of radon.

The authors are pleased to acknowledge their indebtedness to Mr. H. F. Lucas, Jr., for his measurements on the concentration of radon in the air of the steel room.

II. Ra²²⁶ and Th²³² Activity in Humans

C. E. Miller

As discussed in the previous semiannual report,⁽⁶⁾ a relatively large number of individuals who have accumulated significant quantities of either radium or thorium some 30 years ago are the object of a systematic study in this laboratory.

A. Radium Subjects

Eleven women who painted radium watch dials during the 1920's have been brought in for total body gamma-ray measurements. The radium body content of these subjects has ranged from 10^{-9} to 2.5×10^{-6} c of Ra²²⁶. Total skeletal x-rays of twenty of the dial painters have been obtained under the direction of A. J. Finkel, M.D., Director of the Health Division, Argonne National Laboratory. To date, the number of people measured is 68, and the number x-rayed is 36.

In addition, the radium body contents of two individuals who received radium therapeutically 20 to 30 years ago have been determined. One individual (H.T.) contained $4\mu\text{c}$ and the second (C.F.G.) contained $4.2\mu\text{c}$. Two individuals, (B.M.) and (W.F.), reported previously from our laboratory, who received the material from the same source had burdens of $1.2\mu\text{c}$ and $5.2\mu\text{c}$, respectively. Since these cases are discovered usually as a result of clinical or roentgenological symptomatology, it should not be surprising that their burdens are, as a whole, well above those found in cases of industrial exposure. What is comforting, but not reassuring, is the fact that the minimum symptomatological burden has not decreased since 1952.

B. Study of Thorotrast in Humans

The evaluation of the long-term effects of both radium and thorium is complicated by the fact that both elements are the parents of long families of radioactive daughters. The problem is particularly difficult in the case

of thorium since this element decays through a series of 6 others, two of which are long-lived, before it reaches the stable element Pb^{208} . The daughters are metabolized and excreted in different manners. Thus the metabolic pattern of each daughter must be understood in detail before integral dose calculations for particular organs can be made.

Reynolds, et al. (7) calculated the retention and elimination of radium isotopes produced by the decay of thorium parents within the body (calculations and comparison with experimental findings). These results are being verified by clinical studies now in progress following the I.V. injection of Thorotrast to appropriate subjects under medical care. The amount of thorium-232 and of each daughter is determined before the material is administered. The empty vial and hypodermic syringe are measured after the injection to determine the amount of Th^{232} and of each daughter remaining on the walls of the glassware. The quantity of each element administered to the patient is thus known. At a later date, the amount of Ra^{228} , Th^{228} , Ra^{224} , Pb^{212} and Po^{212} present in the liver, kidney and bone is determined.

One case has been studied to date. Measurements have demonstrated that a large fraction ($\sim 75\%$) of the Ra^{228} daughter remained in the vial. Consequently, it is imperative that the empty vials be measured carefully since a small error in the measured value remaining in the vial results in a large error in the estimate of the quantity administered. This situation does not exist in subjects to whom the material was injected many years ago, for most of the Ra^{228} present is the result of growth from Th^{232} retained in the body.

III. Techniques Which May Yield More Accurate Ra^{226} Measurements in vivo

C. E. Miller and O. J. Steingraber

A. Coincidence Techniques

As discussed in the last semiannual report, (6) the practical uncertainty in an in vivo radium determination using the tilting chair technique is about 3×10^{-9} c. This large uncertainty is due to the presence in the human body of small quantities of fission products. For example, the relatively abundant 607-kev gamma ray emitted by RaC' cannot be easily distinguished from the 661-kev gamma ray of Cs^{137} . Consequently, the human's normal Cs^{137} entire spectrum cannot be subtracted accurately from the subject's gross spectrum thus preventing an accurate measurement of the RaB gamma rays at 241, 295 and 351 kev.

Theoretically, the interference of the Cs and K gamma rays may be eliminated by analyzing only those gamma rays which are in coincidence (timewise) with other gamma rays. This is the case of the abundant 607-Mev gamma ray emitted by RaC' , which is always emitted in coincidence with one of several higher energy gamma rays. Since Cs and K are both monoenergetic emitters, only chance coincidences and those gamma rays which undergo a Compton scatter in one crystal and then are absorbed in the second crystal would produce a count. The scattered gamma-ray coincidence can be eliminated by placing the crystals on opposite sides of the subject so that a gamma ray would have to scatter through 180° (thereby losing all but 250 kev) to reach the second crystal. The pulses resulting from these back scattered photons can be eliminated by adjusting the pulse height analyzers to reject all pulses of less than 500 kev.

The number of coincidences can be measured in the conventional manner whereby the pulses from one crystal are used to gate the analyzer, and only the pulses from the second crystal are analyzed as to height. Obviously, the number of detected events will be doubled if the pulses from either crystal can gate the analyzer and the pulses from the opposite crystal are sorted. Equally obvious is the fact that the pulse from one crystal should not be able to gate the analyzer and be itself sorted and recorded.

Fig. 26

The electronic equipment which has been used to evaluate the feasibility of this approach is shown in block form by Fig. 26. The pulses from both crystals are fed to a summing network (Σ) and the sum pulse applied to the input of the 256-channel analyzer. In addition, the individual pulses from each crystal are amplified and analyzed by wide-band single channel pulse height analyzers. The output pulses of the single channel analyzers are applied to the coincidence circuit. The coincidence circuit in turn gates on the 256-channel analyzer and permits it to sort the sum pulse whenever pulses are received simultaneously from both single channel analyzers.

Two NaI(Tl) crystals, one $2\frac{1}{2}$ inches in diameter by $2\frac{1}{8}$ inches and one 4 inches in diameter by $1\frac{1}{2}$ inches thick, were used for the preliminary tests. The pulse height discriminators on the single channel analyzers were set to accept any pulse over

500 kev. The upper gate was not used at this time, however, but the possibility of its use should be kept in mind for this would reduce the background since the cosmic ray showers would be removed.

Fig. 27. Sum-Coincidence Spectrum from Ra C'

a factor of 10. An additional reduction will be realized by placing a plastic anticoincidence shield over the assembly to eliminate coincidences due to cosmic rays.

The counting rate from 1300 grams of natural K (ten times the amount present in the total body) placed between the crystals was found to be equal to background. Thus the small amount of K present in a subject's leg will not yield a detectable counting rate.

This system could lead to a more accurate measurement of the RaC' content of the body if sufficiently large crystals are used.

B. Direct Measurement of the Radon Partition Ratio

It is not possible to calculate directly the quantity of Ra^{226} present in the human body using the gamma-ray spectrum of the whole body. The 188-kev gamma ray which is emitted by Ra^{226} in 5.7% of the disintegrations is obscured by the numerous degraded gamma rays from the radium daughters, from the Cs^{137} , and from the K^{40} present in the subject's body. Thus the amount of Ra^{226} present must be determined indirectly by measuring the quantity of the Ra^{226} daughter products.

The body content of RaB and of RaC' can be measured quickly and with high accuracy using gamma-ray techniques. Unfortunately, the body content of Ra^{226} is larger than the body content of RaB or RaC' since a

The spectrum of the sum pulses when $2 \times 10^{-7}\text{c}$ of radium was placed between the two crystals with a 10-cm spacing is shown by Fig. 27. The distinct 1.38-Mev and 1.73-Mev photopeaks are the sums of the 0.607-Mev plus the 0.77-Mev gamma rays and the 0.607-Mev plus the 1.12-Mev gamma rays of RaC'. A less intense coincidence pair which yields a sum peak at 1.54 Mev shifts the high side of the 1.38-Mev peak. Other coincidence pairs at sum energies of 1.85, 2.12 and 2.2 Mev account for the area under the curve above 1.8 Mev.

The gamma-ray background, outside of the iron shield and without the use of an upper limit gate to remove cosmic rays, is low, namely, equal to the counting rate which is obtained from 10^{-9}c of radium. Placing the assembly inside the iron room will reduce background by

large fraction of the radon is exhaled. Consequently, an accurate Ra^{226} determination requires, in addition to a measurement of the RaB and RaC' , a measurement of either the radon in the breath or a measurement of the $\text{Ra}^{226}/(\text{RaB RaC}')$ ratio (radon partition ratio).

The addition of a second gamma-ray measurement which would provide the radon partition ratio directly has the obvious advantage that the breath measurements would be unnecessary. The interfering degraded gamma rays from the body which swamp the 188-keV gamma rays from Ra^{226} can be reduced by measuring the spectrum of the kneecap, elbow, shin or ankle with a collimated NaI crystal. These volumes contain a large mass of bone with the least mass of surrounding tissue. The $\text{Ra}^{226}/(\text{RaB RaC}')$ ratio found for one of these volumes is thus assumed for the total skeleton. The whole body $\text{RaB RaC}'$ content is then multiplied by this ratio to obtain the total body Ra^{226} content.

The photopeaks from the 188-keV gamma ray from radium as well as the photopeaks from the 241-, 295-, and 351-keV gamma rays of RaB can be distinguished quite clearly in the spectra obtained from the kneecap and ankle (see Fig. 28). It will be necessary to obtain spectra using appropriate phantoms containing both de-emanated radium and radon in order to peel the spectra from the kneecap and ankle apart to determine the ratio.

Fig. 28

Gamma ray spectrum of
subject EVR's kneecap

A phantom has been constructed using Presdwood to simulate the flesh and using a plaster of Paris bone. Results will be given in a later publication.

IV. Follow-Up of U²³⁵ Inhalation Incident

C E. Miller

In a previous report⁽⁶⁾ data were given concerning a machinist who had inhaled U²³⁵ dust during a grinding operation. The quantity present in various volumes of the subject's body had been measured on three occasions during a span of 38 days. The subject was measured a fourth time 98 days after the third measurement. The quantity present in the subject's chest on the four occasions, as well as the quantity eliminated in the excreta between measurements, is given in Table 13. (The excretion measurements were made at the subject's laboratory and are given here with the latter's permission.)

Table 13

Data on U²³⁵ subject

Date of measurement	Total amount of U ²³⁵ in chest area (mg)	Decrease of U ²³⁵ (mg)	Quantity of U ²³⁵ in excreta during these periods (mg)
November 2, 1957	2.75	0.7	0.74
November 19, 1957	2.05	0.58	0.48
December 10, 1957	1.47	0.77	0.78
March 18, 1958	0.7	—	—
Total		2.05	2.00

The four values do not follow a power function but suggest a short half-time in the lungs of 41.5 days for the first 38 days followed by a longer one. Although the average half-time between the third and fourth measurement is 92 days, this would be a minimal value for the slower phase since the U²³⁵ may have been eliminated at the higher rate for a period of time after the third measurement. It is now apparent that a measurement 20 days after the third one might have been of considerable value.

The subject's head and feet, which constitute a fair percentage of the subject's skeleton, did not contain a detectable amount of U²³⁵. Thus, it is doubtful that the longer half-life is particularly influenced by clearance rates from the bone.

The agreement between the decrease in lung content and the quantity present in the excreta demonstrates that the lung measurements are quantitatively correct if the assumption is made that elimination from other tissues is considerably faster than that taking place in the lungs.

V. The Elimination Pattern of Co^{60} Acquired by Inhalation

C. E. Miller and H. A. May

Two employees, herein designated A and B, inhaled a small amount of cobalt dust while unpacking a vessel which had become contaminated with Co^{60} dust during a grinding and polishing operation. The vessel had been scrubbed down several times, then wrapped and moved to another area for further cleaning. The employees inhaled the dust which had remained on the vessel and which became airborne as it was unwrapped.

The total body content of subject A was measured on the 2nd, 4th, 7th, 10th, 18th, and 50th day, after the accident. The distribution within subject A's body was measured on the 3rd, 4th, 7th, and 10th day. The total body content of subject B was measured on the 2nd, 4th, and the 24th day after the incident and the distribution within the body was measured only on the 4th day.

Only the data taken on subject A will be discussed since they are, in essence, representative of both cases. Measurements with the standard chair technique and measurements with a collimated crystal were made over various areas of the body, as illustrated in Fig. 29 with the results shown in Table 14.

Fig. 29

Table 14

Co⁶⁰ activities (in arbitrary units) obtained by scanning subject A

Position	1	2	3	4	5	6	7	8	9	10
Date										
6-9-58	-	-	-	Dust inhaled	-	-	-	-	-	-
6-12-58	22	30	23	28	32		42		170	3.5
6-13-58				18		22	26	26	100	
6-16-58	16			4.7		8.4	10	22	6	
6-19-58	10			5.4		7.7	8.7	5		
6-27-58	7			6.5			7.5			

As shown by Fig. 30 the total body content of subject A decreased with a half-time of 1.6 days from the 2nd day through the 7th day and with a half-time of 42 days from the 17th through the 50th day. The content of the chest seems to be decreasing with a half-time of 52 days and the content of the abdomen with a half-time of 62 days.

Fig. 30

The data of Table 14 disclose that the quantity at position No. 4, probably the lymph nodes, decreased until the 7th day and then increased slightly. This phase may account for the longer half-time found over the chest. The 62-day half-time for the abdomen is probably influenced by the presence of the material in the liver.

VI. In Vivo Measurement of Cs^{137}

C. E. Miller and L. D. Marinelli

The radioactive body contents of a group of individuals, living in the vicinity of our Laboratory, have been measured periodically since March 1955, and reported last in ANL-5755. These subjects, members of our Division, do not inhale or ingest any radioactive materials other than that present in ordinary food, water and air. Thus, the radioactivity present in their bodies should be typical of that present in individuals living in the Chicago area.

The Cs^{137} content of 8 subjects, plotted as a function of time since 1955, is shown in Fig. 31. The Cs^{137} content is expressed in units of $\mu\mu\text{c Cs}^{137}$ per gram of body K to permit a direct comparison between subjects without regard to body musculature. The Cs^{137} content is seen to have increased from March 1955 until May 1956 at which time it leveled off and remained at essentially a constant value until January 1958. Latest measurements suggest that since then the Cs^{137} content has increased.

Fig. 31

The gamma-ray spectra of subject HAM obtained on January 16, 1958 and July 28, 1958 (given in Fig. 32) clearly demonstrate the increase in Cs^{137} content in the presence of constant K^{40} content.

Fig. 32

Details of the Cs^{137} content of 13 subjects on four dates: April 1956, June 1957, April 1958 and July 1958 are given in Table 15. It is obvious that, despite individual fluctuations, the average value has increased from $34 \mu\mu\text{c Cs}^{137}/\text{gK}$ during 1956-1957 to $52 \mu\mu\text{c Cs}^{137}/\text{gK}$ at the present time. Since Cs^{137} has been found to have a biological half-life in man of the order of 150 days, these values should be roughly proportional to the average rate of fallout for the preceding period of 6 months if no abrupt changes have occurred in the dietary habits of the individuals.

Table 15

Cs^{137} in residents of Chicago (42° N)

Females $\mu\mu\text{c Cs}^{137}/\text{gK}$					Males $\mu\mu\text{c Cs}^{137}/\text{gK}$				
	4-56	6-57	4-58	7-58		4-56	6-57	4-58	7-58
K.G.	27	22	33	38	R.R.	21	25	31	39
L.S.	24	31	29	35	W.P.	20	28	35	44
I.S.	24	33	43	52	B.O.	31	31	42	47
C.L.	42	36	51	57	O.S.	30	36	43	48
N.K.	50	50	59	71	H.M.	45	41	65	71
					C.M.	39	41	44	43
Average	36	34	43	51	E.M.	-	45	62	64
					J.C.	-	42	55	51
					Average	33	36	47	52

References

1. Report on Biological, Medical and Biophysics Programs ANL-5456: 107-123 (1955).
2. Report on Biological, Medical and Biophysics Programs ANL-5518: 45-57 (1956).
3. Radiological Physics Division Semiannual Report ANL-5596: 26-33 (1956).
4. Radiological Physics Division Semiannual Report ANL-5679:22-37 (1957).
5. Radiological Physics Division Semiannual Report ANL-5755:47-61 (1957).
6. Radiological Physics Division Semiannual Report ANL-5829:144-165 (1958).
7. Reynolds, J. C., P. F. Gustafson and L. D. Marinelli. Retention and Elimination of Radium Isotopes Produced by the Decay of Thorium Parents within the Body - Calculations and Comparison with Experimental Findings. ANL-5689 (1957).

SOME INVESTIGATIONS ON THE SOURCE OF BACKGROUND COUNTS IN NaI(Tl) CRYSTALS

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

The almost continuous use of the BIM steel room and the 256-channel analyzer for soil radioactivity measurements has shed a considerable amount of light on the nature of the background radioactivity as well. A representative work week will include one to two 16-hour overnight background runs using the Na_3PO_4 soil blank and a daily 1-hour background also using the soil blank. The background over the spectral region from 0.1 to 2.8 Mev is reproducible within a few percent from day to day using the soil blank. However, there are times when a significant departure from the average background counting rate does occur because of contamination of the room air with A^{41} and/or N^{13} from CP-5 and the cyclotron respectively. If A^{41} contamination is present, the background increases at most 10 to 15 percent over the normal value, and somewhat less in the case of N^{13} . Although we hope to catch the presence of such contamination by frequent background runs, a further protection against accidentally assigning A^{41} counts to radioactivity within the sample has been devised. This is simply to determine the concentration of uranium and thorium in the sample by using two spectral regions, one located above 1.3 Mev (the A^{41} photopeak energy) and the other below. Essential agreement between the two sets of concentration values implies the absence of contamination; disagreement is most likely due to contamination and the reading is then repeated. N^{13} contamination is less frequent and must be detected in the background run, or else the computed concentrations of the various fission products will be correspondingly in error.

Representative counting rates from 0.1 to 2.8 Mev are 2400 counts per minute for the soil sample, and 280 counts per minute for the background. Thus in 16 hours there are about 2×10^6 net counts. The observed variation in background due to A^{41} but present in the region 0.6 to 0.7 Mev is equivalent to 4×10^{-11} curie of Cs^{137} per kg of soil.

Some other features of the background spectrum became apparent as a result of the soil program and were investigated in varying degree. These are the line at 0.511 Mev presumably due to positron annihilation, a line in the region 3.2 to 3.4 Mev (hereafter referred to as 3.2 Mev), and a comparison between the background spectrum in lead and in steel shielding. This comparison between the shielding afforded by 9 inches of steel and 4 inches of lead was an outgrowth of a study concerned with the 3.2 Mev line, but will be discussed first because it is of more direct concern to the low-level measurements of soil radioactivity.

*Biological and Medical Research Division

Background Spectra with Steel and Lead Shielding

Spectra using the BIM 5" x 4" NaI crystal were run inside the steel room with the crystal unshielded and with a one-inch shield of mercury around and over the crystal and phototube assembly. The mercury shield consists of a stainless steel shell with 1/8" wall thickness, with one-inch spacing between inner and outer wall which is then filled with triple distilled mercury. A disk of similar construction fits over the top of this cylindrical shield. The spectra thus obtained are shown in Fig. 33 together with the difference spectrum, i.e., the spectrum obtained without the mercury shield minus that using the shield. This difference spectrum clearly shows the principal lines of the radium spectrum which are assumed to be due either to radium present in the steel plates or to radon daughters in the room air or adhering to the inner surfaces of the room. The amount of radium or radon present corresponds to 2.5×10^{-11} curie of radium counted with the 5" x 4" crystal under 50 percent geometry. Fig. 33 also shows a spectrum taken inside the lead cave with 4-inch lead shielding without additional mercury shield. The radium lines are not as pronounced, and a difference spectrum failed to show any discrete structure due to radium.

Fig. 33

High energy gamma-ray spectrum of 5" x 4" NaI crystal; 16-hour runs February, 1958

The radium and/or radium daughters existing within the steel room present a limit to the sensitivity attainable with the present shield. This points to

the desirability of having, in measurement of small samples, a space around the crystal and sample as small as possible in order to limit both the volume of air and the surface area "seen" by the crystal.

Background above 2.6 Mev.

The method used initially to assay gamma-ray activity in soil was to peel the spectrum apart, starting with normalization of thorium at the 2.62-Mev photopeak from ThC'' .⁽¹⁾ The background spectrum was noticed to rise beyond 2.62 Mev and an actual peak appeared in the vicinity of 3.2 to 3.4 Mev (Fig. 33). The main concern was whether or not this line varied in intensity from time to time, either in the sample itself or in the background. If such were the case, this might interfere with the evaluation of the thorium concentration, and in turn invalidate the entire analysis. This line was found to be fairly constant in the background over periods of a few weeks and bore no relationship to either A^{41} or N^{13} concentration; furthermore it was found to vary directly with the 2.62-Mev line from ThC'' in the presence of a soil sample or other thorium containing material. Thus the net spectrum from monazite sand, for example, indicates peaks at both 2.62 and 3.2 Mev. The ratio of peak intensity of the 2.62 to that of the 3.2-Mev line was about 15:1 when the monazite sample placed directly on top of the 5" x 4" crystal. Geometrical considerations are in keeping with the 3.2-Mev line being a sum of the 2.62- and 0.58-Mev gamma rays emitted in cascade during the decay of ThC'' . The matter was dropped at that point, but interest in the subject was renewed after a visit by Dr. Van Dilla from Los Alamos, who mentioned the presence of this line in the background spectrum using an 8" x 4" NaI crystal in the Los Alamos steel room.

Further observations in the BIM steel room continued to show this line. An early speculation involved the possibility of some connection between the steel shield (~ 70 tons) and the 3.2-Mev line. This could come about either through internal contamination of the steel, or by means of some nuclear reaction between iron and high energy particles and/or neutrons. The same 5" x 4" NaI crystal was thus moved to the lead cave and a 16-hour background spectrum taken. The 3.2-Mev photopeak was still present, in fact the intensity appeared to be the same as in the steel room (Fig. 33). This constancy with shielding was further borne out by the use of the above-mentioned mercury shield around the crystal in the steel room. A distinct lowering of the peak followed, but since the counts lost were about the same as those lost at higher pulse heights (Fig. 33), it was concluded that the counting rate in the line did not change but that the background within the same energy limits had been decreased by the presence of the mercury shielding, possibly because of attenuation of cosmic radiation. A test designed to relate this peak to the ionizing component of cosmic radiation is presented elsewhere in this report by Mr. H. A. May. The high capture cross section of Hg for thermal neutrons seems to eliminate the possibility of this peak being due to neutron capture gamma rays.

This points very strongly to the possibility of internal contamination by natural radioelements, either in the NaI itself or in the canning envelope. Although both the uranium and thorium series have either primary or summation gammas of energy greater than 3.0 Mev, their intensity, relative to that of lower energy gammas, is on the order of 10^{-2} to 10^{-4} ,⁽²⁾ whereas in our spectra we observe ratios well within an order of three. The new (UCLA crystal) 5" x 4" NaI also showed a 3.2-Mev peak of somewhat greater intensity than seen with the BIM 5" x 4" crystal. Using the new 5" x 4" crystal as the detector, the old crystal was used as a source and a 64-hour count taken inside the mercury shield. The hope was that if the 3.2-Mev line was a true gamma-ray line or sum peak (2.62 + 0.58 Mev) either the 2.62- or the 3.2-Mev line would be augmented by counts coming from the second NaI crystal. Comparison was made with a 64-hour run using only the new 5" x 4" crystal inside the mercury shield. The presence of the second crystal served only to reduce the counting rate, much as did the mercury. In the region 2.8 to 3.8 Mev 65,001 counts were obtained with the new 5" x 4" crystal in 64 hours, whereas 63,111 counts were accumulated in the same period and under the same conditions with both crystals. From 2.5 to 2.7 Mev a similar effect was noted, the counts in 64 hours going from 11,236 to 10,617. An attenuation was also noted in the region beyond 4 Mev. The geometry was on the order of 25 percent, and the net counts in the region 2.8 to 3.8 Mev were estimated to be 55,300 in the case of the new 5" x 4". This estimate was made by extrapolating the continuum which exists above 4 Mev down below 2.8 Mev. Allowing for the lesser intensity of the 3.2-Mev line in the case of the BIM 5" x 4" crystal (48,000 net counts, estimated as above) there should have been on the order of 10,000 additional counts in 64 hours with the two crystals counted simultaneously. The results thus tend to indicate that the 3.2-Mev line is not due to internal gamma rays in any way.

The possibility remained that the bulk of this peak might be due to alpha particles instead of photons. The light output from an alpha particle in NaI(Tl) is roughly 1/2 that arising from an electron of the same energy (more nearly correct when the alpha energy is greater than 6 Mev). All that was necessary was to find a suitable alpha source to fit the observed peak. The uranium series, namely Ra^{226} and its daughters, was the first suspect. A small crystal was spiked with a bit of radium, and the alpha-beta-gamma spectrum was taken. The alpha-to-electron pulse height relationship was in fairly good agreement with the data of Taylor *et al.*⁽³⁾ However, the radium spectrum did not match that originating in the 5" x 4" crystal; there were peaks above 3.4 Mev as well as below 3.0 Mev, and the beta continuum was considerably more intense in the case of the spiked crystal. Similar testing of the thorium and actinium series remains to be done because most of the characteristics of the spectrum from the 5" x 4" crystal can be accounted for by the daughters of Th^{228} or of U^{235} .

A further thought was that thallium might have a hitherto undetected alpha-emitting isotope; that is, either Tl^{203} or Tl^{205} might be radioactive. Using the observed counting rate (3.2 Mev) as an indication of specific activity in the 5" x 4" NaI crystal and computing the number of thallium nuclei present, a half-life on the order of 10^{17} years was obtained. However, an alpha energy of some 6 to 7 Mev is not in keeping with this half-life according to the theories of alpha decay.

Another facet to the puzzle was indicated by using smaller crystals; two 2" x 2", and one 3" x 2" NaI, none of which showed any peaks beyond the 1.76- or at most the 2.17-Mev lines from Ra^{226} daughters as illustrated in Fig. 34. Since there were no peaks above 2.2 Mev, this rules out radioactivity due to thallium as a source of the 3.2-Mev line. All of the evidence points to this energetic line as being a unique feature of large crystals and/or photomultipliers with some possibility of its being due to internal contamination by some long-lived series.

Fig. 34. Gamma-ray spectrum of new 3" x 2" NaI crystal; 16-hour run BIM steel room inside 1" mercury shield (5-28-58)

A further point should be discussed relative to the source of this line, namely, that its intensity has increased about 20 percent in the last 250 days. Fortunately, some data exist from November, 1957, which show the 3.2-Mev line as well as the cosmic radiation continuum beyond up to 6 Mev. Using the continuum for purposes of normalization, the intensity

as a function of time may be computed. Such an increase in intensity would indicate growth from a long-lived parent. Members of any of the four principal radioactive series may be involved; however, from the standpoint of alpha energies, the thorium and actinium series are most promising. In fact actinium has some advantage in this regard since the most energetic alpha (7.43-Mev from Po^{211}) is within the range of electron energies encompassed by the 3.2-Mev peak as shown in Fig. 35. The thorium series does match at all points fairly well, too, but should also have a peak at almost 5 Mev due to the 8.95-Mev alpha from Po^{212} . This line, however, should be smeared by the 2.25 β -ray spectrum of its parent Bi^{212} , for its half-life is only 0.3 μs , and it is too short to be resolved by a NaI crystal.

Fig. 35

Gamma-ray spectrum of new 5" x 4" NaI crystal. Cosmic-ray component removed. 64 hour run in BIM steel room inside 1" mercury shield (5-23 to 5-26-58)

As shown in Fig. 35 aside from a large peak from 3.0 to 3.60 Mev there are also peaks around 2.5, 2.2 and 1.8 Mev. One might postulate then that the reason that only larger crystals have such contamination is that they are grown or otherwise processed in an area where such radioactivity exists, whereas the smaller crystals are located in a cleaner area. To tie down the source of this line unequivocally will not be an easy task but may well be worthwhile if reduction of background in this energy region is essential. This will be more emphatically true since the intensity is increasing and there is no a priori way of determining its ultimate value at this time.

Line at 0.511 Mev.

The other line which has been investigated to some extent is the one at 0.511 Mev. Although N^{13} is a certain source of such radiation, there is

Fig. 36

Gamma-ray spectra showing
0.511 Mev line; new 5" x 4"
NaI crystal in BIM steel room;
3.25-hour runs. (5-22-58)

always a fairly prominent peak at this energy even when the cyclotron is not in operation. This line again appears to be due to the crystal and/or photo-tube material rather than to external sources. Figure 36 shows this energy region with the new 5" x 4" crystal run in the steel room with and without mercury shielding added. There is only a moderate reduction ($\sim 30\%$) in the true intensity of this line with one inch of mercury, and the difference spectrum does not show any pronounced peak at this energy. It is difficult to imagine that the production of events giving rise to this line would equal approximately the attenuation of the 0.511-Mev line in one inch of mercury. There is an indication that the intensity of the 0.511-Mev line increased when the BIM 5" x 4" crystal was counted by the new 5" x 4" crystal; an increase of almost 2000 counts in 64 hours was noted out of some 16,000 counts assigned to this line with the new 5" x 4" crystal during a 64-hour period. Expressing the intensity in terms of counts/16 hours/kg of NaI, the new 3" x 2" crystal was found to have 1.33 greater counting rate at 0.511 Mev than the new 5" x 4" crystal. More research, using a wider variety of crystals is intended. The most that can be said at present concerning this

line is that it doesn't seem to be related in any way directly to the 3.2-Mev line, nor is it solely a feature of large crystals.

References

1. Gustafson, P. F., L. D. Marinelli and S. S. Brar. Radiological Physics Division Semiannual Report ANL-5829:136-143 (1957).
2. Dzhelepov, B., S. Shestopalova and I. Uchevatkin. "On the 2450-3400 Kev Region of the RaC Gamma Spectrum," Nuclear Physics 5:413-414 (1958).
3. Taylor, C. J., W. K. Jentschke, M. E. Remley, F. S. Eby and P. G. Kruger. Phys. Rev. 84:1034 (1951).

ON THE NATURE OF THE 3.25-Mev PEAK IN LARGE CRYSTALS

H. A. May

A small peak in background counting rate at a pulse height corresponding to total absorption of a 3.25-Mev gamma ray was observed and called to our attention by Dr. P. F. Gustafson of the Biological and Medical Research Division. The same effect is observed in a crystal of similar size (5-inch diameter by 4 inches, Harshaw W338) and to a considerably lesser extent in the 8-inch by 4-inch crystal currently in use in Iron Room I. The three spectra are shown in Fig. 37; the curve for the 8" x 4" crystal is normalized in the 1.5- to 2-Mev region. No attempt has been made to observe the effect in smaller crystals because of its small magnitude. Since this energy is well above that of any known monoenergetic gamma emitter, the source was investigated. The energy given is only approximate, as the observed "peak" is poorly resolved. Table 16 shows representative data for the three crystals over the energy band from 2.5 to 3.8 Mev. All data were taken inside at least 8 inches of Fe shielding with runs of at least 1000 minutes.

Fig. 37

Table 16

Crystal size, in.	Counts/min for underlying smoothed-out curve	Counts/min over smoothed curve
5 x 4 (BIM)	18.6	5.5 ± 1
5 x 4 (W338)	23.2	9.0 ± 1
8 x 4	51	3.8 ± 1

The possibility of this anomaly being connected with the ionizing component of the cosmic ray background was considered. Two coincidence runs were made with the 5" x 4" (W338) crystal and 256-channel pulse height analyzer. The coincidence signal was obtained from a bank of seventeen 2-inch diameter GM tubes lying in a horizontal plane over the outside top surface of a small Fe cave. The first run showed a small increase in the coincidence rate, sharply peaked at 3.27 Mev. There were, however, indications of analyzer malfunction in the upper channels. A second 1500-minute coincidence run, taken after minor circuit adjustments were made, resulted in a smooth curve. Without evidence of a peak, it was thus concluded that the increased counting rate noted is not due to ionizing component radiation. We suspect the presence of a naturally-occurring alpha active impurity in the crystal or reflector material.⁽¹⁾

Background Counts Connected with Cosmic Radiation

Data have been previously reported on the coincidence rate in a 4-inch diameter by $1\frac{1}{2}$ -inch NaI crystal in either of two geometries: 1) coincidence detector outside the shielding in a plane covering only the top surface, as in the experiment above, and 2) coincidence GM tubes within the main shield arranged in an inverted U covering the top and sides of the crystal detector. Experimental details are discussed at greater length in references (1) and (2). Figure 38 illustrates the essentials of each geometry. The differences observed between the two geometries were within the statistical and instrumental uncertainties of the equipment (Table 17). It has been thought advisable to obtain similar data on a larger crystal (5- by 4-inch). The higher counting rate, together with the greater stability of energy calibration inherent in the pulse-height-to-digital type of analyzer, results in a significant reduction of statistical uncertainties. There now seems to be an observable difference in the two geometries with the larger crystal. Table 17 shows the pertinent results, together with a summary of previously-reported data. Most of the error associated with a given counting rate is due to uncertainties in the calibration at the low energy end of the spectrum, which supplies most of the counts. Also,

Fig. 38

Table 17

Geometry	Shield	Energy Range	Coincidence counting rate, counts/min
<u>4-inch diameter by 1½ inch crystal</u>			
1 ^a	Small Fe cave	65-830 kev	23.4 ^b
1 ^a	Small Fe cave	65-1.60 Mev	27.2
2	Room II	65-830 kev	24.5 ^c
2	Room II	65-1.60 Mev	28.0
<u>5-inch diameter by 4 inch crystal</u>			
1	Small Fe cave	65-830 kev	68.2
1	Small Fe cave	65-1.60 Mev	79.4
2	Room II	65-830 kev	57.0
2	Room II	65-1.60 Mev	67.0

^aShielded by an additional 1 inch of Hg inside the coincidence detector.

^bMean of two runs, with 10 μ sec and 100 μ sec resolving times.

^cMean of two 1000-minute runs.

the data were taken in three different shields of quite similar but not identical attenuation characteristics. For these reasons, no probable errors are shown. It is believed that all figures are good to within 10%.

The random coincidence rate will be higher with the coincidence detector outside the main shield (geometry 1) because of the higher counting rate of this detector, but the effect is far too small to explain the observed difference. We believe the difference to be real and to suggest that the primary charged particles of cosmic origin, supposedly detected outside with about 100% efficiency, generate showers of high energy secondaries which are not detected with comparable efficiency by the anticoincidence ring when inside.

References

1. C. E. Miller, Report of Biological, Medical and Biophysics Programs. Part II Semiannual Report of Radiological Physics Division, ANL-5456, p. 109 (July, 1955).
2. H. A. May, Radiological Physics Division Semiannual Report, ANL-5679, p. 73 (February, 1957).

BACKGROUND OF IRON ROOM II

H. A. May

The extended analysis of the natural background within the second Iron Room presented in the preceding semiannual report⁽¹⁾ established that some contaminated material was present. This was readily proven to be in the 1/8-inch lead lining. When this was replaced by a sheet of equal thickness from another source known to be free from radium, the observed background counting rate dropped to a value in agreement with that obtained in Room I.

Because of the pressing nature of other work, we have not made a direct comparison with the 8-inch by 4-inch crystal as in the previously reported work. The equivalence in background count of the two rooms may be inferred by means of less direct measurements, however. Two independent sets of experimental counts show this equivalence and also demonstrate the consistency of results obtained. They thus establish the validity of the various counts involved.

Method 1: A sixty-minute background run was taken in Room I simultaneously with two crystals, size 8- by 4-inch and 9½ by 5-inch. These were separated by approximately two meters, so that the contribution from the potassium-40 and radium in each crystal and phototube to the counting rate of the other would be minimized. The counting rate of the 8- by 4-inch crystal was equal, within statistics, to others taken alone, proving that this effect was not detectable. A 50-minute count in Room II with the 9½- by 5-inch crystal alone resulted in the same observed count, within limits of detection. The exact figures are:

Room I	8- by 4-inch crystal	953 ± 6 c/m*	between 25 and 1575 kev
Room I	9½ by 5-inch crystal	1833 ± 8 c/m	between 25 and 1575 kev
Room II	9½ by 5-inch crystal	1854 ± 10 c/m	between 25 and 1575 kev

*0.9 error

The difference in the last two lines, of the order of 1%, may easily have been caused by residual contamination from build-up of radon daughters in Room II since there was no forced ventilation at the time.

Method 2: In the early phases of testing the 9½-inch by 5-inch crystal, a background determination in Room II with the original contaminated lead lining in place was made. At this time, the crystal was viewed by six 3-inch photomultiplier tubes. A later run with the clean lead lining in place, already referred to above, was made, this time with only five tubes attached to the crystal. The different arrangement was not intentional;

the background runs were actually rather incidental to the testing of various phototube arrays. As a further complication, we have no direct measurement of the result of removing a single 3-inch tube. This was calculated from data taken with a 5-inch by 4-inch crystal and comparison of counting rates of the two crystal sizes with an extended K^{40} source. It should be noted in passing that the contribution of a photomultiplier tube should not be exactly the same if counted in contact with a crystal, but on the opposite face from the phototube supplying the signal, as if optically coupled to the crystal window and forming part of the signal circuit because the collection efficiency of the low energy photons may be expected to differ in the two cases. This is admittedly a rather small effect. The pertinent figures over the energy range previously mentioned, are below. Since the effect of the lead shielding is essentially confined to the energy region below 650 kev (see reference 1, Figure 38, page 148), data within this band would reveal the same difference count.

Room II Old lead, $9\frac{1}{2}$ by 5-inch crystal, viewed by 6 tubes	2270 $\pm$ 7 c/m
Contribution of one 3-inch photo- multiplier and base, less socket as counted by 5-by 4-inch crystal	57 $\pm$ 6 c/m
Same, calculated for $9\frac{1}{2}$ by 5-inch crystal	150 $\pm$ 15 c/m
Room II Old lead, equivalent rate with 5 tubes	2120 $\pm$ 16 c/m
New lead, $9\frac{1}{2}$ by 5-inch crystal, viewed by 5 tubes	1854 $\pm$ 10 c/m
Net reduction, due to clean lead	266 $\pm$ 17 c/m

From this reduction, we now calculate what the difference would have been if the 8-inch by 4-inch crystal had been used. This ratio is calculated, as before, by a comparison of calibration data using K^{40} and radium sources. The relative response ratio, averaged over the energy band, is about 1.3:1. Thus we have:

Net reduction, equivalent, for 8- by 4-inch tube	203 $\pm$ 13 c/m
---	------------------

This is to be compared with an observed difference between Rooms I and II of 197 c/m as reported by Miller - the difference in the totals of columns 4 and 5, Table 41, reference 1.

Reference

1. C. E. Miller, Radiological Physics Division Semiannual Report, ANL-5829, p. 147 (February, 1958).

EVALUATION OF RCA 5-INCH TUBE, TYPE 7046

H. A. May

An RCA Type 7046 photomultiplier tube was obtained for evaluation. This is a 14-stage tube, designed to have good electron collection efficiency and short transit-time spread. The faceplate, of Corning 9741 ultraviolet-transmitting glass, features a curved photocathode and flat outer surface, resulting in a rather massive plate. The tube is inconveniently large, but it was hoped that this might be outweighed by a possible reduction in radioactivity due to the Kovar metal shell and absence of a conventional bakelite plug-in base. The tube, with faceplate, was placed against an 8-inch by 4-inch crystal, separated only by a 1/2 inch Lucite beta-shield. A net increase of 571 counts per minute, with a typical radium spectrum, was observed.

A separate faceplate was obtained* and counted with a 5-inch by 4-inch crystal, yielding a net count of 230 counts per minute, representing a total radium content of 2×10^{-10} curies. The potassium-40 content was below the detection level.

Since the faceplate activity greatly limits the usefulness of this tube in low-level counting applications, only a cursory test of its electrical characteristics was made. It was optically coupled to a 4-inch by 1 1/2-inch crystal and operated with an anode load of 20,000 ohms and applied potentials varying from 1500 to 3000 volts. The best resolution was 12 1/2 % for Cs^{137} at a potential of 1800 volts, slightly better than that attained with an unselected DuMont 6364. Photocathode uniformity was not investigated.

*We acknowledge the interest and assistance of Dr. R. W. Engstrom and Mr. J. L. Weaver of the Tube Division, RCA, Lancaster, Pennsylvania.

OPTIMUM PERFORMANCE FROM LARGE NaI(Tl) CRYSTALS

H. A. May

A great deal of experience with various methods of crystal mounting, reflector materials, light pipes, etc. obtained with crystal sizes up to 3 or 4 inches in diameter is summarized in the literature. Optimum methods have thus been recognized, resulting in uniform results among the many laboratories and workers employing crystal spectrometers of this size. Resolutions of 8 to 9% for cesium-137 are routinely obtained. Most of the credit should be given to the manufacturers of crystals and photomultiplier tubes for the improvement in the uniformity of their products attained in the last few years.

The number of crystals in use with diameters of 8 inches or more is very limited, as is experience with methods of obtaining the best results from them. Davis, Bell, Kelley and Lazar⁽¹⁾ have reported on results obtained with three 5-inch tubes coupled to a $9\frac{1}{4}$ -inch diameter by 9-inch long truncated-cone single crystal. The resolution was 11.8% with a source located in a well at the center of the crystal, and about 15% with a collimated external source. Koch and Foote⁽²⁾ at the Bureau of Standards obtained a resolution of 11% with a 5-inch diameter by 8-inch long composite crystal viewed by eight 2-inch tubes located on both end faces.

A program was undertaken to investigate the resolution of a new $9\frac{1}{2} \times 5$ -inch ANL canned Harshaw ingot, hereafter referred to as crystal "A," with various light collecting systems. There was realization at the outset that one cannot, in the present state of the art, expect every crystal of such dimensions to have a uniformly high performance. The problems of separately evaluating the effects of crystal inhomogeneity, self-absorption of the scintillation quanta, and poor reflector surfaces became apparent as the work progressed. This section presents some of the results obtained.

When the photocathode area is less than that of the crystal window, it is essential that a reflector be placed over the remainder. Teflon, of a selected pure white grade, optically coupled to the crystal window with DC-200 silicone fluid,* is excellent for this purpose. Figure 39 shows an assembly typical of those used for all measurements and found to be quick and convenient. For temporary assembly, the $1\frac{1}{2}$ -inch thick Teflon plate is greased to the crystal face with fluid of 10^6 centistoke viscosity. A temporary clamping plate held against the Teflon with a few C-clamps for a short time speeds the distribution of the thick oil and assures uniform optical contact. The lower part of each tube well, shown in detail, is then filled with 1000 centistoke DC fluid* and the tube lowered into place. If the

*Dow-Corning DC-200 silicone fluid.

Fig. 39

tube is tilted slightly at first, and some excess oil displaced, no bubbles are formed. A spun mu-metal shield is then dropped down over each tube and pressed tightly into the well against the upper ledge. This gives mechanical protection and light shielding for each tube. Since the Teflon is not perfectly opaque, several layers of dark cloth are draped around the tube and electronic preamplifier assembly, and as an extra precaution, a cardboard shipping drum placed over that. The latter measure proved necessary only when operating in a well-lighted location. Since no mechanical clamping is employed, use of this arrangement is limited to the upright position; but as a means of obtaining relative data which are readily reproducible without spending a lot of time in set-up, it is found very satisfactory.

When multiple tubes are used, the tube gains must be closely matched. The degree of matching depends somewhat upon the crystal size and energy of the gamma rays to be detected since these factors are the major ones influencing the spatial distribution of light flux at the crystal window. Six Dumont 6363 tubes were selected from a lot for uniformly high gain and best resolution. The resolution of each one, attached to an excellent $2\frac{1}{2}$ -inch by $2\frac{1}{8}$ -inch crystal, fell between 8.5% and 9%. By plotting the log of the relative gain versus applied voltage, any difference in photocathode efficiency is manifest as a change in slope. The tubes selected had equal efficiencies, a total spread in gain of less than $\pm 20\%$ of the mean, and required an average increase of 86 volts in operating potential to double their gain over the required operating range. The circuit employed a common anode load of one megohm and a positive high voltage supply. Although the string of dividing resistors were all of the same nominal value, the inevitable differences in dynode voltages, due to the use of 10% tolerance

resistors, introduced additional differences in tube gain. To avoid fluctuations due to this source, each tube was always plugged into the same socket. The gain of each tube-socket combination was equalized by selection of a dropping resistor in series with the divider string. In addition, individual potentiometers permitted variations in the focusing electrode potential over a small portion of the total cathode-to-first dynode range, as fine gain adjustments. Although this undoubtedly affects the uniformity of photoelectron collection to some extent, its effect on the over-all resolution is negligible. When the tubes were carefully matched, one could vary the high voltage to give a gain change by a factor of 2 or 3 without any differences in gain among the tubes.

Crystal A was first tested with a single 5-inch tube, also selected on the basis of high gain and good resolution. An uncollimated Cs^{137} source located on the crystal axis within a few centimeters of the $1/2$ -inch Lucite beta-shield was employed for all tests, unless otherwise noted. A resolution of 26.6% was obtained. From this, as well as from the value of the high voltage required, it was apparent that very little of the light released within the crystal was being collected at the phototube. The assembly of six 3-inch tubes previously described was next tried, improving the resolution to 14.5%. The pulse heights from each of the five peripheral tubes were equal within a few percent, whereas that of the center tube was at least 25% higher. When the center tube was replaced by a Teflon reflector, a resolution of 12.8% resulted. The five outer tubes were then shifted toward the center by $1/2$ inch, a second Teflon plate being used. The total light collected, as determined by a voltage reduction of 17 volts required to maintain the same pulse height, was increased by 19%, but unexpectedly, the resolution fell to 14%. These results were tentatively interpreted as evidence of inhomogeneity within the crystal. Further tests of the crystal are under way by the maker in an attempt to help explain the remarkable dependence of resolution upon detector geometry.

While this work was in progress, a second large ($9\frac{3}{8} \times 6$ -inch) crystal with a 45° bevel over 1 inch of the end was borrowed from Harshaw for evaluation and comparison.* It is hereafter referred to as crystal "B." It appeared clearer optically, with its reflecting surfaces more uniform and lacking tint. This superior visual appearance was confirmed by the results obtained when coupled to various phototube assemblies. A limited number of comparative tests were possible during the short period available, and these are summarized in Table 18. The few measurements of resolution made at higher energies are also shown. Had we anticipated the interest shown by others in results at high energies, where the improvement in large crystals should be most noticeable, we would certainly have extended this part of the work.

*Our indebtedness to Dr. E. C. Stewart for the opportunity of testing this crystal is hereby acknowledged.

Table 18

RESOLUTION vs. TUBE AND SOURCE GEOMETRY

A. $9\frac{1}{2}$ x 5 inch, ANL canned, .082" walls and .010" window, #304 stainless steel.B. $9\frac{3}{8}$ x 6 inch, beveled, in .040" copper, Harshaw type B assembly.

Source Geometry	One 5" PM tube		Five 3" tubes on 3-1/4" radius.		Five 3" tubes on 3-1/2" radius, plus one on center		Five 3" tubes on 2-3/4" radius.
source (s) at x	A	B	A	B	A	B	A
26.6%	12.2%	13%	10%	14.5%	9.5%	14.5%
27%	12.1%	10%		10.2%		16%
	peak shift =						
	-4%	+1%	-.25%		-.25%		-.75%
29%	15.5%	11.5%		10%		15.6%
	peak shift =						
	-9.5%	-1.5%	-2.7%		-2.5%		-4.2%
1.46 Mev	9.8%	10.2%		12.6%		
	2.62 Mev	9.5%	7.5%				

The results obtained with multiple tubes coupled to crystal B clearly demonstrated the importance of equalizing tube gains. Once adjusted, there were no observable shift with changes in applied voltage, and results were readily repeated over periods of several hours. We have no direct evidence of longer-term stability. The best Cs^{137} spectrum obtained with six tubes is shown in Fig. 40; it has been normalized and the energy axis adjusted to facilitate comparison with similar spectra in the Scintillation Spectrometry Catalog.⁽³⁾ The major difference noted is the reduced peak-to-valley ratio of the large crystal, (14.5 vs 40) due to the relatively large amount of scattering material present. This consisted of 560 mg/cm² of Lucite (the safety container for the Cs source), plus 450 mg/cm² of copper in the crystal can. A peak-to-total ratio of 0.54 is observed.

It is believed these results are convincing evidence that there are no fundamental reasons for large crystal performance to be inferior to that now readily obtainable from 3- x 3-inch and 4- x 4-inch sizes, at least over the energy range of interest in whole-body counting.

Fig. 40

Comments and Analysis

By utilizing the relative gain *vs* voltage calibration of the phototubes, we may calculate the relative number of photons reaching the photocathode surface, or the relative light output, of each crystal for a given geometry. This is shown in Table 19 for two cases, but a comparison for the six-tube geometry is not included since the anomalous results referred to above make it of questionable significance.

Table 19

Geometry	Relative light output (Crystal B: Crystal A)
One 5-inch tube	1.55:1
Five 3-inch tubes on 3 $\frac{1}{2}$ -inch radius	1.86:1
"Bounce" experiment	1.95:1
Mean value	1.8:1

In a separate experiment designed to measure relative light transmission, scintillations from a $2\frac{1}{2}$ " x $2\frac{1}{8}$ " crystal were "bounced" through each large crystal and collected by a single 3-inch tube diametrically placed. The setup is depicted in Fig. 41. That portion of the crystal window not coupled to crystal or tube had no reflector. The efficiency, or ratio of light collected to that collected when the crystal and tube were coupled directly, is calculated to be of the order of 5%. Because of gamma-ray transmission through the 1-inch Pb walls of the collimator, a spurious spectrum is obtained from the large crystal in addition to the optically coupled spectrum of the smaller crystal. When this directly transmitted background was subtracted, a clearly defined photopeak remained. The height (channel number) of this peak was averaged over several different angular orientations. The comparison in Table 19 shows reasonable agreement between these rather dissimilar methods of measurement.

Fig. 41

The resolution is seen to improve with number of phototubes employed. It might be supposed that this is due solely to the increased number of photons escaping from the crystal to produce photoelectrons somewhere on one of the tubes. That this is not the case may be proven from the following observations. With six tubes coupled to crystal B, the voltage was adjusted to place the Cs peak in channel 237. With the center tube electrically disconnected but left in place, the photopeak dropped to channel 200. This change is within a percent or two of that to be expected on the basis of relative cathode areas. When the center tube was replaced by a Teflon reflector plug, the photopeak returned to channel 237. We regard this as a significant indication of the high optical quality of the crystal and satisfactory reflectivity of Teflon. The results with crystal A, though having a similar trend, were less consistent and demonstrated a marked dependence on the location of the tubes.

In an analysis of the factors contributing to the observed resolution of NaI crystals, Kelley, et al.,⁽⁴⁾ showed that for small crystals (1.25- x 0.5-inch and 3- x 3-inch) the total observed line width may be approximated by:

$$R_{\text{Total}}^2 = R_i^2 + R_{\text{tube}}^2 \quad (1)$$

The term R_i is a constant of the scintillator material, known as the "intrinsic crystal resolution." It was shown to be about 6% at an energy of 660 kev. The term R_{tube} is determined by the statistical nature of the photoelectron release and multiplication processes within the tube, i.e., R_{tube}^2 is proportional to $1/\bar{n}$, where $\bar{n}$ is the average number of photoelectrons per pulse collected at the first dynode. This term was evaluated by light pulser methods and shown to be of the order of 4% for the best 3 tubes at a pulse height equivalent to that of the Cs^{137} photopeak. This value has recently been confirmed by Hickok and Draper⁽⁵⁾ who also reported a mean value of 5.3% for 4 Type 6364 five-inch tubes.

As crystal size increases, the useful light output per scintillation event is reduced for several reasons, principally as a result of some sort of crystal inhomogeneity, self-absorption of light, and losses at reflector surfaces. In addition, if the photocathode area is reduced relative to the total surface area of the crystal, there result a greater average internal path length and a greater number of reflections before light reaches the photocathode. These additional handicaps may not easily be offset by larger photocathodes because photomultipliers presently available show a rather rapid decrease in cathode uniformity and photoelectron collection efficiency vs the size increase. The literature contains little information as to the magnitude of these effects. Hickok and Draper found that the RMS variation in photocathode yield was from 27% to 63%, with a mean of 41%, for the 5-inch tubes mentioned above. In their experiment, the shield or "focus" electrode potential was adjusted to give maximum pulse height. Our own experience indicates that this potential distribution does not necessarily result in the most uniform response. For optimum results, it is probably necessary to determine each tube response individually.

The effect of these factors in reducing the number of photoelectrons per pulse might be accounted for individually by a redefinition of the last term in equation (1) by setting

$$R_{\text{tube}}^2 = \frac{1}{N_0 \epsilon \gamma n_p} \quad (2)$$

where

N_0 = mean number of photons generated by the scintillation per incident gamma photon,

ϵ = efficiency of optical coupling,

γ = photocathode quantum efficiency,

n_p = fraction of photoelectrons reaching the first dynode.*

This expression is of limited practical value because of the difficulty in separately evaluating each factor. Lacking the idealized perfect crystal and unity optical coupling, we chose to compare each crystal-detector arrangement with a "standard" small crystal of good resolution by means of the definition:

$$R^2_{\text{tube}} = \frac{1}{\bar{n}} = \frac{1}{\beta \bar{n}_s} \quad (3)$$

where β is the over-all efficiency relative to the standard crystal, which has a mean photoelectron yield in a selected 3-inch tube, of $\bar{n}_s$. The value of β may be determined with some precision when comparing systems employing the same phototube, but greater uncertainty results when inter-comparing single vs multiple-tube systems. In the present study, the standard chosen was crystal X-110, size $2\frac{1}{2}$ - x $2\frac{1}{8}$ -inch, and β is believed known to $\pm 10\%$. The quantity $\bar{n}_s$ calculated by equation (1) from the observed resolution of 8.5%, when coupled to the median of the size 3-inch tubes used, was 275. The data of Hickok and Draper, using a light pulser, indicate a mean value of 670, while a 4% tube resolution required a value of 620. Hofstadter⁽⁷⁾ states that 1 to 1.5 photoelectrons per kev were released in a 5819 tube coupled to a small NaI fragment, implying a value between 600 and 900. It appears that the standard system chosen is somewhat less than the ideal.

Values of β for other systems were calculated from the ratios of observed relative pulse heights and known tube gain data. Analysis of the observed resolution reveals that the increase in line width is much greater than that predicted by Poisson statistics applied to the reduced value of $\bar{n}$. This nonstatistical distribution in the actual number of electrons collected per pulse may be taken into account by adding a third term R_g to equation (1), giving

$$R^2_{\text{obs}} = R^2_i + \frac{1}{\bar{n}} + R^2_g \quad (4)$$

The added term may be thought of as a measurement of the quality or "goodness" of a particular crystal-detector geometry.

*The terminology is due to Morton.⁽⁶⁾

Table 20 lists the observed and calculated quantities discussed above, for the systems listed in Table 18, plus a few others readily available in our laboratory. For the large crystals, the R_g term is seen to change value far more rapidly than might be expected from the changes in collector geometry. This suggests spatial variations in crystal homogeneity, optical transmission and reflection. Clearly, an improvement in experimental methods and a more rigorous theoretical approach are needed if the performance of these large crystals is to be understood.

The author is pleased to acknowledge the capable assistance of Mr. Orville J. Steingraber in planning the program and taking many of the data.

Table 20

(All data with an uncollimated cesium-137 source approximately 2 cm from crystal face. R_i assumed to be 0.06.)

Crystal size, in.	Detector arrangement	R_{total}	β	$\bar{n}$	R_{tube}	R_g
$2\frac{1}{2} \times 2\frac{1}{8}$ (Std)	1 - 3"	0.085	1.0	275	0.06	0
4 x 5/8	1 - 5"	.103	1.0	275	.06	0.057
4 x $1\frac{1}{2}$	1 - 5"	.13	0.90	250	.063	0.096
5 x 4	1 - 5"	.12	.63	174	.076	0.071
$9\frac{3}{8} \times 6$	1 - 5"	.122	.50	137	.085	0.063
$9\frac{3}{8} \times 6$	5 - 3"	.10	.56	154	.081	0
$9\frac{3}{8} \times 6$	6 - 3"	.095	.56	154	.081	**
$9\frac{1}{2} \times 5$	1 - 5"	.266	.32	89	.105	0.236
$9\frac{1}{2} \times 5$	5 - 3" on $3\frac{1}{2}$ " dia	.13	.37	102	.099	.06
$9\frac{1}{2} \times 5$	6 - 3"	.15	.39	107	.097	.098
$9\frac{1}{2} \times 5$	5 - 3" on $2\frac{3}{4}$ " dia	.145	.43	119	.092	.095
$9\frac{1}{2} \times 5$	4 - 3" on $2\frac{1}{2}$ " dia	.157	.35	96	.102	.103

** R_g^2 is negative

References

1. R. C. Davis, P. R. Bell, G. G. Kelley, and N. Lazar, Response of "Total Absorption" Spectrometers, Transactions of Nuclear Sciences NS-3, No. 4, p. 82 (Proceedings of the Fifth Scintillation Counter Symposium) (November, 1956).
2. H. W. Koch and R. S. Foote, Total Absorption X-ray Spectrometry, Nucleonics 12, No. 3, p. 47 (March, 1954).
3. R. L. Heath, Scintillation Spectrometry Gamma-Ray Spectrum Catalog, IDO-16408 (Phillips Petroleum Co., Atomic Energy Division, Idaho Operations Office, Idaho Falls, Idaho) (July, 1957).
4. G. G. Kelley, P. R. Bell, R. C. Davis and N. Lazar, Intrinsic Scintillator Resolution, Transactions on Nuclear Science NS-3, No. 4, p. 57 (Proceedings of the Fifth Scintillation Counter Symposium) (November, 1956).
5. R. L. Hickok and J. E. Draper, Gamma Ray Resolution of Larger NaI Scintillators, Personal communication, submitted for publication in Rev. Sci. Instr.
6. G. A. Morton, Recent Developments in the Scintillation Counter Field, Transactions on Nuclear Science NS-3, No. 4 p. 122 (Proceedings of the Fifth Scintillation Counter Symposium) (November, 1956).
7. H. I. West, W. Meyerhof, and R. Hofstadter, Detection of X-rays by Means of NaI(Tl) Scintillation Counters, Phys. Rev. 81, p. 141 (1951).

THE EFFECT OF METEOROLOGICAL VARIABLES UPON THE VERTICAL AND TEMPORAL DISTRIBUTIONS OF ATMOSPHERIC RADON

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

I. Introduction

Ever since Elster and Geitel, in a paper written in 1901,⁽¹⁾ reported that the atmosphere contains radioactive substances, many investigators have measured the content of radon and radon daughters in the lower atmospheric layers. Much of this work was carried out by German investigators during the first half of this century, but within the last decade, interest in problems associated with radioactive fallout has given new impetus to the study of the problem in this country. The early work has treated many of the fundamental relations between the meteorological variables and the radon concentrations. Some types of data on radon concentrations are extremely rare. An example of this is data on hourly values of radon concentration and hourly vertical concentration gradients.

The measurements described in this report probably represent the most detailed measurements available showing the relation between the meteorological variables and the vertical distribution of radon with height and time. The facilities available to us, i.e., the instrumentation of the Meteorology Group and the techniques developed by A. F. Stehney and Henry F. Lucas, Jr., provide the wherewithal to fill in some of the gaps concerning this problem.

It is well known that the concentration of atmospheric radon at any location is a function of the following factors:

1. the emanation rate from the soil
2. the atmospheric diffusion rate
3. the advective rate, i.e., the rate at which radon is brought in over the station by the horizontal component of the wind
4. the radioactive decay rate.

The first three factors are markedly influenced by meteorological variables.

Much remains to be learned concerning the mechanisms which remove the radon from the soil. For example the effect of atmospheric pressure changes is considered important by some^(2,3) and discounted as unimportant by others.⁽⁴⁾ The effect of wind speed or soil temperature gradients is also in need of further investigation. It is commonly agreed that rainfall retards the emanation of radon by clogging the capillary pores in the soil, but insufficient data are available to establish quantitative relationships between rainfall or soil moisture and diminution of radon emanation.

The diffusion rate of radon in the atmosphere is primarily determined by the wind speed, atmospheric stability and surface roughness. An added complicating factor was suggested by H. Israel.⁽⁵⁾ He points out that radon may become adsorbed on dust particles, and therefore, the vertical distribution of radon would be influenced by particle pickup, particle fall velocity or sources of particles upwind. Further investigation is needed to provide answers to this problem.

Previous investigators⁽⁶⁾ have shown that the trajectory of the air influenced the radon concentration. However, the emphasis has been on the differences between air of continental and air of maritime origin. The data presented below indicate that substantial horizontal discontinuities may exist in air which has become stagnated in pockets during strong nocturnal terrestrial radiation and is suddenly flushed out.

II. Experimental Procedure

A series of radon concentration measurements at various heights was begun at 0900 CST on May 20, 1958 and continued through 1100 CST, May 21, 1958. Observations were begun 10 minutes before the hour and ended at 10 minutes past the hour, thereby obtaining 27 twenty-minute integrated samples of radon concentration for each level. The first observation, however, started at 0900 and ended at 0920. The measurements were made at four levels: 3 ft, 18.75 ft, 78 ft, and 131 ft above the ground. The air samples were obtained by drawing air through 1/4- and 3/8-inch dia. copper tubing attached to the Meteorology Tower and leading to an instrument shack located at the southeast leg. The radon in the 50- to 100-liter samples of air from each level was quantitatively adsorbed on charcoal cooled with a dry-ice bath using the system previously described.⁽⁷⁾ The volume of each sample was determined by means of wet test meters. The charcoal trap containing the radon was then removed from the sampling apparatus and taken to the radiochemistry laboratory of the Radiological Physics Division for analysis. The radon in each sample was transferred by heating and flushing with helium to an alpha scintillation radon counter.⁽⁸⁾ This system has a sensitivity of 5.46 ± 0.08 cpm/ $\mu\mu\text{c Rn}^{222}$, a reproducibility of $\pm 2\%$, and a background counting rate of about 0.1 cpm. Blank values were less than 10^{-14} c/sample. Counting rates of 16 to 300 cpm were observed which when counted for 50-60 min. result in a 90% confidence level of 2-5%.

III. Discussion of Data

In Fig. 42 is shown a plot of the radon data as a function of time and height. There are several striking features which these data exhibit. First of all, it is obvious that during the daylight hours the concentrations of the radon are quite small and the vertical gradients are very weak. However,

FIGURE 42
HOURLY VALUES OF RADON CONCENTRATION AT INDICATED LEVELS

METEOROLOGY TOWER
MAY 20-21, 1958

as the dark hours approach, the concentrations and the vertical gradients of radon concentration increase markedly. The amount of radon measured at three feet above the ground at night was nearly 20 times as much as that during the day. Also during the night, the concentrations at 3 ft were three or four times those at 131 ft but during the day the concentration at 3 ft seldom exceeded 1.5 times that which occurred at the 131-ft level.

A very interesting feature of this plot is the marked increase in the radon concentrations at all levels which occurred between 0100 and 0200. During this hour, the wind direction changed appreciably (see Fig. 43) thus indicating that the rise in concentrations may have been due to advection.

An examination of Fig. 44 in which the difference between the air temperature and soil temperature is plotted seems to suggest that possibly some of this increase may have been due to increased emanation from the soil because the air within the top layers of the soil became convectively unstable. This is an attractive possibility, but the work of soil scientists on soil aeration⁽⁹⁾ suggests that this was an unlikely cause.

Another interesting feature of Fig. 42 is the time at which the decrease of radon occurs at the various levels. It may be noted that at the lowermost level, the radon begins to fall at 0300 and falls continually on to the early forenoon hours. However, in the uppermost layers, the 131-ft level and 78-ft level, the sharp decrease does not occur at the same time but starts to fall markedly at 0600. This may suggest, but not prove, a reduction in the radon emanation rate near the soil.

Figure 45 shows the hourly values of stability for the following atmospheric layers: 5.5-15.2 ft, 5.5-34.0 ft, 5.5-72.2 ft, and 5.5-144 ft. The values plotted in Fig. 45 represent a 4-minute average of the temperature difference between the top and bottom of each of the above layers. The gross features of the radon concentration curves and stability curves correspond quite well. Note that at 1800 shortly after an inversion set in, the radon concentrations and gradients increased. Similarly at 0600 when the inversion was destroyed in the lower layers, the radon concentration gradients were markedly reduced. The sharp rise in radon concentration between 0100 and 0200 does not appear to be clearly related to changes in stability.

Twenty-minute averages of wind speed at 69 inches, 18.75, 37.5, 75 and 150 ft are shown in Fig. 46. The averaging period for wind speed measurements corresponds to that for the radon measurements. Figure 46 shows strikingly the diurnal variation in wind speed. In the evening the greatest change takes place between 1800 and 2000. This corresponds to the rapid change in stability. However, the greatest changes in radon concentration appear to occur during 1900-2100, i.e., a lag is evident in the buildup of radon. The sharp rise in radon at 0100-0200 corresponds to a drop in wind speed at the 150- and 75-ft levels, but additional data and analysis are necessary adequately to assess the effect of this wind speed change on the radon concentration. Also of interest is the sharp rise in speed at the upper levels at 0200-0400, which corresponds to the increase in stability.

Hourly values of soil temperature and 20-minute averages of air temperature are shown in Figs. 47 and 48. It may be noted that from 0100 to 0200 a sharp drop in temperature (3.7°C) occurred to correspond to the rise in radon. It is of interest that the temperature at 144 ft dropped even more, i.e., 4.8°C , thus reducing the inversion by 1.1°C . At the 18.75- and 37.5-ft levels, the temperature did not drop quite as much as at the surface. The curves showing the hourly values of soil temperature from 1 cm to 29 ft are quite smooth and do not depict the microchanges observed in the air.

The dewpoint temperatures for the two lower levels of 5.5 and 131 ft are plotted as a function of time in Fig. 49. Although the measurements obtained by the Dewcels have an error of $\pm 2^{\circ}\text{F}$, the gross features of the

FIGURE 45
HOURLY VALUES OF ATMOSPHERIC STABILITY AS DENOTED BY TEMPERATURE DIFFERENCE
BETWEEN TOP AND BOTTOM OF INDICATED LAYERS
METEOROLOGY TOWER - MAY 20-21, 1958

FIGURE 46
HOURLY VALUES OF WIND SPEED AT INDICATED LEVELS
METEOROLOGY TOWER - MAY 20-21, 1958

FIGURE 49
HOURLY VALUES OF DEW POINT TEMPERATURE AT INDICATED LEVELS
METEOROLOGY TOWER
MAY 20-21, 1958

changes are portrayed with sufficient accuracy. It is apparent that between 0100 and 0200 a marked rise in water vapor occurred to correspond with the rise in radon content. It is to be noted, however, that the general trend was toward an increase in moisture with time. Nevertheless, throughout this period, the air was quite dry thus contributing to marked terrestrial radiation at night and in turn to a strong nocturnal inversion.

Plots of solar radiation and radiation flux are shown in Figs 50 and 51. These data indicate the absence of clouds, strong incoming solar radiation effects during the day, and outgoing terrestrial radiation at night.

Pressure is plotted in Fig. 52. The changes in pressure during this period were sufficiently small as to have only a very minor effect on the radon emanation from the soil.

IV Comments on Measurements

1. These data strongly suggest that the distribution of radon concentration in the lower layers of the atmosphere is not horizontally homogeneous. During the night when conditions are stable the movement of masses of air may be appreciably slowed down in forested areas or bowl-shaped configuration of terrain. A change in wind direction may flush out such masses and result in marked increases in radon concentration. For

example the air movement through a rectangular forested area would be small when the wind was oriented along its long axis. Should the wind direction change to coincide with the short axis, the ventilation rate would increase, thus flushing out the air in which large amounts of radon had accumulated. It is likely that a phenomenon of this type resulted in the large increase in radon observed between 0100 and 0200.

2. The charts of the temperature difference or atmospheric stability charts and the radon concentration curves show a striking similarity. Although it is well known that radon concentrations increase under stable conditions, the data presented here probably represent the most detailed measurements of the temporal variations of radon and stability.

3. During the day, the concentrations of radon are quite low, as may be seen from Fig. 42. The values observed range from a little over $0.04 \mu\mu\text{c/liter}$ to slightly over $0.1 \mu\mu\text{c/liter}$. During the night, the highest radon concentration recorded at the 3-ft level was $0.96 \mu\mu\text{c/liter}$ while the highest at the 131-ft level was $0.45 \mu\mu\text{c/liter}$. The gradient between 3 and 131 ft during the daylight hours was of the order of $10^{-4} \mu\mu\text{c/liter/ft}$. During the night, the maximum gradient observed was $3.9 \times 10^{-3} \mu\mu\text{c/liter/ft}$. This represents a gradient during the night nearly 40 times as steep as that during the day. Of course, further data are necessary to establish how representative these figures are.

The help in carrying out the measurements rendered by Peter E. Wasko, Frank C. Kulhanek and Gunther A. Zerbe of the Meteorology Group and H. Ilcewicz of the Radiochemistry Group is hereby acknowledged. Thanks are also due to Martha C. Swanson for her part in preparing the figures and carrying out numerous calculations connected with these studies.

References

1. Elster, J., und Geitel, H., "Über eine fernere Analogie in dem elektrischen Verhalten der natürlichen und der durch Becquerelstrahlen abnorm leitend gemachten Luft." *Phys. Z.*, 2:590-593 (1901)
2. Lester, O. C., "A Rapid and Accurate Method of Correcting Measurements with Emanation Electroscopes for Ordinary Changes in Temperature and Pressure." *J. Opt. Soc. Amer.*, 11:637-640 (1925)
3. Wright, J. R., and Smith, O. F., "The Variation with Meteorological Conditions of the Amount of Radium Emanation in the Atmosphere, in the Soil Gas, and in the Air Exhaled from the Surface of the Ground, at Manila." *Phys. Rev.*, (2) 5:459-482 (1915)
4. Baver, L. D., Soil Physics, 3rd edition, Page 210, John Wiley & Sons, Inc., New York, N. Y., 1956
5. Israel, H., "Radioactivity of the Atmosphere," pages 155-161, Compendium of Meteorology, American Meteorological Society, 1951
6. Becker, F., "Messungen des Emanationsgehaltes der Luft in Frankfurt a.M. und am Taunus-Observatorium." *Beitr. Geophys.*, 42:365-384 (1934)
7. Lucas, H. F., Jr., F. I. Ilcewicz, and A. F. Stehney, Radiological Physics Division Semiannual Report, ANL-5679, p. 10 (1956)
8. Lucas, H. F., Jr., "Improved Low-Level Alpha Scintillation Counter for Radon," *Rev. Sci. Instr.* 28, 9:680-683 (1957)
9. Baver, L. D., Soil Physics, 3rd edition, Page 209, John Wiley & Sons, Inc., New York, N. Y., 1956

ARGONNE METEOROLOGICAL MODEL TOWING TANK

P. Frenzen

Introduction

A series of turbulent diffusion experiments in both neutrally and stably stratified fluids were carried out in the Argonne Meteorological Model Towing Tank during the period covered by this report. The principal effects of stability variation on the turbulent field which were observed in these runs may be summarized as follows:

- a) the turbulent energies (mean square velocity fluctuations) measured in the stable cases averaged about half those of the neutral cases; the ratio of the energies of the horizontal and vertical components (an index of isotropy) remained approximately the same in the two types of stratification;
- b) the time required for the vertical Lagrangian autocorrelation function to fall to zero in the stable fluid was found to be less than half of that required for the same component in the neutrally stratified fluid; no significant differences in the horizontal autocorrelation functions' "times to zero" could be detected;
- c) in the neutrally stratified fluid, the ratio of the horizontal and vertical diffusion coefficients was 1.5; in the stable fluid, the ratio was observed to increase to 4.2.

All experiments were carried out in relative mean flow velocities of about 25 cm/sec in the lee of a 3/8-inch bar, 2-inch mesh rectangular grid.

Continuing modification of apparatus and experimental method have reduced the residual mean flow components and increased the time that individual tracer particles remain under observation to twenty-four seconds. Digital computation of the mean flows, the rates of turbulent decay, and the Lagrangian autocorrelation functions have been employed, not only in the determination of the results quoted above, but also in the evaluation of the effects of the several modifications in the apparatus as they were made. At the end of the report period, a preliminary procedure for calculating power spectra of turbulent energies from the autocorrelation functions was being tried out on the analog computer, PACE.

Apparatus

After a number of trial runs under the five-foot long, moving upper lid described in the previous reports⁽¹⁻³⁾ of this series (but carried out twice as far beyond the grid as the limited experiment illustrated in Fig. 72 of Reference 3) had revealed excessive residual, mean flow "surge" components ranging from minus one to plus four millimeters per second, a semi-rigid lid covering ninety-five percent of the free water surface was installed. The only openings left were three-quarter-inch wide slits running the length of the tank along each side; these were provided for two narrow struts which suspend the grid below the lid and carry it through the fluid when the carriage is driven overhead.

The lid itself consists of one five- and two ten-foot long aluminum trays, twenty-five inches wide and three inches deep, which are floated on the towing tank's water surface, along with a five-foot long transparent plastic unit that admits light to the test section. When firmly shimmed and clamped end-to-end, the trays constitute a semirigid lid which damps all but the longest wave components in the tank.

The mean, positive "drift" about which the surging components had been observed to oscillate was reduced to one or two millimeters per second by extending the ends of the grid bars, reducing their previous one-inch clearance at the walls, floor, and lid to one-quarter of an inch, thus decreasing the net flow by restricting the space through which the fluid could return.

Vertical mean flow components associated with the horizontal surging appear to be held to a minimum by the presence of the lid. However, changing tracer-particle buoyancies appear in the analysis as effective mean flows in this direction. An effort was made to begin each series of experiments with slowly sinking tracer (less than one millimeter per second) since the mixture tends to become lighter with time. When, in later runs, the tracer began to rise more rapidly than one millimeter per second, the nitrobenzene-olive oil ratio was readjusted.

Ten-second photographs were taken of the undisturbed tracer distribution before each run. These showed a two-to-one variation in vertical tracer velocities (order of 0.5 to 1.0 mm/s) even in the same sample. The variation is probably due to varying amounts of occluded water within individual oil bubbles and differing drag forces associated with tracer particles of different diameters.

As will be seen in the discussion of the analysis below, it is difficult to compensate for these mean flow effects, especially when they are not constant from one single-particle run to the next. It appears that every effort should be made to eliminate them in the experiment itself by, for

example, stiffening the lid, reducing still further the clearance between grid and tank walls, and improving control over both tracer density and bubble diameter.

Experimental Procedure

A change in the experimental procedure was made in order to avoid a common misinterpretation of G. I. Taylor's statistical theory of turbulent diffusion pointed out by Frank Gifford⁽⁴⁾ Meteorologist in Charge, U. S. Weather Bureau Office, Oak Ridge. Gifford indicated that in many investigations the statistical averaging was not carried out over an ensemble of one-particle diffusion experiments as Taylor intended it should have been, but rather over a group of particles released at one time.

The point concerns the possible lack of statistical independence in the trajectories of groups of particles in the same turbulent field; motions of all particles within a given volume will be influenced largely to the same degree by all turbulence elements larger than that volume. Comparison of the trajectories of neighboring particles observed in the same experiment indicated that this effect did indeed operate, at least on particles separated by not more than a few centimeters in the relatively small scale of turbulence generated in the Argonne tank. For this reason, the experiments were changed so that instead of following many particles in a few experiments, either only one or only a few widely separated trajectories were selected for analysis from each run.

Solenoid circuits were changed so that the camera was held open for the entire period of the experiment, and a new rotating shutter with one large and seventeen small holes at twenty-degree intervals was installed on the one-second timing shaft. This combination resulted in photographs showing trajectories as finely-dashed lines marked with brighter points at one-second intervals. Since entire trajectories are clearly marked on single photographs, measurement of turbulent velocity components can proceed directly without the intermediate operation of identifying individual streaks with their particular trajectories. However, a number of successive experimental runs are now required where one sufficed before.

Experiments consisting of several consecutive rather than single runs necessitate closer control of experimental conditions in order that data obtained in such a series can be legitimately grouped in a statistical ensemble. To this end, a fine speed control was installed on the drive mechanism, enabling more accurate reproduction of mean flow conditions from run to run. In addition, an effort was made to start the carriage from rest at the beginning of each run in the same, smooth manner. Thirty

minutes were allowed between runs for the disturbances in the water to quiet down. Operation of the camera shutter was started by the passage of the carriage over a microswitch; since the carriage speed remained essentially constant, consecutive particle velocities measured along individual trajectories from different runs corresponded to similar consecutive positions relative to the grid and therefore were in similar states of decay. Thus the $(t^{\frac{1}{2}})$ decay correction programmed for the computer could still be applied to the ensemble.

Stable profiles were created in the tank thermally, though it had been thought that it would be necessary to carry out runs of this kind in stratified salt solutions. It was found that by half filling the tank with warm water, letting it stand until quiet, and then slowly slipping cold water in on the bottom, temperature differences of ten or twelve degrees Centigrade could readily be created between the top and bottom of the fourteen-inch deep tank. The profiles observed were "s" shaped, i.e., steep and practically linear in the central regions but approaching isothermal distributions near the upper and lower boundaries. In the series of stable runs providing the results summarized in this report, vertical temperature gradients ($d\theta/dz$) ranging from +0.6 to +0.3 deg C per cm were observed at the level of the center of the test section.

Temperature measurements were made with an array of five mercury-in-glass thermometers, fixed at two-inch intervals of depth, one-half inch from the glass wall near the test section, positions that barely cleared the moving grid bars during a run. They were calibrated in tenth degree C divisions, but readings were estimated to the nearest one hundredth. In an effort to assess the amount of mixing, and hence, the amount of additional work done against the stable density stratification during the decay of each realization of the field of turbulence, lapse rate measurements were taken just preceding and just after each experimental run. But because of the high lag of the mercury-in-glass thermometers (estimated to be at least three or four seconds), and because of the absence of any provision for horizontal space averaging, not to mention the close proximity of the thermometers to the tank wall, the results obtained were inconclusive.

Experimental Conditions

Results presented in this report are based upon the analysis of two groups of "single-particle" diffusion runs, one in neutrally stratified (isothermal) and the other in stably stratified ($\frac{\partial \theta}{\partial z} > 0$) water. The appellation "single-particle" does not strictly apply, for from one to four trajectories were selected from each run; but where more than one trajectory has been used, they have been selected from different parts of the test area photograph in order to approach more or less the requirement of statistical independence on the turbulent trajectories.

There were twenty-four neutral fluid trajectories taken from fifteen runs carried out on two separate days, no more than two from any one run. The runs lasted from seventeen to twenty seconds, twenty-one of them reaching the nineteen-second mark. Since timing for decay purposes necessarily begins from a virtual origin at which the intensity of turbulence would have been infinite (about ten mesh lengths behind the grid in these experiments), the first velocity measured at twenty mesh lengths from the grid was considered to have occurred at $t = 2$ sec, i.e., two seconds after the virtual origin, moving about five mesh lengths per second, has passed. This point, however, appeared to be too close to the grid for homogeneous turbulence to have been fully established; therefore, the first velocity measured in all of the neutral runs was ignored in the autocorrelation computation. This visibly reduced the scatter of points along the curve of the correlation function.

Carriage speeds in the neutral runs averaged 25.4 cm per second, individual values never varying more than plus or minus 0.1 cm per second from that figure. Vertical temperature gradients were very close to isothermal, averaging +0.003 deg per cm; all measurements were within plus or minus 0.002 deg per cm of the mean.

Data for the stable fluid studies were taken from twenty-one trajectories, these selected from ten runs carried out again, on two different days. Four trajectories were taken from one especially good run, three from another, and no more than two each from the remainder. Duration of the runs ranged from seventeen to twenty-four seconds, fourteen of them lasting twenty seconds or more. In this experimental series, the camera was automatically started by the passage of the carriage as described earlier; the switch was set to locate accurately the relative position of the first velocity measurement in each run at thirty mesh lengths from the grid, assigning a time for the first decay correction of four seconds. Since this appeared to be far enough from the grid for the turbulence field to have become uniform, all velocities measured in the stable runs were used in computing the autocorrelations.

Carriage speeds were again within plus or minus 0.1 cm per second of an average speed of 25.4 cm per second. Because of eddy mixing during the runs as well as heat losses at the top and gains at the bottom, the strength of the stable vertical temperature gradients changed from one run to the next, decreasing almost 0.01 deg per cm every five minutes. In the first series, the gradient decreased from 0.50 deg per cm to 0.28 deg per cm between the initial and final runs; in the second, the gradients ranged from 0.62 deg per cm to 0.38 deg per cm. The average gradient, both per run and per trajectory, was 0.42 deg per cm. When more stable runs have been completed, it will be possible to collect and analyze runs with vertical temperature gradients more closely grouped than these in order to investigate not only the effect of stability as compared with neutral stratification, but also the relative effects of differing degrees of stability.

Analysis

The question of the influence of residual mean flow components upon the autocorrelation calculations was the subject of a series of digital computer experiments using the basic data obtained in the runs described above. Difficulty arises because the effects of such residual flows are progressively magnified at later times in the experiment by the velocity decay correction built into the computer program, each observation being multiplied by the square root of the time.

Computations were made in which the mean flow was considered to have been zero; in another trial, mean flows for each time were calculated by averaging all velocities observed at similar times in the different runs of a single set. The autocorrelation curves thus obtained were compared with a third computer run in which a fictitious constant mean flow, about twice as large as the individually calculated mean flows, was assumed. Comparison of the first two cases showed relatively small changes; employing the individually calculated means reduced the times required for the curves to fall to zero from 1.6 to 1.4 corrected* seconds for the correlation of horizontal components in a neutral fluid and from 1.4 to a little less than 1.3 seconds for the correlation of horizontal components in the stable fluid. In addition, the shapes of the curves were altered by these changes in the correlation analysis, becoming more concave (upward) in one case and less in the other.

Assuming a large, constant mean flow (4 mm/s) considerably flattened the autocorrelation curve, preventing it from reaching the $R = 0$ axis during the period of the experiment, never in fact allowing it to fall below $R = 0.4$. This bears some resemblance to the results of E. K. Webb⁽⁵⁾ who found that a positive, linear trend superimposed upon a fluctuating record caused the autocorrelation curve to level off at some value greater than zero. Here, of course, the $(t^{1/2})$ decay correction acting upon the more slowly decaying mean flow imposes a positive, "parabolic" trend.

Experimental Results

The principal results of the analysis of these neutral and stable experimental runs are presented in the four autocorrelation curves of Fig. 53. In each of these, the second of the trial treatments of the mean flow described previously was employed, i.e., mean flows for each time were approximated by the averages of the observations taken at that time.

*The time (ξ) axes of the autocorrelation curves are in units of time altered by the second Batchelor decay correction (cf. ANL-5829) re-defining times as the natural logarithm of the actual time units of the experiment.

FIGURE 53
LAGRANGIAN AUTOCORRELATION FUNCTIONS IN NEUTRALLY AND STABLY STRATIFIED WATER, DETERMINED
FROM DECAY-CORRECTED PARTICLE VELOCITIES MEASURED IN THE LEE OF A RECTANGULAR GRID; $\bar{U} \approx 25.2$ CM/SEC;
GRID BAR = 1CM, MESH = 5.1CM; $\theta/\theta_2 = +0.003^\circ$ PER CM (NEUTRAL) AND $+0.42^\circ$ PER CM (STABLE)

The curves are best compared through the medium of several characteristic parameters which are derived from them. Their values are listed in Table 21 where they may be conveniently compared; their significance and methods of calculation are described below.

$u^* = \left(\sqrt{i} \sqrt{u_i'^2} \right)$: a typical turbulent fluctuation (horizontal), corrected for decay. Standard deviations $\left(\sqrt{u_i'^2} \right)$ of the velocities observed for each time (i) along the several trajectories of each series are regularly computed to measure the rate of decay. When these are multiplied by the square root of the time, their average becomes a typical velocity fluctuation of the decay-corrected turbulence field. Its vertical counterpart is w^* .

u^{*2}/w^{*2} : the ratio of the average energies of the horizontal and vertical components of the decay-corrected turbulence field, an index of the degree of isotropy.

T_0 : the "time to zero," the value in corrected time units (ξ) of the autocorrelation curve's intercept on the $R(\xi) = 0$ axis.

$t_1 = \int_0^{T_0} R(\xi) d\xi$: the Lagrangian scale time, the integrated area

under the autocorrelation curve, approximated here by the assumption that the integral beyond T_0 is negligible.

$\ell_1 = u^* t_1$: the Lagrangian scale, a characteristic length analogous to Prandtl's mixing length since it can be considered a measure of the distance that an average particle moves before its original velocity is lost, i.e., until $R(\xi)$ falls to zero.

$K_x = u^{*2} t_1$: a virtual coefficient of (horizontal) diffusion, again analogous to similar coefficients defined in mixing-length theory. K_z is written for vertical diffusion.

K_x/K_z : the ratio of the horizontal and vertical diffusion coefficients.

Table 21

Series: Component	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)
	Fluctuations (mm/sec)		Relative energies	Scale times (ξ sec)		Scale lengths ℓ_1 (mm)		Diffusion coeffi- cients (mm ² /sec)		
	u^*	w^*		T_0	t_1	$u^* t_1$	$w^* t_1$	K_x	K_z	K_x/K_z
Neutral:										
Horizontal	11.7		1.52	1.40	0.5	5.8		67.0		1.49
Vertical		9.5		1.09	0.5		4.8		44.9	
Stable:										
Horizontal	8.0		1.53	1.26	0.5	4.0		33.5		4.19
Vertical		6.5		0.45	0.2		1.3		8.4	

With regard to the entries in the preceding table, it may be pointed out that even though the absolute magnitudes of individual turbulence characteristics have been influenced by uncompensated mean flows, their relative magnitudes obtained by comparing the characteristics of one series to those derived in the same way from the other are less subject to this source of uncertainty, provided only one assumes that the mean flow effects were more or less comparable in the two sets of experiments. In short, the fact that the first entry in column 3 above is about 1.5 is probably not as significant as the fact that the two entries in that column are practically the same.

Comparing the entries in the first two columns of Table 21 it may be seen that the turbulence generated in the neutral fluid had about twice the average energy (u^{*2}) of that generated in the stable fluid. Since the rates of decay in the two fluid conditions were not found significantly different, it appears that this difference in eddy energy may have prevailed even in the immediate vicinity of the grid; if this is the case, the difference in energies would be the result either of a less efficient initial generation of energy in the boundary layers of the grid bars themselves, or of a more rapid rate of decay in the wake regions close behind the grid.

The almost identical values in the third column, the ratios of horizontal to vertical turbulent energies, are surprising. Available measurements under neutral and stable conditions in atmospheric turbulence would

lead one to expect a marked shift toward anisotropy (increased u^2/w^2) in the stably stratified fluid. Indeed, the lack of isotropy in the neutral case is in itself surprising and since rectangular grids have often been shown to generate locally isotropic fields of turbulence, even dubious. In these experiments, this result is best considered as an indication of the relatively strong effect exerted by the larger, horizontal mean flow components upon the horizontal turbulence component evaluations. The available experimental data are consistent with this conclusion.

However, there is no reason to believe that a similar disparity between the magnitudes of horizontal and vertical mean flows should not also have prevailed in the stable fluid and that, therefore, this observation of fields of turbulence with approximately similar degrees of isotropy (or anisotropy) in both neutrally and stably stratified fluids, though unusual, may be still correct. If so, it might be associated with the absence of vertical shear in the mean velocity profile in the tank since this is a phenomenon present to a marked degree when anisotropic turbulence is observed in a stable atmosphere

The most significant measurement of "time to zero" is the fourth entry (stable fluid, vertical component) of column four. Because of the larger effect of uncompensated mean flows in the computation of the horizontal autocorrelation curves, tending to delay the intercept of the time axis, the other three values of T_0 are probably in reality more closely grouped than they appear here, that is, the two horizontal values should be reduced. Note that the three corresponding Lagrangian scale times, t_1 , in the fifth column are almost identical. (Actually they differ by as much as several hundredths of a second, but this was not considered significant in view of the uncertainty in locating the curves along their entire length as well as the inaccuracies involved in measuring the areas bounded by the curves.) The explanation for the fact that the scale times are identical when the "times to zero" are not can be seen in the shape of the curve of Fig. 53(a); the largest T_0 is associated with the most concave (upward) autocorrelation curve, these two characteristics in effect canceling each other as far as changing the area under the curve is concerned.

Webb⁽⁵⁾ has shown that the Lagrangian scale time of an exponential correlation curve indicates the frequency of a peak in the power spectrum at $1/(2\pi t_1)$. Insofar as the observed autocorrelation curves of these experiments may be considered to be of exponential form, this relation gives peaks at 0.32 cps for both of the components of the neutrally stratified runs and also the horizontal component of the stable runs. The shorter scale time measured in the stable runs' vertical component corresponds to a higher frequency peak, 0.80 cps. These estimates indicate relative damping of longer wave length turbulent components in the stably

stratified fluid, at least in the vertical spectrum. This point will be subject to further investigation when complete power spectra are obtained by the computation of cosine transforms of the autocorrelation curves. At the time of writing, this procedure was being worked out on Argonne Laboratory's analog computer, PACE.

Lagrangian scale lengths, ℓ_L , show the same similarity in the first three entries of columns 6 and 7 of Table 21, but the value determined for the vertical fluctuations in the stable fluid falls off by a factor of three or four. In the parlance of mixing-length theory, particles moving in the stably stratified fluid preserved their original vertical turbulent fluctuation velocities through distances only one third to one fourth as large as those traversed by particles in the neutrally stratified fluid.

Combining the fluctuation energies (u^2) and the scale lengths, we find the virtual diffusion coefficients of columns 8 and 9. Their separate values are sensitive to the two-to-one reduction in turbulence energies observed between the neutral and stable cases. However, the ratios of horizontal to vertical coefficients within each of the two experiments (column 10), eliminating the energy change effect between experiments, still show an increase by a factor of three in the stable fluid; this is due to the larger reduction of the vertical Lagrangian scale in that case.

Conclusions

It may be noted that the 1.4-second time to zero of the horizontal component of the neutral fluid autocorrelation function measured in these experiments is shorter than the 1.8-second time found in trials under similar conditions of fluid stratification which were reported in the previous report of this series (Fig. 73, ANL-5829). The difference illustrates the effect of the reduction in residual mean flows that was achieved by substituting the stationary, semirigid lid for the moving upper boundary "sled" of the earlier experiments.

The only other experimental measurements known to this writer to which these results may be compared are those determined by Vanoni and Brooks⁽⁶⁾ in a water tunnel. Working with neutrally stratified flow moving through a half-inch bar, two-inch mesh grid at about fifty centimeters per second, (these values giving a grid Reynolds number about twice that realized in the current Argonne runs, 25,000 as compared to 12,500), these investigators found "times to zero" of 1.5 and 1.3 seconds for the horizontal and vertical autocorrelation functions respectively. These are quite close to the values of 1.4 and 1.1 seconds listed in Table 21 for similar conditions of fluid stratification, even though Vanoni and Brooks used an entirely different method of minimizing the effects of

turbulent decay. The Lagrangian scale times, as far as they can be estimated from the curves published by Vanoni and Brooks, are also close to those found in the current study, appearing to be only about 0.1-second larger in the horizontal component's case and no more than a few hundredths of a second different in the vertical autocorrelation case.

Because the problem of residual mean flows does not yet appear to have been adequately solved, results in this report in the form of magnitudes of characteristic diffusion parameters must be viewed as preliminary estimates. Ratios of these parameters, where such treatment reduces the mean flow effect as described earlier, can be considered to carry greater weight.

In the continuing experimental program, further efforts to minimize the residual flows by stiffening the lid and reducing the clearance of the grid at the tank walls will be made. In addition, the experiments will be tried at a higher level of turbulence by increasing the carriage speed and/or using a grid with larger diameter bars. Note that the average corrected turbulent fluctuations of the order of 6 to 12 mm/sec in these experiments derive from observed velocities only one-fifth as large toward the end of the trajectories, twenty-five seconds after the passage of the virtual origin.

Steadier thermal stratification conditions will be needed in order to obtain a number of runs for each of several degrees of stability; for this purpose the top and bottom of the tank will be insulated and an external heat source and sink arrangement will be added. A temperature-measuring system capable of horizontal space averaging will also be required. With these improvements, the experimental comparison of the neutrally and stably stratified fluid cases can be continued and eventually expanded to include the case of unstable stratification.

Acknowledgements

Because of the statistical nature of the research program in the Argonne Meteorological Towing Tank, the cooperation of the Applied Mathematics Division in supplying digital computation facilities has been of invaluable aid; the problem would not be feasible without them. Special thanks are due Mrs. Cynthia Chamot of that division whose interest in and efforts in behalf of the program have been largely responsible for the quality and quantity of analyzed data obtained to date.

Similarly, data reduction could not have proceeded with anything approaching the accomplished speed without the use of digital coordinate readout equipment extended by the Cloud Physics Laboratory, University of Chicago, and more recently by the Reactor Engineering Division, ANL.

References

1. Frenzen, P., Radiological Physics Division Semiannual Report, ANL-5679, p. 95 (1957).
2. Frenzen, P., Radiological Physics Division Semiannual Report, ANL-5755, p. 77 (1957).
3. Frenzen, P., Radiological Physics Division Semiannual Report, ANL-5829, p. 218 (1958).
4. Gifford, F., Jr., Personal communication.
5. Webb, E. K., (1955) Autocorrelations and Energy Spectra of Atmospheric Turbulence; Tech. Paper No. 5, CSIRO, Div. Meteorol. Phys., Melbourne, Australia, 28 pp.
6. Vanoni, V. A. and N. H. Brooks, (1955) Report No. E-46, Hydro. Lab. Cal. Inst. of Tech., 51 pp. (ASTIA Reprint AD 66182).

ATMOSPHERIC DIFFUSION STUDIES

Peter E. Wasko and Harry Moses

A. Introduction

Objectives of the research described in this report are the determination of the ability of the lower atmosphere, in various meteorological states, to diffuse materials released into it and the increase of our understanding of the physical processes involved.

Photographs of smoke plumes obtained under various meteorological conditions were analyzed with those objectives in mind. To date the analysis has been aimed at obtaining a clear picture of the smoke plume characteristics and behavior. To do this, graphs of smoke plume measurements, such as horizontal width, thickness, rate of growth (divergence), are presented. One feature of the analysis has been the separation of the larger-scale diffusion, as shown by the meander of the smoke plume, from the smaller-scale diffusion as shown by the growth of the smoke plume around the center line. However, additional diffusion measurements made under different meteorological conditions must be made before it is possible adequately to relate the effect of meteorological variables on the diffusion process.

B. Procedure

The equipment (meteorology smoke stack, the K-24 aerial cameras, photogrammetric analyzer, and IBM-650 computer) used in the procedure from the observation to the computation stage has been discussed in previous reports (see ANL-5679, ANL-5518, ANL-5456, and ANL-5755). However, a brief review of the observational setup is essential as background for the reader.

Two K-24 aerial cameras facing each other and tilted 30° with respect to the horizontal are positioned on a 400-foot base line located 80 feet from the meteorology stack. These cameras, designated the A and B cameras, photograph the plume from opposite sides. A third K-24 aerial camera designated as the C camera is placed equidistantly from the other cameras at 40 feet from the stack and directed vertically upward so that the bottom of the plume is photographed.

Two planes of reference are used in this work. The first is called the datum plane which may be defined as that vertical plane going through the smoke stack, containing the C camera's axis, and bisecting the 400-foot base line of the A and B cameras. The second plane called the base line

plane is the vertical plane containing the base line between the A and B cameras. A Cartesian frame of reference is used in carrying out these analyses. In this coordinate system the origin is at the top of the stack, the X axis is horizontal and in the datum plane, the Z axis is vertical and also in the datum plane, and the Y axis is perpendicular to the X and Z axes.

Before each run, camera locations are selected which allow the smoke plume to lie as close as possible to the datum plane. The area around the smoke stack has been surveyed and camera locations determined for 18 different 400-foot base lines. Consecutive base lines make an angle of 20° with each other. The midpoints of the base lines lie on a circle having a radius of 80 feet with the stack as center. The data from these photographs are analyzed by means of a photogrammetric plotter and key-punched on cards. Calculations are performed by an IBM-650 computer.

C. Status of Work

In a previous Argonne report, ANL-5755, some results of the run made on October 5, 1956 were presented. To date, two additional runs have been completely analyzed. Five more runs are being processed in the IBM computing section.

The time required to reduce the data on the photogrammetric plotter represents a serious bottleneck to the progress of our work. At the present time, it takes over a man-month to analyze a single run. A single run usually consists of 90 sets of photographs taken at varying intervals.

D. Techniques of Analysis

1. Analysis of photographs from the A and B cameras

In the analysis of these data, simultaneous sets of the photographs taken by the A and B cameras are projected on a translucent screen from opposite sides. The screen is adjusted to a position corresponding to the datum plane by bringing the projected image of the smoke stack from each film into coincidence. The screen is then moved so that the tops of the plumes coincide at a distance of 2.5 feet from the stack. The Z value of the top of the plume with respect to the stack's top is then recorded. Also, the Y position of the screen is recorded. The screen is then moved until the bottoms of the plume coincide and the Z and Y positions are again recorded. This process is repeated for a distance of 5 feet from the stack and 5-foot intervals thereafter to a distance of about 160 feet.

The measurements from the A and B camera photographs are obtained by means of a cursor, riding on the translucent screen. Information concerning the position of the X and Z positions of the cursor are transmitted by synchromotors to Veeder Root counters which display numerical values corresponding to the X and Z distances. The screen itself is also connected through a synchromotor to a Veeder Root counter enabling the determination of the Y positions. These data are then recorded and transferred to cards for processing through the IBM-650 computer.

2. Analysis of photographs taken from the C camera

For the analysis of these data the translucent screen is first placed in a position to correspond to the distance of the horizontal plane through the top of the stack. Measurements are taken of the edges of the plume at 2.5 feet, 5 feet, and every 5 feet thereafter to the end of the photograph. The screen is moved, wherever necessary, to agree with the position of the plume center line above or below the top of the stack as calculated from the A and B camera photographs. An average of the Z values of the top and bottom of the plume is taken as the plume's center line position. The mechanical arrangement using the cursor, synchromotors, and Veeder Root counters are also used in recording the C-camera data.

E. Results and Discussion

1. Diffusion about Plume's Center Line

A quantitative determination of the rate of diffusion about the center line of the plume may be obtained from the measurements showing how the horizontal width and the thickness of the plume varies along its center line. From these measurements, it is possible to obtain several derived parameters such as the cross-sectional area along the plume's center line or the divergence of these variables.

In this report are presented the data of three runs. One of these runs, that of October 5, 1956 was presented in ANL-5755; however, some of these results are repeated for the purpose of comparison with the two other runs which were made on August 15 and September 28, 1956. The associated meteorological and experimental parameters are presented in Table 22.

A refinement in the analysis technique has shown that the values for thickness presented previously were too high by about 13%. These have been corrected in this presentation.

Table 22

Meteorological and Stack Parameters

Date	August 15, 1956	September 28, 1956	October 5, 1956
Time (CST)	1005 - 1034	1423 - 1447	1500 - 1512
Stack height	111 feet	111 feet	111 feet
Stack flow rate and Effluent temperature at top of stack	4200 CFM; 96° F 3000 CFM; 100° F 2000 CFM; 104° F 1500 CFM; 100° F	4400 CFM; 100° F 2500 CFM; 114° F 2000 CFM; 132° F 1500 CFM; 133° F	2500 CFM; 115° F
Weather	Scattered clouds	Scattered clouds	Clear
Ground condition	Dry	Dry	Dry
Ambient air temperature at 111 feet	81° F	75° F	66° F
Mean wind speed at 111 feet	9 mph	14 mph	11 mph
Mean wind direction at 111 feet in degrees	260	180	100
Temperature at 144 feet minus Temperature at 5.5 feet	-1.3° C	-1.3° C	-0.8° C
Solar radiation on horizontal surface	1.24 Langleys/min	0.83 Langleys/min	0.58 Langleys/min

a. Horizontal Width

The horizontal width along the plume's center line for the three runs are presented in Figs. 54a-c. The horizontal width is defined as the horizontal dimension of the plume in the plane perpendicular to the plume's center line at the point of measurement. All three runs plotted on the same chart are shown in Fig. 54d. These curves show a considerable amount of similarity both in the magnitude of the horizontal width and the rate at which it increases along the center line. However, close examination of these data indicates that the run of September 28th showed somewhat smaller widths than those of the other two. In Figs. 54a-c are presented also the standard deviations of the horizontal width. The number of cases which were used to obtain the averages and standard deviations ranged from 23 to 62.

b. Plume Thickness

The plume thickness, as used here, denotes the dimension of the plume in the plane perpendicular to the plume's center line which is at right angles to the horizontal dimension. If the center line deviates from the horizontal, then the direction of the thickness length deviates from the vertical. The mean plume thickness based on 21 to 62 cases as a function of distance along the center line is shown in Figs. 55a-c. The standard deviation is also shown on these charts. In Fig. 55d is plotted the mean

FIG. 55a
AUGUST 15, 1956
1005-1034 CST

FIG. 55b
SEPTEMBER 28, 1956
1423-1447 CST

FIG. 55c
OCTOBER 5, 1956
1500-1512 CST

FIG. 55d
COMPARISON OF MEAN THICKNESS OF PLUMES

thickness for all three runs. It is rather interesting to find that the magnitudes of the plume thicknesses differ by so little for given distances along the center line.

c. Cross-Sectional Area

The cross-sectional area is defined as the area formed by the intersection of the plume with the plane perpendicular to its center line. This area is assumed to be elliptical and is calculated from the expression

$$\frac{\pi \times \text{thickness} \times \text{horizontal width}}{4}$$

Since the cross-sectional area is a derived quantity, it is not surprising that its variation along the plume's center line is similar to that of the horizontal width and thickness. These curves are shown in Figs. 56a-d.

d. Ratio of Plume Horizontal Width to Thickness

The ratio is also a derived quantity. The mean ratio of the horizontal width to thickness along the plume's center line is shown in Figs. 57a-c. It is to be noted that the curve for the experiment of September 28, 1956 is appreciably different from those of the other two in that the horizontal width remained relatively smaller at corresponding points compared to those of the other two runs. The standard deviation of the mean ratios is also shown on these charts. The experiments of September 28th and October 5th show a marked tendency for the horizontal width to be smaller than the thickness since the curves fall significantly below the ratio equal unity line. This tendency is also exhibited by the measurements of August 15th but to a lesser extent. With the data at hand, it is too early to be able to ascribe the variation of meteorological parameters to the difference observed in the variation of thickness and width with distance along the plume's center line.

e. Divergence Calculations

The term divergence as used here represents the percentage rate of change of the variable with distance along the plume's center line. Divergence calculations were made for the following variables: 1. horizontal width, 2. thickness, and 3. cross-sectional area. As indicated in an earlier report, ANL-5755, the divergence of a cross-sectional area represents the rate of entrainment of air by the small-scale eddies into the plume. Divergence of the area was calculated from the expression

$$\frac{\Delta \ln A}{\Delta S}, \text{ which is approximately equal to the conventional form } \frac{1}{A} \frac{\Delta A}{\Delta S}.$$

Divergence calculations for the three experiments are shown in Figs. 58a-c. The striking feature concerning these curves is the general similarity among all three. It is to be noted that the divergence rate levels off at about 20 feet or roughly 13 stack diameters along the plume's center line.

2. Diffusion due to Meander of Plume

The meander of the plume is caused by the large-scale eddy motions in the atmosphere as distinguished from the smaller-scale eddies which cause diffusion about the center line of the plume itself.

The displacement of the plume center line as a function of both time and distance from the stack defines the meander of the plume. In studying this phenomenon the spatial distribution of the points of intersection of the plume's center line with a sphere of a given radius were computed. The sphere was centered at the top of the stack. Photographs of the plume were made every fifteen seconds. Concentric spheres having radii of 2.5 feet, 5 feet, 10 feet, and every 5 feet thereafter to 160 feet were used in this study.

The points of intersection of the plume center lines and the sphere were expressed in a spherical coordinate system where R equals the radius, ϕ the latitude and θ the azimuth angle. For a given spherical shell R is constant, and then the position of any point is a function of ϕ and θ . The distribution of intersection points may be transformed or mapped on a plane using a rectangular x, y coordinate system according to the transformation

$$\begin{aligned} y &= R \phi \\ x &= R \theta \cos \phi \end{aligned}$$

The origin of the x, y coordinate system may correspond to the centroidal position of the intersection points on the sphere.

If it is assumed that these points are normally distributed, a reasonable assumption, and a normal correlation surface is constructed to represent the bivariate frequency distribution, we have

$$z = \frac{1}{2\pi\sigma_x\sigma_y(1-r^2)^{1/2}} \exp \left\{ -\frac{1}{2(1-r^2)} \left(\frac{x^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2} - \frac{2rxy}{\sigma_x\sigma_y} \right) \right\} \quad (1)$$

where

z = the theoretical frequency

σ_x = the standard deviation of the x components

σ_y = the standard deviation of the y components

and r = the correlation coefficient between the x and y values.
[See reference (1).]

For constant values of z , equation (1) represents a series of ellipses any one of which has the form of

$$\frac{x^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2} - 2r \frac{x}{\sigma_x} \frac{y}{\sigma_y} = \lambda^2$$

where λ is a function of σ_x , σ_y and r . The area of any ellipse is given by

$$\frac{\pi \lambda^2 \sigma_x \sigma_y}{(1-r^2)^{1/2}}$$

The probability that a point will fall within the ellipse obtained for an assigned value of λ is

$$\frac{2\pi \sigma_x \sigma_y}{2\pi \sigma_x \sigma_y (1-r^2)} \int_0^\lambda e^{-\frac{\lambda^2}{2(1-r^2)}} \lambda d\lambda = 1 - e^{-\frac{\lambda^2}{2(1-r^2)}}$$

If we assume that the meander area corresponds to the ellipse containing 95% of the observations we have

$$1 - e^{-\frac{\lambda^2}{2(1-r^2)}} = 0.95 \text{ or } \frac{-\lambda^2}{2(1-r^2)} = \ln 0.05 = -2.9957$$

$$\lambda^2 = 5.9914 (1-r^2)$$

Because the range of latitude and azimuth angles are relatively small, the areas of the ellipses on the x,y plane are nearly equal to the corresponding areas mapped on the sphere. The difference does not exceed 2 or 3%. It is, therefore, valid to consider elliptical areas obtained on the plane as the plume meander areas on the sphere.

In Fig. 59 are plotted the meander areas as a function of radial distance from the stack for the three experiments. The data were further divided according to stack velocities. This, of course, was necessary since the plume center lines are elevated with greater stack velocities, thus altering the location of the intersection points of the plume center line and the spherical surface.

The magnitudes of the meander areas are several times larger than the cross-sectional areas of the plume for corresponding distances. Further preliminary calculations show that the divergence or the percent rate of change of the meander areas is larger than that of the plume area itself. This is significant since the ratio of the meander area to plume area must, therefore, increase with distance. At about 65 feet from the stack the meander area is about 10 times or an order of magnitude greater. This result indicates that, for a range of distances from a source of contamination, the concentration average as given by Sutton's formula or discussed in the literature is really made up of high concentrations for relatively short intervals separated by periods of zero or very small concentrations.

Another interesting phenomenon shown by these data is the suppression of the vertical component of the motion in the large eddies responsible for the meander. It may be recalled from Fig. 57 that the vertical dimension of the plume exceeded the horizontal dimension, but in case of the meander area a test for circularity shows that this area is circular.

A test for circularity given by Mauchly⁽²⁾ and described by Brooks and Carruthers⁽³⁾ may be expressed by

$$Le = \frac{2 \sigma_x \sigma_y \sqrt{1 - r_{xy}^2}}{\sigma_x^2 + \sigma_y^2}$$

where the symbols have the usual meanings.

If $Le > 20 \frac{1}{N-2}$ then the distribution is circular at the 95% level. N is the number of cases. An analysis of the data shows that the distribution of the intersection points is significantly circular, thus indicating a significant difference in the shapes of the cross-sectional and meander areas for these data.

The effect of stack velocity on the height of rise is shown in Figs. 60a and b. Unfortunately the stack temperature did not remain constant although the variation was not too great. It is to be noted that for the lower stack velocities there was a tendency for the plume to fall below the stack top. Additional data are necessary before quantitative formulations of the relationships can be made.

References

1. Rietz, H. L., Mathematical Statistics, The Carus Mathematical Monographs, No. 3, The Open Court Publishing Company for the Mathematical Association of America, 1927, pp. 104-109.
2. Mauchly, J. W., "A Significance-Test for Ellipticity in the Harmonic Dial," Terrestrial Magnetism and Atmospheric Electricity, Washington, D. C. 45, 1940, p. 145.
3. Brooks, C. E. P. and N. Carruthers, Handbook of Statistical Methods in Meteorology, Her Majesty's Stationery Office, London, 1953, p. 233.

STAFF ACTIVITIES

January-June 1958

Am. Phys. Soc., New York, Jan. 29-31, 1958. "Bromine Delayed Neutron Precursor of 1.4-sec Half-life." G. J. Perlow and A. F. Stehney.

American Radium Society, Hollywood Beach, Florida, March 26-28, 1958. "Natural Radioactivity in the Human." L. D. Marinelli.

American Water Works, Illinois Section, Chicago. March 26-28, 1958. "The Natural Radium Content of Public Water Supplies in Illinois." A. F. Stehney and H. F. Lucas, Jr.

Health Physics Society, University of California Radiation Laboratory, Berkeley, California June 9, 1958. "Low Intensity Spectrometry of the Gamma Radiation Emitted by Human Beings." C. E. Miller.

LaSalle County Medical Society meeting at ANL. June 12, 1958
"Contemporary Radioactivity of the Natural Environment." A. F. Stehney.
"Microradiography of Bone and Problems of Dosimetry " R. E. Rowland.
"Wider Implications of Current Studies of Radioactivity " L. D. Marinelli.

PUBLICATIONS

January 1, 1958 - August 1, 1958

- Gustafson, P. F., L. D. Marinelli and S. S. Brar. Natural and Fission-Produced Radioactivity in Soil. *Science* 127:1240-1242 (May 23, 1958).
- Gustafson, P. F., L. D. Marinelli and S. S. Brar. The Use of Gamma-Ray Spectroscopy in the Determination of the Gamma-Ray Dose Due to Radioactive Fallout Present on the Ground. *Rad. Res.* 9:125 (July 1958) (Abstract).
- Jowsey, J., R. E. Rowland and J. H. Marshall. The Deposition of Rare Earths in Bone. *Rad. Res.* 8(6):490-501 (1958).
- Marinelli, L. D. The Use of Low Level Gamma-Ray Spectrometry in the Field of Nuclear Medicine. *Minerva Nucleare* 1(4-5):201-207 (October 1957).
- Marinelli, L. D. The Relevance of the Skeletal Burden of Thorotrast Patients to the Problem of Chronic Toxicity of Bone Seekers in Man. *Radiology* 70:93-94 (January 1958) (Abstract).
- Marinelli, L. D. Radioattività del corpo umano. Estratto dagli Atti del Congresso Scientifico - Sezione Nucleare - indetto in occasione della IV Rassegna Internazionale Elettronica Nucleare-Giugno 1957, pp. 533-549.
- Marinelli, L. D. An Estimate of Bone Cancer Incidence in Man as a Function of Radium Burden. *Rad. Res.* 9:149 (July 1958) (Abstract).
- Marinelli, L. D. and J. E. Rose. Occurrence of Cs^{137} in the Atmosphere, Biosphere, and Its Uptake and Behavior in Man. Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the U. S. Eighty-fifth Congress. First Session on The Nature of Radioactive Fallout and Its Effects on Man. Part I, 1957. U. S. Govt. Printing Office, Washington, D. C., 1957. pp. 764-765.
- Marinelli, L. D. (Contributing Author) NBS Handbook. No. 63. Protection Against Neutron Radiation up to 30 Mev. Nov. 22, 1957.

- Miller, C. E. Measurements of Radioactivity in the Lungs. Rad. Res. 9:154, (July 1958). (Abstract).
- Perlow, G. J. and A. F. Stehney. Bromine Delayed Neutron Precursor of 1.4 Sec Half-Life. Bull. Am. Phys. Soc. 3:6 (January 1958). (Abstract).
- Rowland, R. E., J. Jowsey and J. H. Marshall. Radon Escape from Bone Mineral. Rad. Res. 8(4):298-306 (1958).
- Wanta, R. C. and H. Moses. Report on Meeting of Ad-Hoc Committee on Scaling Mesometeorological Phenomena. ANL-5869. June 1958.

TO BE PUBLISHED

- Jowsey, J., R. E. Rowland, J. H. Marshall and F. C. McLean. The Effect of Parathyroidectomy on Haversian Remodeling of Bone. *Endocrinology*.
- Jowsey, J. Radioelement Distribution in Skeletal Tissue. *Atompraxis*. 1958.
- Lucas, H. F., Jr., and F. H. Ilcewicz. The Natural Ra^{226} Content of Illinois Municipal Water Supplies. *J. Am. Water Works. Assoc.*
- Marinelli, L. D. Radioactivity and the Human Skeleton. *Am. J. Roentgenol. Ra Therapy and Nuclear Med.*
- Marshall, J. H., V. K. White and J. Cohen. Microscopic Metabolism of Calcium in Bone. I. Three Dimensional Deposition of Ca^{45} in Canine Osteons. *Rad. Res.*
- Marshall, J. H., R. E. Rowland and J. Jowsey. Microscopic Metabolism of Calcium in Bone. II Quantitative Autoradiography. *Rad. Res.*
- Marshall, J. H., J. Jowsey and R. E. Rowland. Microscopic Metabolism of Calcium in Bone. IV. Ca^{45} Deposition and Growth Rate in Canine Osteons. *Rad. Res.*
- Marshall, J. H., R. E. Rowland and J. Jowsey. Microscopic Metabolism of Calcium in Bone. V. The Paradox of Diffuse Activity and Long Term Exchange. *Rad. Res.*
- Miller, C. E., and L. D. Marinelli. Low Intensity Spectrometry of the Gamma Radiation Emitted by Human Beings. *Geneva Conference Proceedings*, 1958.
- Rowland, R. E., J. H. Marshall and J. Jowsey. Radium in Human Bone: The Microradiographic Appearance. *Rad. Res.*
- Rowland, R. E. Quantitative Microradiography of Bone. Medical Physics. Edited by O. Glasser.
- Rowland, R. E., J. H. Marshall and J. Jowsey, Microradiography and Micro-Dosimetry of Human Bone Burdened with Ra^{226} . *Radiology*. (Abstract).
- Rowland, R. E., J. Jowsey and J. H. Marshall. Microscopic Metabolism of Calcium in Bone. III. Microradiographic Measurements of Mineral Density. *Rad. Res.*

- Rowland, R. E., J. Jowsey and J. H. Marshall. Structural Changes in Human Bone Containing Ra^{226} . Geneva Conference Proceedings, 1958.
- Shonka, F. R., J. E. Rose and G. Failla. Conducting Plastic Equivalent to Tissue, Air and Polystyrene. Geneva Conference Proceedings, 1958.
- Stehney, A. F. and G. J. Perlow. Identification of Delayed Neutron Activities by Chemical Separation. Geneva Conference Proceedings, 1958.
- Wasserman, R. H., C. L. Comar and J. Jowsey. Quantitative Studies on Skeletal Accretion and Resorption in Domestic Animals. Geneva Conference Proceedings, 1958.