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THREE DIMENSIONAL DEPOSITION OF CALCIUM-45 IN CORTICAL BONE OF DOGS*

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I Introduction

The microscopic pattern of deposition of the calcium-like elements in bone is of interest both in the study of the biological mechanisms of bone calcification and in the evaluation of the long-term radiation damage to bone produced by internally-deposited radium or strontium. The nonuniformity of the pattern is often so great that sampling and counting techniques are of little use except for the determination of the average deposition or retention. Microscopic deposition of radioactive elements has therefore been studied by film autoradiography. It is the aim of our work to use autoradiography quantitatively to investigate the microscopic dynamics of calcium metabolism in bone. This is the first step, a three-dimensional mapping of some of the regions of most active calcium deposition in bone. To our knowledge, three dimensional autoradiographic mapping of bone has not heretofore been attempted.

The deposition of Ca^{45} in cortical bone may be conveniently considered in terms of three components. First is the general diffuse component which spreads through nearly all the cortical bone in relatively uniform concentration. Second is the deposition by appositional growth at the periosteal or endosteal surfaces. Third is the deposition in haversian systems or osteons. It is the three-dimensional description of this third component that forms the subject of this report.

The cortical bone examined included sections from two adult mongrel dogs, one young, the other middle-aged. The sections were taken from three bones, the humerus and the left and right femurs, and from two locations, the middle of the shaft and the end of the shaft.

Each dog had been intravenously injected with a tracer dose of Ca^{45} . When viewed on a cross-section autoradiogram, a calcifying osteon appeared as an intense spot or "hotspot." These hotspots were followed in serial cross-section autoradiograms by two techniques, one which gave 50μ intervals and the other which gave 150μ intervals. In the first case, the behavior of each

*Part of the material in this report was derived from research performed while the senior author was at the Radioactivity Center of the Department of Physics, Massachusetts Institute of Technology.

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hotspot was compared with the corresponding osteon architecture for a depth of half a millimeter in order to reveal what relations might exist between the region of active calcification and the diameter, length, and branches of the osteon. In the second case, cross-section autoradiograms were followed for almost a centimeter from which a map was constructed to show the extent and ramifications of hotspots in three dimensions.

II. Method

The 50μ interval autograms were made by the method of serial facing, described in detail elsewhere.⁽¹⁾ Briefly, serial Ca^{45} autograms with corresponding photomicrographs of cross sections of undecalcified bone were made by mounting thin blocks and facing one surface repeatedly on a lathe. Initially, each cross-section block was about 1 mm thick. Each facing on the lathe removed 50μ of bone. Each face was autoradiographed on Eastman No-Screen Autoradiographic plates. The two dogs received about $50\mu\text{c}$ Ca^{45} per kilogram body weight which required an exposure time for each autoradiogram of about 20 hours. The autoradiographic plate and the bone block, which was mounted on a microscope slide, were held during exposure in a right-angled jig identical with the slide holder on the microscope. Alternate examination of the plate and the bone under the microscope gave unambiguous identification between the hotspots and their corresponding osteons. The 50μ interval between successive faces was chosen to yield a maximum of information about the continuity of an osteon's pattern of calcification. The range of Ca^{45} beta particles in bone is such that the half-value thickness for absorption in bone is roughly 30μ . Thus, intervals less than 50μ , though feasible, were unnecessary. Intervals much larger than 50μ might have allowed us to miss significant variations in the deposition.

The results obtained by serial facing are given in Sections III and V.

Having established the three dimensional continuity of the Ca^{45} deposited in haversian systems over many 50μ intervals, we then used a direct serial sectioning technique in order to follow the hotspots for larger distances. Undecalcified, dehydrated bone, embedded in bioplastic, was sectioned at 300μ intervals using a circular saw 150μ thick. Autoradiograms were then made at 150μ intervals by sandwiching the 150μ thick bone sections between two autoradiographic plates. When the plates were analyzed under the microscope, every other plate was turned upside down so that the hotspots upon adjacent plates could be superimposed. Since the majority of hotspots changed very little from plate to plate it was simple to maintain the identity of each hotspot on the successive plates. The analysis of the plates was greatly facilitated by projecting each plate at a magnification of 20 on the ground glass of an Eastman Contour Projector. The distribution of hotspots was traced in black pencil and the relative intensities of the hotspots

was recorded in a color code directly on the ground glass. When the next plate was projected and superimposed upon the tracing, the modification in the tracing necessary to make it correspond to the new plate could be made. Tracing paper copies of each distribution were kept so that the three dimensional extent of all the hotspots followed could be reconstructed. The results are given in Section IV.

III. Serial Facing Results

All of the data in this section were obtained from the left and right femur of an adult mongrel dog (MIT No. C119), judged to be in middle age and sacrificed 14 days after injection. One of the cross-section autograms from this dog is shown in Fig. 1. Six cross-section blocks were serially faced, four of them, CF-35, 36, 37, 38, were adjacent sections from the middle of the shaft of the right femur. Block CF-79 came from the middle of the shaft of the left femur and Block CF-97 from the distal end of the shaft of the left femur. About 10 of the hottest spots from each of these 6 blocks were followed for from 9 to 14 serial faces as illustrated by Table 1.

TABLE 1

Calcification length from serial facing

Block No.	S*	D	d	Sd	$\frac{Sd}{L}$	$\frac{(Sd/L - D)^2}{Sd/L}$
CF 35	7	0	18 mils**	126 mils	2.4	2.4
CF 36	5	3	18	90	1.7	1.0
CF 37	7	2	18	126	2.4	0.1
CF 38	14	4	18	252	4.8	0.1
CF 79	14	10	22	308	5.8	3.0
CF 97	15	6	28	420	7.9	0.5
		$\sum D = 25$			$\sum Sd = 1322 \text{ mils}$	$\chi^2 = 7.1$

$$L = \frac{Sd}{D} = \frac{1322 \text{ mils}}{25} = 53 \pm 11 \text{ mils} = 1.3 \pm .3 \text{ mm}$$

Probability that repetition of the observations would show greater statistical deviations than $\chi^2 = 7.1$ is $P = 0.7$ so the given data is self-consistent.

*For explanation of column headings see text immediately following table.

**25.4 μ = 1 mil

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Column S shows the number of spots that were chosen at the start, Column D, the number of the initial spots which were observed to come to an end and Column d the total depth to which each block was faced. Each facing removed 2 mils (50μ) of bone so, for example, block CF35, with $d = 18$ mils, was faced 9 times, while block CF-97 with $d = 28$ mils was faced 14 times.

From the observed number of hotspots which end in a given depth of bone we can calculate approximately the average length of the actively calcifying region in an osteon. Let us assume for the moment that every hotspot is the same length, an assumption which will be shown later to produce no appreciable error. Let us call this the calcification length. Now, although the number of hotspots in a cross section of cortical bone is somewhat greater near the ends of the bone than it is in the mid-shaft, the variation of the average number of hotspots in a cross section is very small over a distance of a millimeter (40 mils). Therefore, if every system had the same calcification length and the actively calcifying regions were distributed at random along the length of the bone, then the situation could be schematically represented as in Fig. 3. If the number of hotspots present in any given cross section is S and the number of hotspots which have ended within a distance d below the given section is D, then the average calcification length is $L = \frac{Sd}{D}$. Using this relation, the data in Table I give $L = 53 \pm 11$ mils or $1.3 \pm .3$ mm. The chi square test⁽²⁾ for the statistical consistency of this data gives $P = 0.7$ which means that considering the number of hotspots followed, the deviations in the data in Table I are purely random, and there is no significant difference between the calcification lengths that would be derived from the different blocks considered separately. This calcification length, L, was derived with the oversimplified assumption that all hotspots were the same length. However, if we take a normal distribution of lengths with a standard deviation half the mean value and perform the same analysis indicated by Fig. 3, the value for the average calcification length is only about 20% larger than that derived above. Such a distribution of lengths about the average is a reasonable assumption (verified in Section IV). Therefore the best value for the average calcification length derived from serial facing is $L = 1.6 \pm .4$ mm. This value compares favorably with the more direct results of the next section.

IV. Serial Sectioning Results

All of the serial sectioning results were obtained on a 1 cm length of a quadrant from the mid-shaft of the right humerus of a young adult mongrel dog. The dog (MIT No. C-120) was sacrificed 12 hours after

Fig. 1. Ca^{45} autoradiogram from middle-aged dog (C119), midshaft of femur.

Fig. 2. Ca^{45} autoradiogram from midshaft of femur of young dog (C120).

Fig. 3. Derivation of calcification length from serial facing.

Fig. 4. Ca^{45} autoradiogram from a quadrant of the midshaft of the humerus of dog C120.

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injection.* One of the serial autoradiograms from this dog is shown in Fig. 4. Every detectable hotspot in this quadrant of the cross section, 53 hotspots in all, was followed for 23 serial sections above and 28 serial sections below the master section shown in Fig. 4. The vertical spacing between serial autoradiograms was 150μ so that the total length of bone analyzed was 7.8 millimeters. Figure 5 shows the results of this 3 dimensional reconstruction. Hotspots which interconnect have been placed side by side in Fig. 5 so that the whole 3 dimensional extent of the Ca^{45} deposition could be portrayed clearly in 2 dimensions. The widths of the actively calcifying regions are drawn approximately to the same scale as the length.

Fig. 5. Three dimensional map of the hotspots shown in Fig. 4. Fig. 4 is the cross section view of the hotspots at the level of the master section, Number 24.

Each active osteon is drawn vertically in a single column, even though individual osteons may not run perfectly parallel to the axis of the bone. No attempt has been made to distinguish between actively calcifying systems that appear on the autoradiogram merely as a spot and those that are large enough and well enough resolved to appear as a crescent or a ring. The

*This short time interval was chosen in connection with other work. It should not affect the present results which do not deal quantitatively with hotspot intensities.

widths of the activity distributions shown in Fig. 5 were obtained by subtracting $100\ \mu$ from the width of the hotspot on the autoradiogram in order to account approximately for the effect of resolution; the value $100\ \mu$ was subtracted because this is the spot width that would be produced by any active region much less than $100\ \mu$ in diameter: no spot smaller than $100\ \mu$ in diameter was observed with the present technique. The most intense Ca^{45} concentrations are drawn in black with regions of lesser concentration in grey. Points along a particular system at which active canals branch off but were not followed are indicated by crosses. Active branches which lead toward but did not reach the master section were not followed; they appear as short dashed lines.

Figure 6 shows the distribution of hotspot lengths as measured from Fig. 5. The length of every hotspot which appeared in the master section was measured from the point the activity was first detected above the master section to the point where the activity disappeared below the master section. In those cases in which one of the master section hotspots connects laterally with another hotspot not present in the master section, the length of the hotspot refers only to the length before the active branching occurred. For example, in Fig. 5 there is one case in which activity was continuously followed through all 52 sections, but the hotspot and haversian system found in Section 1 was connected to the hotspot in Section 52 by a hot lateral branch at Section 51. Four of the 53 hotspots present in the master section continued beyond the limits of the 7.8 millimeter distance analyzed. These systems are recorded with arrows pointing to the right in Fig. 6. The mean length of the hotspots as shown in Fig. 6 is about 2 mm. This value is somewhat larger than that found by the method of Section III, but considering that the present analysis was performed on a younger animal which was undergoing much more rapid formation of secondary haversian systems (compare Fig. 2 with Fig. 1) and therefore has many systems in the slowly-calcifying final stages of growth, the results are remarkably consistent. In Fig. 5 it can be seen that the diameter and the intensity of the activity distributions varies considerably along the whole length of a calcifying osteon. However, there are considerable distances along which the intensity of calcification is relatively constant. Figure 7 shows the distribution in length of the most actively calcifying regions, indicating a mean length of somewhat less than 1 mm.

V. Comparison of the Ca^{45} Deposition with Osteon Architecture

Serial photomicrographs and corresponding autoradiograms of 33 osteon hotspots in dog C119 were prepared by the serial facing technique. The results can be described by considering the behavior of the 33 hotspots under several categories

Fig. 6. Osteon calcification length. Data from Fig. 5.

Fig. 7. Most active calcification length. Data from Fig. 5.

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(1) Hotspot disappearance accompanying a branch or junction of haversian canals: There were nine examples of this type of behavior in two of which the joining canal itself was hot and in seven of which the joining canals were not hot. Figure 8 shows a good example of hotspot disappearance (or appearance) associated with a branching point in which the branches are cold.

(2) Hotspot appearance as one moves along a resorption cavity into a region in which calcification is occurring: The central spot in Fig. 11 is a good example of this type, of which there were four examples. Note that it takes 200-300 μ of travel along the resorption cavity until the hotspot is symmetrical. If a high resolution autoradiogram were made at a level between the first appearance of the hotspot and the symmetrical region, the osteon would be found to be calcifying only around part of its circumference. Most calcifying osteons when viewed more than a canal diameter away from the onset of calcification along the canal show a relatively symmetrical pattern of calcification.

(3) Hotspot appearance as one moves along a 20 μ diameter canal into an enlarged diameter canal that is calcifying; no evidence of branches. Two examples of this type were found.

(4) Disappearance of the hotspot and coincident disappearance of the osteon itself: Fig. 9 is an example of this type. As far as can be ascertained from the photomicrograph of the bone, not even the central canal of the osteon continues through the bone. The whole calcifying system seems to come to a dead end. Six examples of this type of behavior were found. Feeling that it is quite possible that a continuing canal might have been missed due to the thickness of the bone section being examined, one of us (J. Cohen) has proceeded with more extensive work on this point and on the general reconstruction of osteons by sectioning decalcified bone. Dead end canals, which show by the intense concentrations of Ca^{45} that they are calcifying and not resorbing, might show that the network of osteons in cortical bone in part resembles the growth of a tree. It should be emphasized, however, that dead ends are infrequent.

(5) Hotspot unchanged through all the sections examined in spite of hot branching canals: The right hand spot in Fig. 11 is one of the two examples of this type.

(6) Hotspot unchanged in spite of cold branching canals: There were two examples of this type.

(7) All other continuous though not necessarily constant intensity hotspots, eight in all. Figure 10 is an example of this type. Note that there is a definite correlation between diameter and hotspot intensity in Fig. 10. If we consider the large cold canal which crosses the lower sections in Fig. 10, this hotspot might also be classified under Type (6).

Figs. 8, 9, 10, 11. Serial photomicrographs and corresponding autoradiograms for osteon hotspots in dog C119. Note that the scales for corresponding pictures in Fig. 8, 9, and 10 are different so as to include more than one hotspot for reference.

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MICRONS

Fig. 10

Fig. 11. (see also Figs. 8, 9, and 10)

VI. Conclusions

1. Rapidly calcifying regions in haversian systems in dogs have an average length the order of 1-2 mm. Note that this length is roughly ten times the average osteon diameter.

2. The majority of osteons in the animals studied calcify all around their circumference at the same time. The sections in which calcification was observed to occur only in part of the circumference of the osteon are shown to lie near the end of the region of active calcification as one moves along a canal. The length of this nonsymmetrical region is comparable with the canal diameter.

3. No significant correlation was found between the locations of vascular connections of an osteon canal and the limits of actively calcifying regions. On the contrary, regions of active calcification were found to end under many different conditions, which suggests that the local mechanism for determining the location of new resorption cavities and consequently the subsequent location of actively calcifying regions is perhaps not dependent upon details of the vascular network in bone such as the locations of branching vessels.

4. A few hotspots and their accompanying osteons were found to end blindly as cross section serial faces were followed. The presence of Ca^{45} activity two weeks after injection in dead-end osteons shows that they were calcifying and not resorbing.

5. A derivation of the rates of osteon calcification from hotspot intensities (now in preparation) would be meaningless unless the hotspots were selected for three-dimensional continuity, because a significant fraction of the hotspots in any given cross section of bone will be due to junctions or branch points or will lie at the rapidly varying edge of calcification in an otherwise continuous osteon.

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GAMMA-RAY EMITTING ELEMENTS PRESENT IN SOIL

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

A gamma-ray spectrum of present-day surface soil reveals the presence of several lines, the intensities and energies of which correspond neither to K^{40} nor to those of the uranium or thorium series. These additional lines are due to fission product fallout from bomb tests as demonstrated by Fig. 12 which shows the gamma spectrum of soil taken in situ on the Laboratory grounds outside of the Biology Building on April 7 of this year. A 2-1/4 in. x 2 in. NaI crystal was placed 4 cm above the surface of the ground for this measurement, and the data were collected and analyzed by an Argonne Type 256 channel analyzer. For comparison, a spectrum was taken of a concrete floor using the same crystal placed 4 cm from the floor. The two spectra were normalized at the 1.46 Mev peak from K^{40} . The high degree of matching between the spectra obtained by this normalization indicates that the amounts of thorium, uranium, and K^{40} present in soil and in concrete are quite comparable in this instance. The spectra differ around 500 and 750 kev, which differences are due to Ru-Rh^{103,106} and Zr-Nb⁹⁵, respectively, as has been previously demonstrated in carpet dust.⁽¹⁾ Since the sources involved here have considerable thickness, there is a predominance of scattered radiation relative to that directly incident on the crystal and thus the observed increase in counting rate at energies below 500 kev. This situation is magnified further by the crystal efficiency which is greater at lower energy. The net result is that little detail is obtainable from bulk material below 500 kev.

However, if one examines a finite source of soil rather than an infinite plane source the low energy end of the spectrum contains much less scattered radiation. This is illustrated in Fig. 13 which shows the result of using a 200 g soil sample. The sample comes from the top 1.5 cm of soil taken from behind D202. This soil was dried, ground, and screened through 40-mesh screening. Then 200 g were put in a polyethylene bottle and placed on top of a 5-in. x 4-in. NaI crystal canned in electrolytic copper. The spectrum was run with both detector and soil inside the steel room in the basement of the Biology Building.** A second 200 g soil sample, obtained from 10 cm beneath the surface, was prepared and counted for the same period of time (20 hours). The difference spectrum is plotted in Fig. 13. In addition to Ru-Rh and Zr-Nb, the 135 kev line due to Ce^{144} is now apparent. The order of magnitude of activities present in this sample are: Ce^{144} 0.44 $\mu\mu\text{c/g}$, Ru-Rh¹⁰⁶ 1 $\mu\mu\text{c/g}$, and Zr-Nb⁹⁵ 3 $\mu\mu\text{c/g}$. There is no definite

*From the Division of Biological and Medical Research.

**The Bio-Med steel room⁽²⁾ has 9-in. steel walls, floor, and ceiling, but otherwise is dimensionally equivalent to the Radiological Physics steel room.⁽³⁾

Fig. 12. Gamma-ray spectrum of concrete and soil;
2 $\frac{1}{4}$ in. x 2 in. NaI crystal

Fig. 13. Fission gamma-ray spectrum of top 1.5 cm of soil at
Argonne National Laboratory (5-21-57); 200 g sample;
5 in. x 4 in. NaI crystal.

indication of Cs^{137} , which is not surprising since because of its longer half-life and subsequent lower specific activity it is 150-fold less active than Zr^{95} . Activity-wise this would amount to $.02 \mu\mu\text{c/g}$ or in the 200 g sample, this corresponds to about 400 counts in the 20 hour period.

The depth distribution of fallout activity in soil was determined by taking successive $1\frac{1}{2}$ in. layers of soil down to a depth of 12 in. This analysis was carried out on soil from Argonne Park, which was covered with thick grass and a layer of dry organic material. Table 2 indicates the percentage of the total fallout activity present in the various layers. From this data it appears that fallout forms primarily a surface deposit, and activity monitoring and dose calculations are somewhat simplified since it is not necessary to make extensive absorption corrections.

TABLE 2

Profile of fallout radioactivity in soil

Layer	Per cent of total activity
Grass & organic material	62
Soil 0 - $1\frac{1}{2}$ in. depth	27
Soil $1\frac{1}{2}$ - 3 in. depth	8
Soil 3 - $4\frac{1}{2}$ in. depth	3

We wish ultimately to make quantitative measurements of fission product activity present in soil. As a start Na_3PO_4 has been used as a mock soil. This compound has roughly the same density and atomic number as soil and contains no observable radioactivity. A 200 g sample of mock soil is then used for background purposes and samples with monazite sand or carnotite added are used to obtain the uranium and thorium spectra with the same amount of scattering and absorption as is found with the true soil samples. The characteristic K^{40} spectrum is obtained by measuring a 200 g sample of KCl.

The soil gamma spectrum may then be peeled apart by proper use of these 200 g samples of natural activity.^(4,5) Briefly, the procedure is as follows: The gamma spectrum of a 200 g soil sample is run with the gain set so that the 2.62 Mev line from ThC'' is visible. With background subtracted, the monazite spectrum is normalized to coincide in intensity with that from the soil at 2.62 Mev, and the normalized monazite spectrum subtracted off. The resultant net spectrum is that of soil minus the

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contribution due to the thorium series. A similar procedure is carried through for uranium and K^{40} until a spectrum due to fission products is obtained. By taking the highest energy line in this spectrum it is possible to peel it apart as well. This may be done quantitatively by using sources of known activity. Thus the amount of activity of the various isotopes may be determined per 200 g. If sampling is done over a known area, one may arrive at activity per unit area.

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POSITIVE ION EMISSION

H. A. Schultz

Introduction

In a halogen-sensitive positive ion emitter, the ionizable material is released from a source, transported to a heated surface, and ionized; then the positive ions are collected by an electric field.⁽¹⁾ Some recent work on two phases, release, and transport of ionizable material is here reported.

Improved Source of Ionizable Material

The source of ionizable material is a small platinum boat located near the bottom end of a vertical filament. Material is released when the boat is heated by passing an electric current through its walls. Sketches and further description are given in earlier reports.⁽²⁾ To assure constant ion current, the temperature of the boat is regulated by a control circuit.⁽³⁾ The steady-state operation was very good, but unfortunately there were initial transients which obscured the chemically-induced transients which should be studied. The control circuit was improved first,⁽³⁾ then the boat. In the earlier boat design, the establishment of steady state thermal conduction down the current leads was accompanied by a transient ion current.⁽⁴⁾

The effect of the current leads has been minimized in the present design by reducing the cross section of the portions adjacent to the boat. To accomplish this, the boat has been made three times as long, but with only the middle third open at the top, loaded with material, and connected to the control bridge by voltage leads.⁽³⁾ The outer thirds are curved around in the final design so that a top view of the assembly resembles a C somewhat, with the convex side nearest the filament. The two platinum voltage leads, also one Pt-Rh thermocouple lead, are 1-mil wires, spot welded to the concave side of the C, at the ends of the active portion of the boat. Special techniques were developed during the fabrication, thermal calibration, and loading of this boat.

The operation of the new boat has been quite satisfactory. Admittedly, the design is a compromise, since there is a possibility that the outer thirds of the boat will become extra sources of ionizable material. In over 1500 hours of operation, there has been no evidence that this has caused trouble.

Transport of Ionizable Material

When a positive ion emitter is used to measure the concentration of a tracer, as in atmospheric diffusion studies, air at atmospheric pressure is drawn through the equipment. With this in mind, positive ion emitters studied in the Laboratory have normally been operated in a gas at atmospheric pressure. Even when this gas is chemically inert, it plays an important role in the process, since the ionizable material and the ions subsequently formed must both travel through this gas.

Accordingly, the ion current depends upon the transport properties of the gas, that is, upon the viscosity, thermal conductivity, and coefficient of diffusion. The rate of transfer of ionizable material from the boat to the filament depends not only upon diffusion, but also upon the convection currents in the gas. Convection is a hydrodynamic process; it depends upon the properties of the gas, and also upon the geometry of the equipment and the temperature distribution.

The effect of the physical properties of the gaseous medium is shown in Table 3, a comparison between helium and argon, two inert monatomic gases, both purified in the same way. The experiments were performed in the same closed vessel. The average filament temperature, the average boat temperatures and the collecting potentials were maintained the same in the two cases. The potential was great enough to eliminate space charge.

TABLE 3
Comparison of ion currents and rates of heat loss in
argon and in helium
(Other conditions held constant)

Item	In helium	In argon	He/A ratio
1. Observed steady state ion currents	9.1×10^{-10} amp	1.6×10^{-10} amp	5.7
2. Calculated relative diffusion coefficient, at 550° K	2.09 cm ² /sec	0.525 cm ² /sec	3.99
3. Calculated relative diffusion coefficient, at 1300° K	8.80 cm ² /sec	2.22 cm ² /sec	3.92
4. Observed filament power dissipation	34.6 watts	10.7 watts	3.23
5. Calculated rate of heat loss by gaseous conduction	19.6 watts	2.4 watts	8.2
6. Calculated loss by radiation	1.5 watts	1.5 watts	1.0
7. Estimated loss by conduction in the leads	3.0 watts	3.0 watts	1.0
8. Calculated loss by free convection	9.5 watts	3.4 watts	2.8
9. Heat loss not accounted for above	1.0 watts	0.4 watts	2.5

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The steady state ion current was 5.7 times as great for helium as for argon (Table 3, Item 1). This is at least partly due to the greater ease with which molecules can diffuse through helium. The material which evaporates from the boat diffuses into a gas stream which is rising vertically, then the vapor diffuses horizontally within this stream so that some of the ionizable particles strike the filament. As a measure of the relative ease of diffusion through the two gases, Items 2 and 3 give the coefficients of diffusion of argon in helium and argon calculated for 550°K (approximately the boat temperature) and 1300°K (approximately the filament temperature). The calculations followed formulas in the Appendix, using force constants* known to give correct values at 0°C.⁽⁵⁾ Of course, the ionizable material is not exactly identical to argon. However, it does not appear to be possible to choose any reasonable set of force constants and mass for the ionizable particles, to make the diffusion rates alone differ by as much as 5.7 to 1. Possible reasons are (a) perhaps quantum mechanical rather than classical methods should be used for helium,⁽⁶⁾ and (b) the diffusion process is superimposed upon a mass flow (convective) process, which may be different in the two gases.

The rate of heat dissipation, like the ion current, depends upon the physical properties of the gas. In order to maintain the same average filament temperature, more than three times as much power is required in helium as in argon (Table 3, Item 4). The greater thermal conductivity is responsible for a large share of this. Values of thermal conductivity used in the calculation of Item 5 were obtained by the formula in the Appendix, with force constants from Hirschfelder, et al.⁽⁷⁾ The variation of thermal conductivity with temperature was taken into account in the calculation.

Heat loss by radiation and by thermal conduction in the leads is independent of the gas, except for a second order effect due to a somewhat different temperature distribution along the filament in the two gases. The loss by radiation was calculated by the usual T^4 law, taking into account the emissivity of platinum.⁽⁸⁾ Heat loss by conduction in the leads was estimated by reference to an experiment performed in vacuum.

The heat loss due to free convection (Item 8) was calculated by use of the empirical relation given in the Appendix, with the constants of the gas evaluated at the temperature of the filament. This procedure, and the results, must be regarded as somewhat questionable. The constant in the empirical relation was obtained by studies on two liquids, water and ethylene glycol.⁽⁹⁾ Its applicability to other materials is based on similarity considerations which are well developed in hydrodynamics but which should be used with caution in comparing liquids and gases. Although the gas next to the filament is by far the most important to the convection process, it is not correct to neglect the variation of the physical properties of the gases with temperature.

*Following Hirschfelder, et al.,⁽⁵⁾ the term "force constants" is applied to the two parameters, σ and ϵ , of the intermolecular potential function. See Appendix.

The process of convection, which has not yet been evaluated in detail, is important in the transfer of ionizable material to the filament, as well as in the transfer of energy away from the filament. It appears to be possible, in principle, to solve the convection problem much more completely, by following the method used by Furry, Jones, and Onsager⁽¹⁰⁾ in the study of the problem of thermal diffusion. Since the required integrals do not, in general, lead to closed forms, numerical integrations would be necessary in order to calculate the complete pattern of mass flow parallel to the filament.

Acknowledgment

Mr. William Prepejchal contributed numerous useful ideas and much careful workmanship, especially in the design, fabrication, and calibration of the new boat.

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Appendix

1. The transport coefficients of the gases were calculated to a first approximation by formulas based on classical collision theory, assuming a Lennard-Jones type of potential function.⁽¹¹⁾

Viscosity of a pure gas:

$$[\eta]_1 = \frac{266.93 \times 10^{-7} \sqrt{MT}}{\sigma^2 J_2(T^*)} \quad \text{g/cm-sec}$$

Thermal conductivity of a pure gas:

$$[\lambda]_1 = \frac{15}{4} \frac{R}{M} [\eta]_1 \quad \text{cal/cm-sec-deg}$$

Coefficient of diffusion of a binary mixture:

$$[D_{12}]_1 = \frac{0.0026280 \sqrt{T^3 (M_1 + M_2) / 2 M_1 M_2}}{p \sigma_{12}^2 J_1(T_{12}^*)} \quad \text{cm}^2/\text{sec}$$

Coefficient of self diffusion:

$$[D]_1 = \frac{0.0026280 \sqrt{T^3/M}}{p \sigma^2 J_1(T^*)} \quad \text{cm}^2/\text{sec}$$

The notation is as follows:

M, M_1, M_2 = molecular weights, g/mole

T = absolute temperature, deg K

R = gas constant, cal/mole-deg

p = pressure, atmospheres

σ = finite value of the internuclear distance at which the potential energy is zero, in angstroms

T^*, T_{12}^* = reduced temperatures (dimensionless), equal to $\frac{kT}{\epsilon}$, $\frac{kT}{\epsilon_{12}}$, respectively, where ϵ is the depth of the intermolecular potential well and k is Boltzmann's constant, measured in units so that ϵ/k is in deg. $J_2(T^*)$, $J_1(T_{12}^*)$ and $J_1(T^*)$ are integrals which measure the deviation of the molecular model from the idealized rigid sphere model.⁽¹²⁾ (The notation of Hirschfelder, et al. was simplified at this one point.)

The Lennard-Jones type potential function used had the form

$$\phi(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]$$

where

r = intermolecular distance. (13,14)

2. Heat transfer by convection was approximated by using the empirical relation given by Jakob⁽⁹⁾ for vertical cylinders

$$N_{Nu} = 0.726 (N_{Gr} \cdot N_{Pr})^{1/4}$$

where

N_{Nu} , N_{Gr} , and N_{Pr} are the dimensionless groups, the Nusselt number, the Grashof number, and the Prandtl number, respectively.

The constant 0.726 was obtained from measurements on water and ethylene glycol in the laminar range. For comparing two gases, the coefficient of heat transfer by convection was written.

$$h = 0.726 \left(\frac{g\beta \Delta T}{L} \right)^{1/4} \left(\frac{\rho^2 \lambda^3 C_p}{\eta} \right)^{1/4} \text{ cal/cm-sec-deg}$$

in which the numerical factor and the first parenthesis are the same for the two gases.

The notation is.

g = acceleration of gravity, cm/sec²

β = volume coefficient of expansion of gas, per deg

ΔT = temperature difference between filament and gas approaching lower end of filament, deg

L = length of filament, cm

C_p = specific heat of gas at constant pressure cal/gram

ρ , λ , η = density, thermal conductivity, and viscosity of the gas evaluated at the temperature of the filament, units as in Appendix, Section 1.

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DRIFT VELOCITY OF ELECTRONS IN GASES

J. C. Bowe

A. Comparison of Experimental Results with Theory

Results obtained in nitrogen and the rare gases have directed interest to a study of the correlation between our measurements and the experimental electron scattering data. Now that drift velocity data within a few per cent accuracy are available, it is possible either to test the validity of the theory which relates the two sets of data, or to infer information concerning the cross sections for elastically and inelastically scattered electrons.

Computation of electron drift velocity curves requires a knowledge of (1) the energy distribution of electrons in a uniform electric field, and (2) the cross section for momentum transfer, $Q_D(\mu)$, as a function of energy. No experimental measurements of the former are available but a large number of independent theoretical derivations predict a Druyvesteyn or a generalized Druyvesteyn distribution. There is also a scarcity of angular cross section data for both elastic and inelastic scattering in the low energy region due to the experimental difficulties which are encountered.

There seem to be only three previous calculations of drift velocity as a function of E/p which take into account the variation of Q_D with energy. These were made by H. W. Allen (1937),⁽¹⁾ by D. Barbiere (1951),⁽²⁾ and by W. J. Graham and A. J. Ruhlig (1954).⁽³⁾

H. W. Allen, using the theory as developed by Morse, Allis, and Lamar⁽⁴⁾ and the scattering data of Ramsauer and Kollath,⁽⁵⁾ calculated drift velocity curves for helium, neon, and argon for $E/p = 0$ to 3.5 volts/cm/mm Hg, taking into account the effects of inelastic collisions. Unfortunately, this paper is too sketchy in parts to permit one to know exactly what was done. For example, it is not known at which values of E/p the effects of inelastic collisions were included in the computation. The temperature at which Q_D is evaluated is not stated, thus making the E/p values uncertain by about 10%. Also, it is not evident what procedure was followed to represent the scattering data as a function of E/p since such data are originally determined as a function of electron energy.

In taking into account the effects of inelastic collisions, the following approximations were made: (1) the elastic collision cross section is reasonably constant, and (2) since the inelastic collision cross section is not known, the distribution function is arbitrarily cut off at an energy E_1 , which lies between the first resonance potential and the ionization potential. The value of E_1 , is then used as an adjustable parameter to give the best fit between theory and experiment. An adjustable parameter of this type makes it

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difficult, if not impossible, to correlate the scattering data with the drift velocity data. The value of the adjustable parameter was chosen to fit the calculated "temperature" curves with the measurements of Townsend^(6,7) and Bailey.⁽⁸⁾

Allen's curve for helium, however, is in remarkably good agreement with our measurements. In argon, the agreement is good. In neon, the curve has the same general shape as ours but is much lower.

D. Barbieri, using the theory as developed by T. Holstein⁽⁹⁾ and the scattering data of Ramsauer and Kollath,⁽⁵⁾ calculated drift velocity curves for helium and argon for $E/p_0 = 1$ to 4 volts/cm/mm Hg, taking into account inelastic collisions by assuming a constant cross section for all energies above the excitation potential. Curves for the limiting values of excitation cross section equal to zero and infinity were computed. The experimental curves lie within these limits.

Graham and Ruhlig calculate drift velocity curves for helium, neon and argon for values of $E/p_0 = 0.1$ to 2 volts/cm/mm Hg, following the method of Morse, Allis and Lamar,⁽⁴⁾ and taking into account elastic collisions only. A set of composite scattering data compiled by Westin⁽¹⁰⁾ was used in the calculations. The curve for helium has the same shape as ours, but lies lower, indicating that the values of $Q_D(\mu)$ used were too large. The curve for neon agrees extraordinarily well with our curve. The curve for argon is in agreement with our measurements, but unfortunately was computed over the limited range of $E/p_0 = 0.1$ to 1.0 volts/cm/mm Hg.

B. Work in Progress

Calculations of drift velocity curves have been initiated by Rose A. Carney* with the following objectives in mind: (1) extend calculations to lower values of E/p , (2) attempt to determine the sensitivity of the drift velocity data to the presence of inelastic collisions, (3) with the aid of the experimental curves and the calculated curves, attempt to extract information concerning the inelastic collision cross section, (4) calculate a drift velocity curve for nitrogen (there is at present no calculated curve for nitrogen whereas experimental agreement among various investigators is within a few per cent), and (5) calculate a drift velocity curve for helium.

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C. Experimental Progress

A gridded chamber was assembled for the purpose of obtaining measurements on the diffusion of the electron cloud. Such information would allow computation of the electron "temperature." The chamber has not as yet been tested.

A Tektronix 531 oscilloscope was procured and used to repeat measurements with purified helium. Results were identical with those obtained using the Tektronix 517.

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DELAYED NEUTRONS FROM 15.5-sec Br^{88} *

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

A description is given of a gas-flow system which permits quick chemical isolation of bromine fission fragments. Using this system, Br^{88} was found to be a delayed neutron precursor resulting from the fission of U^{235} . The half-life measured by neutron counting is 15.5 ± 0.4 sec, in agreement with that previously measured for Br^{88} by radiochemical techniques. It is a relatively abundant source of delayed neutrons, producing per fission about three times the number of neutrons in the 54-sec group having Br^{87} as a precursor. Br^{88} is the first definitely established case of a delayed-neutron precursor of odd proton and odd neutron number. Qualitative arguments are given to show that such nuclei are about as likely to be delayed neutron precursors as are those of odd Z and even N, although only the latter have been reported previously.

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CALIBRATION OF DIBUTYL PHOSPHATE PROCEDURE FOR ISOLATION OF Th^{228} FROM BONE

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A series of calibration tests have been performed for the dibutyl phosphate method of isolating Th^{228} from solutions of ashed bone. As outlined previously,⁽¹⁾ the method consists of extracting Th from a 2M HNO_3 solution into dibutyl phosphate, allowing 3.64-d Ra^{224} to grow into the organic phase, milking the Ra^{224} into a HNO_3 solution, and precipitating the Ra^{224} with BaSO_4 on a platinum planchet to provide a source suitable for counting alpha particles. An empirical relationship between the counting rate observed for the Ra^{224} preparation and the disintegration rate of the Th^{228} in the original sample is required because of possible losses in the analytical procedure and self-absorption of alpha particles in the sample mount. Assay of pure Th^{228} standards is difficult because of radioactive decay products, and the most direct procedure of obtaining Th^{228} of known disintegration rate seems to be preparation from a weighed amount of Th^{232} of known age. By "age" we mean the time elapsed since the thorium was purified from its ore.

Two samples of Th^{232} of reasonably well known ages were obtained from the Massachusetts Institute of Technology through the kindness of Dr. Robert A. Dudley. An average of 0.93 α counts per minute (cpm) of Ra^{224} per $\mu\mu\text{c}$ Th^{228} was obtained with a standard deviation of 0.04 units for 10 analyses of these samples using approximately 2 $\mu\mu\text{c}$ amounts of Th^{228} . These analyses were performed both with and without ashed bone present. Because of the growth of radioactive daughters of Ra^{224} , the counting rate actually observed for the BaSO_4 mounts is about 3 α cpm per $\mu\mu\text{c}$ Th^{224} . Thus 0.01 $\mu\mu\text{c}$ Th^{228} provides a counting rate almost equal to the background rate of 0.04 cpm, which is obtained for the counters.

A locally purchased sample of beef bone used for testing the procedure was found to contain 0.25 $\mu\mu\text{c}$ Th^{228} per gram of ashed bone during the summer of 1956; however, subsequent analyses have shown growth to a value of 0.37 in April and May of 1957. Since the presence of Ra^{228} is indicated, analyses for this isotope will be made. If these values are not typical for cattle, it would be of interest to determine the geographical origin of this particular animal.

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MASS PER UNIT VOLUME OF BONE MINERAL

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

Microradiographs of bone sections delineate quite markedly, small changes in mineral density. This is particularly evident in radiographs of cross sections of cortical bone where haversian systems of various mineral densities are located adjacent to one another. By measuring the photographic density of these systems and comparing these densities with those recorded under a step wedge on the same plate, it is possible to compute the mass of mineral per unit area of the densitometered regions. The theoretic basis of this method has been described in a previous progress report.⁽¹⁾

To obtain the value of the mass of mineral per unit volume of bone requires one additional piece of information, the thickness of the section. This is not as easy to obtain as might be assumed, for one has no sure way of proving that there are not fluctuations in thickness across a cut or ground section. The ideal instrument would measure the thickness of the sections at the same location as it was densitometered, and would give the average thickness over the same area as the densitometer beam illuminated.

Now in operation is an instrument which approaches this requirement; it measures the maximum thickness of a 40 micron diameter spot, while the densitometer employs a 10 micron diameter spot. It works as follows: the substage condenser of a Leitz Ortholux microscope is removed, and on it is mounted a cone. The end of this cone was ground flat and smooth, as viewed through a 22 x Ultropak objective (surface illumination). The diameter of the flat end is 40 μ . Employing the vernier of the microscope, we can reproducibly locate the position of proper focus of this surface within plus or minus one micron in a series of trials.

To measure the thickness of any given region of a bone section, the section is held horizontally over the point, and the point raised until it touches and lifts the bone. The objective lens is focused on the top surface of the bone, then the bone is removed, and the objective lens moved down to focus the top of the pointer. The difference in the two vernier readings is the thickness of the desired region of the bone section. Such a reading is made for every location densitometered.

In practice, the intensity of the x-ray beam is not uniform over a very large area, so that the densities of regions relatively close together are compared with a step wedge placed very near to these regions. A most convenient method is to make a hole in the step wedge and place a small segment of bone into this hole, so that the radiograph of the bone is within the radiograph of the wedge. If the hole in the wedge is made in the step under which

the photographic density is most nearly equal to that of the bone, the uncertainty due to the variation in x-ray intensity is very nearly eliminated. This approach also minimizes difference in response of the photographic emulsions due to unequal thicknesses over various portions of the plate.

Repeated measurements have shown that the intensity of our x-ray tube varies across the field in one direction, but that it is remarkably uniform in a perpendicular direction. For example, if the tube is so oriented that the variation in intensity is a maximum in the vertical direction across the beam, the variation in intensity is a minimum in the horizontal. Hence, the step wedge is mounted to lie horizontally in the beam, so as to obtain minimum variation in intensity from step to step. Since the bone lies within the step closest to its average density, and since it is seldom necessary to obtain a reading of more than two steps above and below this density, the excursion of the densitometer beam from the bone need not be more than a few millimeters.

The following table includes the results obtained from a series of femur sections from dogs of various ages.

TABLE 4

Highest and lowest values of mineralization found in
cortical bone sections from the femur of dogs of
various ages.

Age of dog	Least dense bone	Most dense bone
6 weeks	1.16 g/cm ³	1.54 g/cm ³
1 year	1.10	1.52
5 $\frac{1}{2}$ years	1.23	1.53
11 years	1.41	1.71

The results indicate that the variation in degree of calcification does not vary widely with age. That is, newly formed bone has a minimum density varying from 1.10 g/cm³ in a year-old dog to 1.23 g/cm³ in the 5-year-old dog. The fact that no bone of this low density was found in the 11-year-old dog may be due to the fact that there is relatively little rebuilding in the older animal, so that the chance of finding a closing haversian system with new bone is much lower. The degree of mineralization of the most dense bone is seen to be constant from the puppy to the 5-year-old animal, with a noticeable increase in the oldest animal.

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It must be remembered that these figures refer to the degree of mineralization of bone volumes 10 microns in diameter and 100 microns long. In the puppy there is very little highly calcified material, while in the oldest animal there is very little bone of low calcification.

Robinson and Elliott⁽²⁾ have shown how the density of dog bone varies with its water content. Puppy bone was shown to have a lower density, and lower mineral density, than the bone from the older animals. The fact that we find the absolute values of mineral densities to be the same in puppies and older animals does not contradict this, as can be seen from an examination of Figs. 14 and 15. In the puppy bone, Fig. 14, there is a good deal of bone of low density, and the bone is seen to be quite vascular, while the bone of the 5-year-old animal is composed primarily of bone of high density, with greatly reduced vascularity.

Fig. 14. Microradiograph of a section of femur diaphysis of a 6 week old dog.

Fig. 15. Microradiograph of a section of femur diaphysis of a $5\frac{1}{4}$ year old dog.

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THE MECHANISM OF RADON RETENTION IN BONE

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

When radium is incorporated in bone, its daughter, radon, is observed to escape from the bone and the animal. The fraction escaping depends upon the age of the deposit and the kind and age of the animal. It has been assumed that this escape was due to the recoil of the radon atom,⁽¹⁾ as the radium alpha particle was emitted, which allowed the radon atom to escape from the hydroxyapatite crystal into which the radium had been built. After reaching the blood stream, the radon atom would be swept out of the bone and into the lungs in a time very short compared to its half-life, 3.84 days.

The range of the recoiling radon atom is probably 200 Å in hydroxyapatite, a value computed from Bohr's equation⁽²⁾ for the stopping of a heavy ion with low initial velocity. Recent work of Duncan⁽³⁾ has verified the validity of this equation and indicated that the Flugge and Zimens equation⁽⁴⁾ for recoiling atoms, in which the range was dependent upon the square of the recoil energy, was probably incorrect.

In an effort to understand the physical details of the escape mechanism, a one-year-old mongrel dog was injected with Ra^{226} ($4.4 \mu\text{C}/\text{kgm}$) and sacrificed one month later. Twelve cortical bone sections one cm long were cut from the femur and tibia, the marrow cleaned out, and the sections stored in ethyl alcohol for a month. Before any measurements were made, the sections were soaked for 24 hours in acetone to remove fat. These twelve sections were divided into four groups of three each, and subjected to the following treatments:

- Group 1: Controls, left in air at room temperature.
- Group 2: Temperature studies, one held at 0°C , one at 50°C and one at 250°C .
- Group 3: Ashed at 600°C overnight.
- Group 4: Ethylenediamine extracted for 5 days.

Radon retention was determined by measuring with a NaI(Tl) crystal the relative intensities of the radium gamma ray (188 keV) and a RaB (351 keV) and a RaC (607 keV) gamma ray, as described by Gustafson and Marinelli.⁽⁵⁾ The system was calibrated by precipitating radium as the sulphate, sealing it in a glass vial, and measuring the ratios of the gamma rays as a function of time. One tenth μC of the same radium source as was employed for the injection of this animal was used for the calibration, this being very close to the quantity of radium in each one-cm length of shaft. The method of measurement is highly sensitive at low values of radon retention, below 40 per cent, but as the value of retention approached 100 per cent, this method becomes quite insensitive. However, since the radium content of the bone is constant,

the intensity of the RaC gamma ray (607 kev) is a direct measure of the radon retention. The relation between the 607 kev gamma intensity and the radon retention is obtained at a low value of radon retention by the ratio method.

The results on the four groups of bone sections are as follows.

- Group 1: Constant retention of 22 per cent for 2 months.
- Group 2: 0° C for 17 days: retention dropped to 21 per cent, returned to original value at room temperature.
50° C for 14 days: retention increased to 35 per cent, did not return to original value at room temperature.
250° C for 17 days: retention increased to 40 per cent, maintained this value at room temperature.
- Group 3: Retention increased in one case to 84 per cent, in the other two to 92 per cent.
- Group 4: Retention increased to values of 50, 59, and 60 per cent.

The size and shape of the crystal of bone are in controversy, with the electron microscope showing both a tabular form, with dimensions of $100 \times 200 \times 500 \text{ \AA}$ (6) and a cylindrical form, $200 \text{ \AA}$ in length and $65 \text{ \AA}$ (7) in diameter.

In either case, but particularly the latter, the recoiling radon atom must escape from the crystal in which it originated. However, its range in the organic material between crystals will be greater than in the crystal, (for example, in water the range is $550 \text{ \AA}$). Since we know that a given volume of mature bone is approximately 40 per cent hydroxyapatite, there exists a very good probability that the recoiling atom will not stop in the organic material but will enter an adjacent crystal. If the organic material were to be removed and only empty spaces left between the crystals, then every recoiling atom would come to rest in a crystal from which it could escape only by diffusion.

In the case of the bones ashed at 600°C , the water, CO_2 , and organic material are removed, so that recoil from one crystal into another would take place in every case. However, it is known that the elevated temperature will result in crystal growth, (8) so that the resultant effect must be due to both the above effect and increased retention due to failure to recoil out of the parent crystal. While the bone is at 600°C , the radon retention is observed to be very close to zero, and only increases to 90 per cent retention after the bone is cooled.

The ethylenediamine treated bone did not retain as much radon as the ashed bone; this may be interpreted as being due to less crystal growth (the temperature is not above 115°C) as well as less removal of material by this method.

After the sections had been in the extraction soxhlet for five days they were placed in running water for two days. At the end of this period the radon retention was measured and found to be 22 per cent. On succeeding days radon retention was found to increase until it reached its final value of 60 per cent. Since our method of measurement depends upon the ratio of the number of daughter products of radon present to the number of radium atoms, and since the radon daughters grow into a sample with the half-life of radon, a rapid increase in radon retention produces a measured increase of the form $(1 - e^{-\lambda t})$. Analyzing the increased radon retention of the ethylenediamine extracted bone with this in mind, it becomes apparent that the radon retention did not increase during the period of extraction or washing, but only after the bone was allowed to dry in the air.

This effect was found to be partially reversible, for if the above-described bone was placed in water for several days, and the pressure cycled between the vapor pressure of water and the atmosphere, the radon retention dropped to 30 per cent, then returned to 60 per cent after drying in the air.

On the basis of the recoil mechanism, this behavior can be understood by considering that when the bone is dry the radon recoils must stop within a crystal, for their range in air is several thousand times their range in the bone crystal. When water is present between the crystals, the probability of the recoiling atom stopping between the crystals increases considerably, and thus the radon retention decreases. However, the role of adsorption of radon on the crystal surfaces remains to be evaluated, and experiments are now in progress to determine what fraction of the retained radon is due to this process.

This work has shown that radon can escape from a one-centimeter-long diaphysis cross section of bone, without the benefit of a circulatory system, with the same efficiency as it escapes from the whole animal. Thus the same ratio of daughter products exists in radium burdened bone in vitro as in vivo. Miller and Hoecker⁽⁹⁾ essentially found this to be true in their study of radon retention in rat bone.

From the experiments on radon retention performed by Mays, et al.⁽¹⁰⁾ it appears that freezing bones without alcohol or acetone soaking will result in increased retention of radon. We would predict, however, that as long as the bones were not subjected to elevated temperatures, acetone soaking before they were embedded for autoradiographic studies would return the radon retention to its original value, for the increase may well be due to the high solubility of radon in fat.

To determine if embedding increased radon retention, a bone embedded in methyl methacrylate was cut into 25 slices, each 100μ thick, and pressed against a photographic plate. At the end of two weeks the radon retention of the 25 slices was 35 per cent instead of the normal 22 per cent.

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This indicates that the alpha particles found in an autoradiograph of an embedded bone section are due to the radium content of the bone plus 35 per cent of the short-lived daughter products. In computing the dose to the bone from such an autoradiograph, it has often been assumed that the observed alpha tracks came from radium plus 100 per cent of the daughter products (except RaF). It is now apparent the radon retention will be strongly dependent upon the method of preservation of a bone sample.

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BACKGROUND MONITORING

H. A. May

Continuous monitoring of the ionization in air from natural background sources has been carried on for approximately one month. Ion current is collected in an unsealed 4.7 liter chamber and recorded with a vibrating-reed electrometer and recording galvanometer. The chamber is approximately air equivalent, consisting of 1/8-in. thick Bakelite walls, having 38% of the surface area cut away, and supporting an aluminum foil (5 mg/cm²) wall. It was not deemed feasible to locate it completely in the open away from the Laboratory buildings without requiring considerable effort to make the system weatherproof. Location to date has been in a 30 x 30 x 60 ft long aluminum balloon hangar, with the chamber supported 95 cm above the 6-in. concrete floor. All equipment is placed on a transfer table and is readily moved outdoors without disturbing continuous operation. No change in reading with location has been detected.

A 1½-in. x 1-in. NaI (Tl) crystal and a 3-in. photomultiplier located 15 cm above grade outdoors in a weatherproof housing, driving a linear amplifier, logarithmic counting-rate meter and recorder, is used to give an indication of any change in air or surface gamma activity, such as might be expected from A⁴¹ originating from the nearby reactors, or from fallout from weapons tests. The crystal detector records continuously, indicating qualitative changes in ionization level; the ion chamber may then be easily moved outdoors for quantitative determinations. Average integral counting rate about 150 kev is 3000 c/m.

The following preliminary results have been noticed:

(1) The ion chamber readings tend to follow a daily cycle, caused principally by fluctuations in the atmospheric radon level, which may readily enter the ionized volume. A minimum reading generally occurs between 1200 and sunset, with a gradual rise to a maximum between dawn and 0900. Extent of the maximum depends upon local meteorological conditions, specifically upon extent of temperature inversion, and following precipitation may remain practically constant for 24 hours. No obvious correlation with temperature or atmospheric pressure has been noted. These observations completely agree with the findings of Lucas, Kulhanek, et al.⁽¹⁾ Table 5 summarizes data accumulated to date:

TABLE 5

Average daily minimum	11.3 ± 0.49 ion prs/cm ³ /sec
Average daily maximum	21.8 ± 0.48
Minimum single observation	6.0
Maximum single observation	32.5

The data are uncorrected to standard temperature and pressure. Statistics are expressed as the standard deviation from the mean. To convert to mr/year, multiply the figures by 15.1.

(2) With favorable local meteorological conditions, i.e., wind from the south at very low velocity so that atmospheric mixing and diffusion are minimized, the counting rate from the small NaI crystal has been observed to double, presumably as a result of air-borne activity from CP-5, EBWR, the cyclotron, or other experimental assemblies in the Laboratory. Three such events have been observed during one overnight interval, with the increased counting rate persisting for about 30 minutes for the longest "burst." This seems to have been so short that no detectable amounts of activity were introduced into the iron room; hence positive verification of the suspected cause (namely, A^{41}) by scintillation spectrometry⁽²⁾ has not been accomplished.

Comments

Small changes in background ionization level resulting from fallout are obscured by widely varying radon levels. Thus, determination of the background ionization level from radioactive elements present within and upon the ground plane appears to be feasible only if the varying radon level is known from separate radon-in-air analyses. The minimum levels reported above are higher than those previously reported elsewhere, for comparable locations.⁽³⁾ The extent to which these minima are influenced by residual radon in the air, or by systematic errors as yet unidentified, is unknown. Further experiments, including sealed chambers, are planned to evaluate these variables.

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THE ABSORBANCE CHANGE OF IRRADIATED
ORGANIC SOLVENTS. PART IV*

I. B. Berlman

In Part III, the absorbance change produced in aerated xylene samples by massive Co^{60} gamma-ray doses is discussed. In this section the study has been extended to include results on evacuated xylene samples. Some work has also been done on irradiating frozen xylene.

Xylene was irradiated in evacuated and sealed glass vials with massive doses of Co^{60} gamma rays. Each vial contained approximately 8 cc of doubly distilled p-xylene. Absorbance measurements were made on a Beckman DK-2 spectrophotometer,** using 1 cm absorption cells. Unirradiated xylene was used as the reference solution. Both irradiated sample and unirradiated reference were aerated before the absorbance measurements were made. Curve II of Fig. 16 indicates the type of absorbance curve obtained under evacuated conditions; curve I, in contrast, is the type of curve obtained for a similar sample which was aerated before irradiation. Both samples were given the same dose of approximately 1.3×10^6 r. Furthermore, the absorbance of the aerated samples at a given wavelength in the region of 3500 Å increases with dose in an almost linear fashion (see Part III), but under evacuated conditions the absorbance rises much more rapidly than the first power of the dose. The exact relationship between the absorbance and the dose and a general explanation of the absorbance curves is being studied further. However, some explanation can be given for the difference in the 3300 Å region between the two curves of Fig. 16 and for the different relationship between absorbance and dose for the aerated and evacuated samples. Assuming that absorption in the 3300 Å region is the result of the formation of large molecular products, then the lesser absorption exhibited by the aerated sample can be explained by the scavenger effect of the oxygen present, by which certain radiation radicals are in some way prevented from becoming parts of larger molecules. This explanation seems to be in accord with "oxygen effects" in other experiments (1)

*Part I appears in Report on Bio-Med and Biophysics Programs, ANL-5518, p. 8 (1956).

Part II appears in Rad. Phys. Div. Semiannual Report, ANL-5596, p. 3 (1956).

Part III appears in Rad. Phys. Div. Semiannual Report, ANL-5679, p. 5 (1957).

**Made available through the courtesy of Warren Buck of the Physics Division.

Fig. 16

Fig. 17

Since the absorbance curve for an evacuated sample increased so rapidly with dose in the 3200-3600 Å region, it seemed possible that for a large gamma-ray dose a sufficiently large yield of molecular products would be formed to permit the investigation of the possibility of fluorescence of the products. If the radiation products formed in the vacuum-irradiated samples fluoresced, the emission band of the fluorescence would be on the long wavelength side of the absorbance band. A xylene sample that had received approximately 10^8 r was available. On illuminating this sample with the 3650 Å region line of mercury, an emission band appeared in the 3700-4700 Å. The measurement was made on the Beckman DK-2 spectrophotometer. More exact information concerning the emission spectrum must wait until the photomultiplier of this instrument is calibrated with respect to its wavelength response.

Several of the evacuated and sealed xylene-containing vials were frozen by packing them in dry ice. They were then irradiated while still in the solid state. After irradiation the vials were allowed to reach room temperature and the absorbance measurements were made on the liquid phase. The absorbance change is reduced as is indicated by Curve I of Fig. 17. Curve II is the absorbance change for an evacuated sample irradiated at room temperature. Irradiated ice has fewer radiation products than irradiated water.⁽²⁾ The reduced effects produced in the solid state may be due to dissociation effects.⁽³⁾ Xylene molecules may be dissociated by ionizing radiation, but the products are frozen in place so that on warming they are still close enough to recombine. This interpretation of the results is strengthened by the fact that frozen xylene that is irradiated while in the solid state produces a large number of low energy light flashes on warming to the melting point. The light is assumed to be due to the recombining products and/or radicals. Just freezing and melting xylene does not result in the emission of a measurable amount of light. Whether this thermoluminescence is due to just a temperature rise of the solid or must also involve a change of state (melting) has not as yet been determined.

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THE HUMAN SPECTROMETER

I. Introduction

C. E. Miller

During the past six months, the Human Spectrometer has been devoted largely to the study of certain features of Cs^{137} fallout and its uptake by humans. Thus, the gamma-ray body activities of 11 subjects from Central and South America and of 7 Marshallese have been measured and the trend of cesium burden of our control subjects has been brought up to date. In addition, extensive measurements have been made of the Cs^{137} contents of filter papers collected from an air monitoring program dating back to 1953.

The radium body burdens of two former dial painters have been determined. Studies of the relationship between total and exchangeable K in humans have been started. The gamma-ray spectrum emitted by the liver of a Tridacna clam collected in the Bikini lagoon is given.

The low-energy, gamma-ray background of the Iron Room has been reduced by about 20 per cent with the addition of 1/8 in. of lead to the floor and one wall.

II. In Vivo Measurements of Cs^{137}

C. E. Miller and L. D. Marinelli

Cs^{137} Trends in Humans

The build-up of Cs^{137} as a function of time in four control subjects was reported in graphical form in the preceding Semiannual Report. (1) Because of its obvious importance, all pertinent data from which these curves were plotted have been re-examined in the light of subsequent experience. Figure 18 consists of new curves based on these data incorporating the following changes and additions.

(1) The January, 1956, point on the CEM curve has been dropped. The data had been determined using a 4-in. by 4-in. NaI(Tl) crystal on loan from the Harshaw Chemical Company through the courtesy of E. C. Stewart. This point had been incorrectly plotted since the crystal efficiency factor for the standard 4-in. by 1½ in. crystal was used.

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Fig. 18. Cs^{137} trends in humans

(2) The February, 1956, point on the CEM curve has been dropped since an exhaustive examination of the operating log book disclosed that the pulse height analyzer had malfunctioned on other runs during that day.

(3) The June 28, 1955, point on OJS has been corrected for an arithmetical error.

(4) Data taken from April 1, 1955, until July 1, 1955, were not included in the original graph since the subjects had been measured while wearing their own street clothes which may have carried contamination. Since these values are the only existing measurements of Cs^{137} body burdens during this time, the data have been re-examined. In cases where the K body burden has agreed with subsequent values taken under standard conditions, the early data have been assumed to be correct and have been added to the curve.

(5) Curves for five additional subjects have been added. Several runs which gave higher than normal potassium values for female subject KG were dropped. These high values were discovered to be due to the subject's wearing a foundation garment which had a measurable amount of potassium.

(6) The curves have been extended to June 1957.

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We conclude that the average Cs^{137} body burdens have remained essentially constant since the spring of 1956. The increase of LS's burden, for example, is offset by a decrease in subject KG's burden. These changes can be ascribed to dietary fluctuations.

The estimate of radioactive contamination of the human body under continual intake of radioactive foods and water would be greatly aided by the establishment of a significant parameter describing the radioactivity of both. Thus, in the last few years, great efforts have been expended in establishing whether the Sr^{90}/Ca ratio in the diet leads to a reliable evaluation of Sr^{90}/Ca in the human skeleton. Our first efforts along these lines concerning Cs^{137} led to the speculation as to whether the Cs^{137}/K in the diet could be regarded as a reliable parameter for the estimate of Cs^{137} body burden. Contrasted to the findings with the Sr^{90}/Ca , the Cs^{137}/K in the body was found to be 2.2 times the ratio in the urine of a single individual. (ANL-5679, p. 29-30, June-December 1956).

Taking advantage of the constancy of Cs^{137} burden experienced in the last year, the excreta of two of the control subjects (CEM and OJS) were collected for several days and analyzed for Cs^{137} and K. The pertinent data obtained are shown in Table 6.

TABLE 6

Subject	CEM	OJS
g K/day in urine	2.53	2.0
g K/day in feces	.65	.4
$\mu\mu\text{c Cs}^{137}/\text{day}$ in urine	33.5	40
$\mu\mu\text{c Cs}^{137}/\text{g K}$ in urine	13.2	20
ratio $\frac{\text{Cs/K in body*}}{\text{Cs/K in urine}}$	2.6	1.9

*This ratio has been called assimilation ratio
in the body of the paper.

An assimilation ratio of 1.9 was found for subject OJS and 2.6 for subject CEM. The average value is 2.2 which is in agreement with the previously reported value for RER. Owing to the low radioactivities due to K^{40} and Cs^{137} in the feces of these subjects and the relatively high content of Ra^{226} therein, due to natural intake, it has not been possible to determine the fecal Cs^{137}/K by simple gamma ray analysis alone. Efforts to measure this ratio are currently under way. Additional pertinent information is given later in the body of this report, concerning studies on Marshallese subjects of relatively high Cs^{137} content, who, however, could not be considered in equilibrium with their environment at the time of measurement.

World Wide Values

The Cs/K ratios present in the 13 Argonne control subjects (Chicago) are given in Table 7. These values range from 22 to 50 $\mu\mu\text{c Cs}^{137}$ per gram of body potassium with an average value of 36 $\mu\mu\text{c Cs}^{137}/\text{g K}$, irrespective of sex. The spread from the lowest to the highest value yields a ratio of 1 to 2.3.

This ratio was also determined for 11 Central and South American subjects during their visit to our Laboratory. These data are tabulated in Table 8 according to geographical latitude. The values vary from 7 to 21 $\mu\mu\text{c Cs}^{137}/\text{g K}$ with an average value of 14.6 $\mu\mu\text{c Cs}^{137}/\text{g K}$ and yield a spread of 1 to 3.

Since the spread of each group is approximately the same and the dietary influence is pretty well established, the conclusion could be drawn that the subjects with the larger Cs^{137} body burdens were the larger consumers of meat and dairy products as is invariably the case with North American control subjects. The diets of the Argentine subjects, however, include meat and dairy products in quantities wholly comparable to the U.S.A. diets and, moreover, their Cs^{137} body burdens are the lowest of the group. This fact points to the possibility of a real difference in hemispherical fallout.

The values found for people from other areas of the world are given by Table 9. The average for the European subjects is 32 $\mu\mu\text{c Cs}^{137}/\text{g K}$, essentially identical to the Chicago value. The Asian subjects who include little or no milk in their diets are significantly lower.⁽¹⁾

The similarity between the European and Chicago and continental U.S.A. values⁽²⁾ suggests that the fallout in the Northern Hemisphere is approximately uniform.

Although the evidence gathered on a few subjects is certainly not conclusive, it does, nevertheless, focus attention on the various factors affecting the geographical patterns of fallout which have been considered for the case of Sr^{90} .⁽³⁾

The Cs^{137} Atmospheric Content as a Function of Time

In conjunction with our studies of the time trend of Cs^{137} content of humans at Chicago it was thought advisable to inquire into the Cs^{137} content of the atmosphere at the same site. The gross amount of radioactivity present in the air has been measured at Argonne National Laboratory for the past several years. Air pumps located at several different sites at Argonne pull approximately 15 m^3 of air per hour through HV-70 counting filter papers. The filters from some air pumps were changed daily and some were changed weekly. The weekly filter papers were bunched into monthly groups and the gamma-ray spectra of each bundle obtained

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TABLE 7

CHICAGO SUBJECTS (42°N) DURING JUNE 1957

FEMALES	$\mu\mu\text{c Cs}^{137}/\text{gK}$	MALES	$\mu\mu\text{c Cs}^{137}/\text{gK}$
K.G.	22	R.R.	25
L.S.	31	W.P.	28
I.S.	33	B.O.	31
C.L.	36	O.S.	36
J.J.	43	H.M.	41
N.K.	50	C.M.	41
		E.M.	45
AVERAGE 36		AVERAGE 35	

TABLE 8

DATA ON FOREIGN SUBJECTS

COUNTRY	LATITUDE	DATE	$\mu\mu\text{c Cs}^{137}/\text{gK}$
CUBA	22°N	5-23-57	20.0
EL SALVADOR	13°N	"	8.3
COLOMBIA(4,000')	4°N	"	21.0
ECUADOR(10,000')	0°N	"	13.0
BOLIVIA (13,000')	15°S	"	13.0
BRAZIL	20°S	"	16.0
"	30°S	"	21.2
URUGUAY	34°S	"	11.6
ARGENTINA	35°S	"	7.3
"	35°S	"	7.6
CHILE	37°S	"	22.0
AVERAGE			14.6

TABLE 9

DATA ON FOREIGN SUBJECTS

	COUNTRY	SUBJECT	DATE	$\mu\mu\text{c Cs}^{137}/\text{gK}$	$m\mu\text{c Zn}^{65}$
EUROPE	ENGLAND	T	5-16-56	33.5	
	"	R	7-13-56	34.7	
	FRANCE	J	9-21-56	32.7	
	BELGIUM	F	10-30-56	26.5	
	SWEDEN	N	11-29-56	32.2	
ASIA	AUSTRALIA	P	3-27-57	50.0	
	INDIA	V ₀	12-18-56	18.9	
	"	V ₀	"	20.8	
	JAPAN	S	7-26-56	24.5	53.2
	INDONESIA	S	8-10-56	13.9	52.1
OCEANIA	"	M	"	8.5	
	MARSHALL IS.	RC(US)	4-5-56	26.7	4.2
	"	TT(US)	"	31.7	5.3
	"	*10	"	65.0	29.5
	"	6	"	69.0	73.0
	"	9	"	73.2	29.5
	"	4	"	79.0	29.5
	"	7	"	95.5	62.1
	"	5	"	1610.0	482.0
	"	8	"	2720.0	229.0
UNITED STATES AVERAGE				34.0	

Various amounts of 285-day Ce^{144} - Pr^{144} , 1-year Ru^{106} - Rh^{106} , 27-year Cs^{137} -2.6 m Ba^{137} , and 65-day Zr^{95} - Nb^{95} are present on the filters. The 65-day Zr^{95} - Nb^{95} completely masks the presence of the Cs^{137} on all filters collected since August, 1956. The amounts of Cs present on these latter papers have not been determined to date, although the spectra will be peeled apart when time permits. Unfortunately, all papers collected prior to March, 1953, except for the month of June, 1952, are missing. The amount of Cs^{137} per paper, expressed in arbitrary units, is given by the upper curve of Fig. 19.

Fig. 19. Cs^{137} activity in filter papers

The Cs^{137} atmospheric contents as shown by Fig. 19, increased by a factor of about 6 in the spring of 1953 and remained at this higher level until January 1955, except for an upward excursion immediately following Operation Castle, March 1954. It is of decided interest that the atmospheric Cs^{137} content, at Chicago at least, did return to its 1953 value in the latter part of 1954. The 1955 excursion is due to the 14 U.S.A. continental tests (Operation Teapot) and the early 1956 excursion is probably due to a combination of the American tests in the Pacific and the Russian tests. The various Russian, English, and American tests have taken place with such regularity since 1955 that the Cs atmospheric content has not returned to its 1953-1954 plateau value. The Cs present on the papers collected since August 1956 is completely masked by Zr^{95} - Nb^{95} from Russian tests during the last half of 1956; efforts will be made to measure it by nondestructive tests.

In view of its possible usefulness, the average shape of 3 cesium trend curves (Subjects OJS, KG and LS) has been plotted as shown at the lower right hand curve of Fig. 19. It is hoped that upon closer examination of the various factors influencing fallout and its transmittal to man, it will be possible to offer, at some future date, an explanation of the behavior of the lower curve.

III. Measurements on Some Residents of the Marshall Islands

C. E. Miller and O. J. Steingraber

Seven normal, healthy male residents of the Marshall Islands, under the medical supervision of Dr. Robert A. Conard (RC, Table 9) of the Brookhaven National Laboratory and accompanied by Mr. Jack Tobin (JT, Table 9) District Anthropologist for the Marshall Islands, were flown to this Laboratory on April 4, 1957 for whole body gamma-ray spectroscopy.

Details on mode and extent of exposure, body content from bioassay, and clinical course are described elsewhere.^(4,5)

Four of these men (Nos. 4, 7, 6, 9, Table 9), residents of Rongelap, and two others (Nos. 5 and 8), residents of Uterek, were among those evacuated to Kwajelein from the area of heavy fallout some 48 to 78 hours following the thermonuclear detonation of March, 1954. About 3 months later, Nos. 5 and 8 were returned to Uterek and the others moved to Majuro. The seventh man (No. 10) from Majuro, not exposed, served as a control although his diet was not typical since he worked at Trust Headquarters as did Mr. Tobin (No. 11, Fig. 20).

Each subject's gamma-ray spectrum (see Fig. 20) was obtained using the standard chair position. Since the body burdens of the two Uterek subjects (Nos. 5 and 8) were relatively high, their gamma-ray spectra could be accurately measured utilizing the one-meter arc technique. The body burden of subjects 5 and 8 was then calculated using the one-meter arc technique described in an earlier report ⁽¹⁾ The one-meter arc net $K^{40} + Cs^{137}$ spectrum of OJS (previously determined) was subtracted from the spectrum of Subject 5, whereupon the curve labeled " K^{40} subtracted" in Fig. 21 was obtained representing the Zn^{65} and "excess" Cs^{137} activity. A Zn^{65} phantom spectrum and a Cs^{137} phantom spectrum were obtained using presdwood phantoms of appropriate thickness. The Zn^{65} phantom spectrum was normalized so that the area under the Zn^{65} photopeak matched the area under the subject's Zn^{65} photopeak and the normalized spectrum subtracted. This yielded the curve labeled " $0.48 \mu c Zn^{65}$ subtracted" in Fig. 21. The Cs^{137} phantom spectrum was then normalized to match the area under the subject's remaining Cs photopeak. This normalized Cs^{137} spectrum was subtracted from the above resultant spectrum and the residual curve labeled

Fig. 20. Net in vivo gamma-ray spectra of Marshallese

Fig. 21. Analysis of gamma-ray spectrum of Marshallese Subject No. 5

Fig. 22. Net in vivo gamma-ray spectra of Marshallese Subject No. 5 (K^{40} subtracted)

Fig. 23. Net gamma-ray spectra of Marshallese Subject No. 5

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 "0.21 μc Cs^{137} subtracted" was obtained. This residual spectrum below .4 Mev is undoubtedly due partly to the fact that the Cs and Zn phantom spectra are not exactly identical to the in vivo Cs and Zn spectra. It is felt, however, that the burdens of two subjects calculated in this fashion are correct to at least 5 per cent.

Calculating the body burdens of the five subjects who had been measured only in the standard chair presented a problem. The standard chair position cannot be calibrated with a source and a presdwood phantom since all volumes of the body are not at the same distance from the crystal, a condition which is assumed when a person is placed on the one-meter arc.

Since administration of the pertinent isotopes to the subject was deemed unadvisable, the standard chair position was cross calibrated to the one-meter arc position using the gamma-ray spectra of Subjects 5 and 8 in the following manner. The appropriate K gamma spectrum of a control subject of the same height and weight was subtracted from the gamma-ray spectrum obtained with Subject 5 in the chair and on the arc. The resulting net in vivo Zn^{65} and Cs^{137} spectrum, from 600 kev to 1.3 mev, obtained with the chair position was 4.1 times the net in vivo Zn^{65} and Cs^{137} spectrum obtained on the arc. (See Fig. 22) The same ratio of 4.1 was found for the spectra of Subject 8.

The standard chair gamma-ray spectra of the five subjects were analyzed using the identical peel-off procedure discussed above for the one-meter arc technique above and demonstrated with Fig. 21 except that the part of the spectra falling below 600 kev was neglected and the chair to one-meter arc ratio was taken into account.

The values calculated for each subject were given by Table 9 above.

To investigate the gross distribution of Zn^{65} in the body, gamma-ray spectra were obtained of Subject 5's feet, abdomen, and head by merely placing the crystal near the sites in question. The results, shown in Fig. 23 demonstrate that $\text{Zn}^{65}/\text{Cs}^{137}/\text{K}^{40}$ ratios do not vary widely.

In order to determine the effective biological half-life of the isotopes involved, the Marshallese were supplied with suitable containers and requested to submit their excreta. However, due to sight seeing trips, reporters, newsreel people and other extraneous activities, some samples were lost, and no samples could be regarded as representative of excretion rate over a definite period of time.

In order to gather at least some approximate information the samples were treated as follows: the K, Cs^{137} , and Zn^{65} content of each specimen was measured and the Cs^{137} and Zn^{65} values were expressed in terms of $\mu\mu\text{c}$ per gram of K. Since the Marshallese were on a standard American diet for

about 10 days, they were assumed to be in equilibrium with our diet and to eliminate (as the average of our control group) 2 grams of K per day in the urine and 0.5 gram per day in the feces. Knowing the K contents of the samples collected, this assumption permits a not-too-far-fetched extrapolation to the daily excretion rate.

The Zn^{65} data are given in Table 10. An average of $6.7 \mu\mu\text{c}$ of Zn^{65} per $\text{m}\mu\text{c}$ of Zn^{65} present in the total body is eliminated with each gram of K in the feces. The Zn^{65} amount present in the urine is negligible. On the basis of 0.5 gram of K eliminated per day by the fecal route, $3.4 \mu\mu\text{c}$ Zn^{65} per $\text{m}\mu\text{c}$ Zn^{65} in the body would be eliminated per day. The biological half-time for the element is thus 200 days. An effective biological half-time of 110 days results by adding the effect of the physical half-life of 245 days. This is considerably longer than the published values.

TABLE 10

Subject	Feces	Urine	Body burden	$\mu\mu\text{c}$ Zn^{65} per gram K in feces
	$\frac{\mu\mu\text{c} \text{Zn}^{65}}{\text{g K}}$	$\frac{\mu\mu\text{c} \text{Zn}^{65}}{\text{g K}}$	$\frac{\text{m}\mu\text{c} \text{Zn}^{65}}{\text{m}\mu\text{c} \text{Zn}^{65}}$	$\frac{\text{m}\mu\text{c} \text{Zn}^{65} \text{ in body}}{\text{m}\mu\text{c} \text{Zn}^{65}}$
4	no data	-	29.5	-
5	3340	82	482	7
6	536	-	73	7.4
7	236	-	62.1	3.8
8	1340	42	229	5.9
9	183	-	29.5	6.3
10	280	-	29.5	9.7
				average 6.7

The Cs^{137} data on two subjects are given in Table 11; they similarly are based on the American average of 2 grams of K eliminated in the urine per day and 0.5 gram of K in the feces per day. The effective biological half-time was 92.4 days for Subject 5 and 157 days for Subject 8. The average of these values is in essential agreement with values found under controlled conditions.

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TABLE 11

Subject	Feces $\frac{\mu\mu\text{c Cs}^{137}}{\text{g K}}$	Urine $\frac{\mu\mu\text{c Cs}^{137}}{\text{g K}}$	Body burden μc	Daily excretion $2 \times (\text{urine}) + 0.5 (\text{feces})$ $\mu\mu\text{c Cs}^{137}$
5	825	575	0.22	412 + 1,150 = 1,562
8	1040	638	0.41	520 + 1,276 = 1,796

IV. Exchangeable Potassium in Humans

C. E. Miller

The measurements of exchangeable potassium in humans are based on the following simplified model. The normal K present in humans is considered to be present in two fractions or pools. The atoms of K in one pool are bound within the cells and are not immediately replaced by other atoms of K from the blood stream. The atoms of K in the second pool are exchangeable and are constantly being replaced by new atoms of K. The K present in the urine has thus originated from the exchangeable K pool.

The exchangeable pool is measured in the following fashion. Each subject is given 5 μc of K^{42} at 9:00 A.M. All urine voided from 9:00 A.M. until 10:00 P.M. is collected in one vessel and the amount of K^{42} eliminated recorded. The first void the next morning is collected in a second container, and all urine voided thereafter until the subject is measured is collected in a third container. The amount of K^{42} present in the body at any time is thus known. All K^{42} has been mixed into the pool of exchangeable K by the definition of exchangeable K. The ratio of K^{42} to normal K present in the urine the following morning should equal the ratio of K^{42} to normal K present in this so-called exchangeable pool. The amount of K^{42} present in the urine determined by gamma-ray analysis, and the amount of normal K present is measured with a flame photometer. The amount of K present in the exchangeable pool is found by multiplying the K^{42} present in the body by the ratio:

$$\frac{\text{normal K in the urine}}{\text{K}^{42} \text{ in the urine}}$$

This value of K is then compared with the total body potassium as measured by the in vivo gamma-ray techniques.

On the basis of repeated tests performed on five subjects it can be said that the exchangeable potassium is of the order of 90 per cent or more of the total potassium. It seems certain, however, that because of either

instrumentation irreproducibility or more likely, unsuspected diuretic factors, the measured specific K^{42} activity in the urine is likely to vary considerably in the same individual. It is felt, therefore, that greater accuracy in the determination of the exchangeable fraction must await clarification of the apparent irreproducibility of the specific K^{42} activity of the urine.

V. Radium in Humans

C. E. Miller

There is seemingly a tendency to relate the radium burdens of former dial painters to the length of their employment in the radium dial industry. Information elicited during conversations with some of these former dial painters indicates that such a relationship does not exist.

One individual disclosed that some of the girls painted their fingernails, eyebrows, and even streaks in their hair with radium dial paint before going out on social dates. A second subject stated that the painters who taught new employees would eat a quantity of radium dial paint from a spatula to convince them that the material was harmless.

Considering such practices, it should not be considered unlikely to find an individual with a current burden of $2 \mu\text{c}$ of Ra^{226} as a consequence of an employment period of only three months (prior to 1930) in the radium dial painting industry. To our collection of cases containing Ra^{226} as a result of either employment or therapy two subjects have been added bearing 2.45 and $1.15 \mu\text{g}$ respectively.

VI. Radioactivity in the Liver of a Tridacna Clam

C. E. Miller

On June 30, 1956, Dr. Theodore R. Folsom of the Scripps Institute of Oceanography, La Jolla, California, sent two samples from the liver of a Tridacna (killer) clam to this Laboratory for gamma-ray analysis. This clam had been collected May 18, 1956, prior to the tests of that year in the Bikini Lagoon, 1000 feet northwest of the shore of Bikini Island at a depth of 10 feet. The weight of the whole clam meat was estimated at 1.8 to 2.0 kg wet weight. The weight of the part saved for assay (commonly called the liver) was 171 grams wet weight and 29 grams dry weight, of which two samples weighing 5 and 7 grams were used for measurement.

The gamma ray-scintillation spectra obtained from the 7-gram sample is illustrated in Fig. 24 wherein the top curve is the spectrum obtained from the gross sample. The low energy photopeak was identified as Co^{57} and

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the upper two peaks as partly, at least, Co^{60} since appropriate tests demonstrated that the 1.17 Mev and the 1.33 Mev peaks were in coincidence. The area under the 1.33 Mev peak represents 0.18 μc of Co^{60} as of May 6, 1957, indicating that the entire liver contained 1 μc of Co^{60} .

To check on the possible presence of Fe^{59} , Dr. Jacob Sedlet performed chemical tests separating the Fe into one fraction and Co into another. No activity appeared in the Fe fraction while the Co fraction yielded the spectrum shown by the middle curve of Fig. 24. The fact that the low energy gamma ray quantitatively follows the Co^{60} fraction demonstrates that the low energy gamma is in fact due to Co^{57} .

The Co supernatant was permitted to stand overnight yielding a second precipitant. The filtrate yielded the gamma-ray spectrum shown by the curve labeled "Zn fraction" in Fig. 24. A small amount of Co^{60} is present and about 0.1 μc of Zn^{65} as of July 5, 1956. The remaining supernatant yielded the gamma ray spectrum shown by Fig. 25. Trace amounts of Manganese-54, Zn^{65} , are evident and the photopeak at 510 kev is identified as Na^{22} .

VII. Background Studies

C. E. Miller and O. J. Steingraber

Reduction of Background of Iron Room

It was reported in the last semiannual report⁽¹⁾ that surrounding the crystal with either 1/16 or 1/8 in. of virgin lead reduced the gamma-ray background at the lower energies by approximately 250 c/m.

The floor of the room was covered with 1/8 in. of lead since previous experiments had indicated that the addition of a greater thickness did not further reduce the background. The gamma-ray background of the 8-in. by 4-in. NaI(Tl) crystal was thus reduced by 10 per cent within the energy band from 75 kev to 375 kev.

The addition of 1/8 in. of lead to the west wall further reduced the remaining gamma-ray background by another 10 per cent. The remaining walls will be covered in turn and subsequently reported.

Radioactivity in Welding Rods

All materials utilized in the building of a low level counting facility should be checked for activity, and among those counted for use in the construction of Steel Room II were a number of welding rods, which yielded very definite radium and possible potassium spectra. Further investigation indicated that the activity is in the flux and the amount present varies with the type of rod.

Fig. 24. Gamma-ray spectra of clam liver - Sample B

Fig. 25. Spectra of residue of clam liver

Of the three types measured, the approximate amounts of radium found are tabulated as follows:

<u>Type of rod</u>	<u>$\mu\mu\text{c Ra/rod}$</u>
6010	6
6012	60
6026	100

Type 6010 was considered acceptable. Competent welding will leave very little flux trapped in the bead and the welds will be primarily in the corners of the room, so comparatively little activity will be added to the room background.

Acknowledgement

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

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METEOROLOGY

I. Atmospheric Diffusion Studies

P. E. Wasko and Harry Moses

A. Introduction

Studies of smoke plumes reported in the literature have been based on either multiple exposure pictures, time exposure photographs or measurements of concentration averaged over a specified time period. Information obtained by investigations of this type is based on the combined effect of the diffusion of the smoke particles within the plume and at its boundaries and also the displacement of sections of the plume or its meander. Smoke diffusion measurements made by the Meteorology Group through the use of the meteorology stack, K-24 aerial cameras and the photogrammetric analyzer have allowed separation of the meander component from the diffusion about the plume itself. A resume of some of the analyses obtained with our equipment is presented in this report.

B. Procedure

The meteorology stack, the K-24 aerial cameras, and the photogrammetric analyzer have been discussed in previous Argonne reports.⁽¹⁻³⁾ However a brief description will be presented here in order to provide an adequate background for the reader. Two K-24 aerial cameras facing each other and tilted 30° with respect to the horizontal were positioned on a four-hundred-foot base line located eighty feet from the meteorology stack. The cameras photographed the plume from opposite sides. A third K-24 aerial camera was located equidistant from the other two at forty feet from the stack and was directed vertically upward so that the bottom of the plume was photographed. The data obtained from these measurements with the aid of the photogrammetric analyzer were placed on IBM cards and analyzed by means of the IBM 650 computer. Tabulations were obtained through the use of an IBM 407 tabulating machine.

A series of 81 photographs taken on October 5, 1956 forms the basis for these studies. In this series of photographs, the first 45 were taken at $3\frac{3}{4}$ -second intervals, the next 31 were taken at $7\frac{1}{2}$ -second intervals, and the remaining were taken at 15-second intervals. However there was a 55-second break after the 45th picture and a 75-second break after the 76th measurement. The summaries discussed in this report were based on pictures which were taken every 15 seconds, i.e., every fourth picture in the first group, every second picture in the second group, and consecutive pictures in the third group. A total of 33 photographs was used. The meteorological measurements and the experimental parameters prevailing during the time of the run are tabulated in Table 12.

TABLE 12

Meteorological and stack parameters during period of study

Date	October 5, 1956
Time	1500 - 1512 CST
Stack height	110 feet
Stack flow rate	2500 CFM
Effluent temperature at top of stack	115° F
Weather	Clear
Ground condition	Dry
Ambient air temperature at 110 feet	66° F
Wind speed at 110 feet	11 mph
Wind direction at 110 feet	100°
Stability of 5 5-110 feet	-2.2° C/100 meters (superadiabatic)
Solar radiation on horizontal surface	0.58 langley/min.

C. Types of Analyses

1. Time Series Representation

The 650 computer and the 407 tabulator were programmed to plot on IBM paper sheets the actual values of particular variables such as plume thickness as a function of distance from the stack along the plume's center line. In this presentation the abscissa represents time and the ordinate the distance from the stack. Numbers plotted in a vertical column thus represent the measurements of a single instantaneous plume. The position of the numbers starting at the bottom represents distance from the stack. The space between the column of figures represents time intervals between photographs. It thus becomes possible to draw isopleths of the variable plotted. Presenting the data in this fashion enables one to tell at a glance the similarity between the consecutive plumes which result from large scale eddies acting over periods of time longer than the time interval between pictures. However, very often photographs taken only a few seconds apart show plumes with markedly different geometric configurations. With this technique, errors in computation or data evaluation stand out quite noticeably when data from adjacent photographs are compared.

2. Plume Averages

To study the characteristics of the plume, averages were calculated of a number of pertinent variables for various distances from the stack along the plume's center line. The following plume characteristics were investigated:

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1) Horizontal width: This parameter denotes the horizontal dimension of the plume in the plane perpendicular to the plume's center line at the point of measurement.

2) Thickness: This parameter 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.

3) Cross-sectional area: This is the area formed by the intersection of the plume with the plane perpendicular to its center line. The area is assumed to be elliptical and is calculated from the expression:

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

4) Height of center line above stack.

5) Percentage rate of change of cross-sectional area with distance along the plume's center line

6) Percentage rate of change of horizontal width with distance along the plume's center line.

7) Percentage rate of change of thickness with distance along the plume's center line.

The percentage rate of change of the cross-sectional area represents the rate of entrainment of air into the plume and is a measure of the rate of eddy diffusion. This divergence of the area was calculated from the expression $\frac{1}{A} \frac{\Delta A}{\Delta S}$ where A denotes the cross-sectional area and S denotes the distance along the plume's center line.

The average horizontal width and thickness as a function of S are presented in Figs. 26 and 27. Following the rapid increase in about the first 20 feet, the thickness and horizontal width increase more or less uniformly with distance. The initial rapid increase in all probability is due to the stack-induced turbulence. Of interest are the bumps in the curve. These may be due to a variety of causes such as faulty operation of the photogrammetric analyzer, a few plumes with extremely large or small thickness and width values at the distances in question, or perhaps terrain effects. Also noteworthy is the manner in which the standard deviation increases. A calculation of coefficient of variation (100 x the ratio of the standard deviation to the mean) indicates that this statistical parameter increases with distance.

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Fig. 26. Plume's horizontal width as a function of distance along plume's center line.

Fig. 27. Plume's thickness as a function of distance along plume's center line.

The average ratios of the horizontal width to thickness were calculated and are plotted in Fig. 28. It appears that these ratios remain relatively constant throughout the region of measurement.

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Fig. 28

Ratio of plume's horizontal width over thickness as a function of distance along plume's center line.

The average height of the plume's center line as a function of distance from the stack is shown in Fig. 29. It is evident that most of the rise has been completed in 30 to 35 feet. The rapid rise in height beyond 150 feet is open to question. Attention is called to the nearly linear rate of increase of the standard deviation with distance along the plume.

Fig. 29. Plume's height above the top of the meteorology stack as a function of distance along plume's center line.

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Figure 30 shows the manner in which the cross-sectional area increases with distance from the stack. The exponential rate of increase is what one would expect from the appearance of the curves of thickness and horizontal width.

The effects of the increased zone of turbulence near the stack are depicted most strikingly by the curves of Fig. 31. In these curves the change with distance in the divergence of the area, the thickness, and the horizontal width are shown. It appears that this zone of rapid mixing ends at about 20 feet or slightly over 13 stack diameters from the stack. Further analyses of this type are necessary to determine whether the oscillations in the curves are meteorologically significant.

To present additional information on the behavior of the plumes studied, frequency polygons showing the distribution of the plume's cross-sectional areas at distances of 40 and 70 feet are shown in Fig. 32. Also the horizontal widths at distances of 10, 40, and 70 feet are to be found in Fig. 33. As seen from the curves of standard deviation versus distance, the dispersion increases with distance from the stack. It is to be noted that all of the distributions are skewed to the right.

Similar curves for the thickness and height of the plume are drawn in Figs. 34 and 35. These curves are for distances of 40, 70, and 130 feet from the stack. As is the case for the curves of horizontal width, the curves of thickness are skewed to the right. However, the frequency polygons of the plume's height appear to be more symmetrical.

The detailed analysis of this series of smoke plume photographs provides hitherto unavailable information on the characteristic of the smoke plume such as the entrainment rate, variation with distance of the horizontal width and thickness, and the diffusion in close proximity to the stack. Also, information of plume characteristics is derived with the effect of meander eliminated. In addition to the diffusion within the plume at its boundaries and the meander effect, a third factor to be considered is suggested by the skewness to the right of the frequency distributions of plume's thickness, width, and cross-sectional area. It is evident that a process operates sporadically to cause a marked increase in diffusion over a given section of the plume. This might be explained by the appearance of momentary gusts of scale length and intensity which are particularly effective. However, this bears further investigation.

Since the programming on the IBM 650 computer has been completed for this type of study, the analysis of the remaining smoke plume photographs taken under a variety of meteorological conditions should proceed more rapidly.

Fig. 30. Plume's cross sectional area as a function of distance along plume's center line.

Fig. 31. Divergence of the plume's horizontal width, thickness and cross sectional area as a function of distance along plume's center line.

Fig. 32. Frequency polygon of plume's cross sectional area at 40 feet and 70 feet along plume's center line.

Fig. 33. Frequency polygon of plume's horizontal width at 10 feet, 40 feet, and 70 feet along plume's center line.

Fig. 34. Frequency polygon of plume's thickness at 40 feet, 70 feet, and 130 feet along plume's center line.

Fig. 35. Frequency polygon of height of plume's center line above the top of meteorology stack at 40 feet, 70 feet, and 130 feet along plume's center line.

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II. Concentration of Atmospheric Radon

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

A. Introduction

The measurements of atmospheric radon under the direction of A. F. Stehney and H. F. Lucas are described in the previous semiannual report.⁽⁴⁾ These observations began in August 1956 and have been continued to the present time.

Radon samples are obtained through an intake duct located three feet above ground and one foot from the east wall of a one-story building, Building 203. Water and carbon dioxide of the sampling air are removed by drawing the air through Drierite and Ascarite. The radon is absorbed in a charcoal trap immersed in a dry ice bath. A zinc sulfide scintillation counting chamber previously described is used for measuring the amount of radon present in the sample. Samples are obtained daily during the work week, Monday through Friday, for the three 8-hour periods: 0045-0845, 0845-1645, 1645-0045.

The quantity of radon present in a given volume of air near the ground surface is a function of: (1) the amount released from the soil, (2) the rate at which the radon is diffused, and (3) the amount of radon which is advected by the wind from other locations. In this report only the first two factors will be considered. No data are available on the advection factor for the Laboratory site at the present time.

B. The Effect of Wet Ground

One of the factors which is important in determining the amount of radon released to the atmosphere is the quantity of water in the soil in either liquid or solid form. During periods of appreciable precipitation and in the time immediately following precipitation when the ground is quite wet, a marked reduction is observed in the amount of radon measured. To study this effect, the radon measurement data were grouped into two categories. In the first group were placed those measurements which were taken when there was no precipitation within the 8-hour sampling period or in any of the two preceding 8-hour periods.

In the second category were placed both the data taken in periods where 0.10 in. or more precipitation fell and the data for the two subsequent 8-hour periods. If precipitation occurred in amounts less than 0.10 in., the data for the 8-hour period in which it occurred and those of the two subsequent 8-hour periods were not considered.

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Relative frequency distributions of radon concentration for each of the three 8-hour sampling periods and for dry- and wet-ground conditions are presented in Figs. 36-38.

From these figures, it is apparent that the most pronounced effect of wet ground occurs during the night when the atmospheric diffusion rates are low. However, the means of the distributions are lower for each of the sampling periods for wet-ground conditions.

By utilizing the soil temperature measurements, the data were tabulated for periods when the temperature of the soil one centimeter below the surface was 32° F or lower than 32° F. There does not appear to be a marked difference in the distribution of the data under these conditions. However, this study sheds no light on the problem since the temperature of the ground at the Meteorology Laboratory may have been at or below freezing while the temperature of the ground near Building 203 was above freezing because of the presence of this heated building nearby.

C. Seasonal Effects

There appears to be a seasonal variation of atmospheric radon concentration. Larger values appear in the summer and fall months than in the late winter and early spring. The monthly average and monthly maximum and minimum values of radon concentration are presented in Figs. 39-41. In this analysis the effect of precipitation is not considered. Separate graphs are presented for each of the 8-hour measuring periods.

In the first 8-hour sampling period covering night and early morning conditions, both the mean values and maximum values are quite high in the late summer and fall and drop steadily through April. In May, however, the curves start back up again. During the late summer and fall, the strong nocturnal inversions which occur tend to reduce atmospheric diffusion, and the dry ground supports radon release. These factors may explain the high values during the night at this time. During late winter and spring the moist ground apparently inhibits the emanation from the soil thereby resulting in low radon concentration values.

In the second 8-hour measuring period covering the daylight hours the average radon concentrations are appreciably lower than during the preceding 8-hour period. This is probably due to the effect of increased diffusion under the usual daytime conditions of atmospheric instability. The monthly averages increase from September to December when a maximum is reached and then decrease. The averages for the third 8-hour period 1645-0045, the late afternoon and evening, are generally intermediate between those of the other two.

Fig 36 Relative frequency distribution of radon concentration Measuring period 0045-0845, August 1956-May 1957 The ground was considered dry when no precipitation occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods The ground was considered wet when precipitation of 10 inch or more occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods

Fig 37 Relative frequency distribution of radon concentration Measuring period 0845-1645, August 1956-May 1957 The ground was considered dry when no precipitation occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods The ground was considered wet when precipitation of 10 inch or more occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods

Fig 38 Relative frequency distribution of radon concentration Measuring period 1645-0045, August 1956-May 1957 The ground was considered dry when no precipitation occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods The ground was considered wet when precipitation of 10 inch or more occurred within the 8-hour measuring period or within either of the two preceding 8-hour measuring periods

Fig. 39. Monthly maximum, mean, and minimum values of radon concentration in $\mu\mu\text{c/liter}$ for the period September 1956-May 1957. Period of measurement: 0045-0845.

Fig. 40. Monthly maximum, mean, and minimum values of radon concentration in $\mu\mu\text{c/liter}$ for the period September 1956-May 1957. Period of measurement: 0845-1645.

Fig. 41. Monthly maximum, mean, and minimum values of radon concentration in $\mu\mu\text{c/liter}$ for the period September 1956-May 1957. Period of measurement: 1645-0045.

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D. Radon Concentration as a Function of Wind Speed and Stability

In Figs. 42-46 are presented the results of the analyses of the radon concentration as a function of atmospheric stability and wind speed. Atmospheric stability is determined by means of temperature difference measurements obtained by two thermocouples, one at 5.5 feet above the ground and one at 144 feet on the meteorology tower. The stability values represent the mean of eight readings averaged over two minutes taken approximately on the hour. The wind speed data are taken from the anemometer at the 19-foot level on the meteorology tower and represent the mean of eight 10-minute averages. The 10-minute wind-measuring period started at 5 minutes to the hour in each case. Wind speed class intervals at 5 miles per hour were used. Values of radon concentration were averaged for each of the stability (temperature difference) intervals.

It is apparent that when all of the data are plotted on the same chart (Fig. 42) there is considerable scatter. This scatter is noticeably reduced as seen in Figs 43 to 46 when the data are stratified according to wind speed. Figures 43 to 46 show that radon concentration is positively correlated with stability and is markedly affected by wind speed. This is another way of indicating that the radon concentration is related to the diffusive power of the atmosphere since the diffusion rates also depend upon atmospheric stability and wind speed.

When the wind speed is 10 miles per hour or greater, the mechanically induced turbulence tends to prevent the buildup of an inversion with the result that the range of stability values is quite limited. If vigorous mixing takes place in an air layer, thermodynamic considerations indicate that the vertical temperature gradient within this layer approaches the adiabatic temperature gradient. Thus, the onset of moderate winds would therefore tend to destroy an inversion that may have been present.

It would therefore be expected that under windy conditions the radon concentration values would be low owing to the increased diffusion rates effected by the reduction in atmospheric stability and the increased mechanical turbulence. Figures 45 and 46 bear out this reasoning.

An examination of the data both at Argonne and elsewhere leaves no doubt that meteorological variables affect both the rate of radon issued from the soil and the rate of diffusion once the radon enters the atmosphere. Some meteorological variables such as atmospheric stability or soil moisture produce a more marked effect than do others on measured values of radon concentration.

It is possible, by multiple regression techniques, to determine the effect of each pertinent meteorological variable on the concentration of atmospheric radon at a given height above ground. A multiple regression analysis would then yield a prediction equation from which one could

Fig 42 Relation between average radon concentration and atmospheric stability for various wind speed categories August 1956-May 1957

Fig 43 Relation between average radon concentration and atmospheric stability for wind speed category 0-4 miles per hour, August 1956-May 1957

Fig 44 Relation between average radon concentration and atmospheric stability for wind speed category 5-9 miles per hour, August 1956-May 1957

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Fig. 45. Relation between average radon concentration and atmospheric stability for wind speed category 10-14 miles per hour, August 1956-May 1957.

Fig. 46. Relation between average radon concentration and atmospheric stability for wind speed category 15 miles per hour or greater, August 1956-May 1957.

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determine the radon concentration with a fair degree of accuracy if information on atmospheric stability, wind speed, soil temperature, rainfall, pressure, etc were available. The facilities available at Argonne, the 650 IBM computer or the machines in the Applied Mathematics Division, can be utilized for carrying out an analysis of this type without an undue amount of effort.

III. The Argonne Meteorological Model Towing Tank

Paul Frenzen

Introduction

The Argonne meteorological model towing tank has been constructed as an alternative to a wind tunnel for model studies of turbulent diffusion. Planning and initial construction phases of the program have been summarized in an earlier publication in this series.⁽⁵⁾

During the period covered by this report, preliminary trials of the equipment have been carried out, structural modifications indicated by these trials have been made, and techniques of observation and analysis peculiar to the apparatus and the proposed experiments have been largely perfected.

Apparatus

Modifications made in the tank and its auxiliary equipment as a result of preliminary trials include the following: 1) installation of diagonal braces to reduce the spreading of the sidewalls when the tank is filled to operating depth (see Fig. 47); 2) installation of water line filters to remove sediment and rust; and 3) installation of a set of low-speed change gears between the hydraulic transmission and the cable-driving sheave to improve control of the carriage speed at low setting.

Preliminary runs made with a coarse, rectangular mesh grid (one-inch bars, three inches between centers) also revealed a greater tendency for the generation of long gravity waves than had been anticipated, resulting in a surging component lengthwise along the channel. Such waves are usually prevented in incompressible fluid experiments by restricting the free surface with a rigid lid; this would be complicated in this tank by the presence of struts which necessarily extend down into the fluid from the overhead carriage. As an experimental compromise, the arrangement shown in Fig. 47 was constructed. The carriage was extended to ten feet in length, and a plexiglass lid half an inch narrower than the tank was hung below its entire length. A sled-like bow placed just ahead of the grid reduces entrance effects as the forward motion of the carriage brings water through the turbulence-generating grid and into the test section under the rigid lid. This device, though introducing an unusual, moving upper boundary

Fig 47 Towing carriage with ten foot aluminum and plexiglass "sled" suspended below. Note bow and rectangular grid (3/8 inch bar, 2 inch mesh) mounted at front. Long unistrut member at upper rear carries adjustable cams which operate camera and timer. Also visible are the diagonal braces added to reduce spreading of sidewalls under load.

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condition, effectively damps the vertical amplitude of the waves in the test section proper and greatly reduces it in the free surface regions at either end. However, vertical motions still remaining in these last-named regions contribute, through continuity, to some degree of horizontal surging in the test section.

Reducing the drag of the grid by reducing its solidity ratio (area occupied by bars/total grid area) will decrease the surging by decreasing the amplitude of the waves generated. Accordingly, the grid shown in Fig. 47 has recently been installed. Its 3/8-inch bars and 2-inch mesh have a solidity ratio of 0.34 as compared with 0.56 for the earlier, coarse grid. Solidity can easily be further reduced by increasing the grid's adjustable mesh length, but the intensity of turbulence generated by the grid will also be reduced accordingly.

Quantitative evaluation of the additional velocity components due to the surging motion will be obtained by taking averages of large numbers of velocity measurements and retaining their signs. Since the direction of flow cannot be determined in simple streak photos, it is necessary to use a streak-flash or some equivalently coded exposure technique. A rotating shutter recently installed will supply these data (see below).

In the event that the velocity components along the length of the channel do not have an average value near zero, or alternatively, a nearly constant, non-zero value that can be subtracted from individual measurements as a mean flow, then it appears that it will be necessary to install a rigid lid along the entire length of the channel. Some consideration has been given to such an installation, and it appears to be entirely feasible.

Observations and Analysis

A successful combination of lighting and tracer has been found in a pair of 250 watt mercury arc lamps illuminating droplets of a solution of about one part nitrobenzene to two parts olive oil. The tracer is mixed for minimum sinking speed (order of one mm/sec or less) and is injected into the experimental area with a long, ten-mil hypodermic needle. The droplets deflect light through an angle between incident and emergent rays of approximately 87 degrees; so that with properly oriented lights and camera, they are seen as brilliant points of light, each an image of the source.

Essentially two dimensional photographs of an approximately six-inch square vertical section (x,z) of the tank are obtained by using a fifteen-inch lens and removing the camera to a position twelve feet from the tank. Parallax error due to particle motion across the tank (y direction) is reduced to a maximum of a few per cent, i.e., only at most a few per cent of any v component appears in u and/or w. Errors due to vibration at the end of this long optical lever were minimized by fixing the camera to a rigid framework (Unistrut) loaded with several lead bricks.

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Trial evaluation of the first streak photographs, carried out manually on a projector viewer supplying a ten-inch square image, pointed up the difficulties involved in extracting statistically significant amounts of data by such methods. Therefore, arrangements have been made to employ digitized, coordinate read-out equipment (Benson-Lehner Oscillograph Analyzer and Reader, OSCAR) at the Cloud Physics Laboratory, Department of Meteorology, University of Chicago. Since readings will be taken from prints, a counter has been installed where it can be seen in every photograph, thus preventing confusion among prints in the analysis.

In order to obtain successive, accurately timed, instantaneous positions of particles moving on long trajectories for the evaluation of Lagrangian correlation coefficients as described below, a rotating shutter synchronized with the Robot camera's automatic film traversing mechanism has been installed. It will also supply the velocity component data necessary for the evaluation of the surging component described above.

The shutter consists of a fourteen-inch disc arranged to rotate in front of the camera lens at one revolution per second. The disc has three windows at 120-degree intervals and a fourth window centered at an intermediate, 60-degree position. Hence the path of a particle moving at nearly uniform speed from left to right appears thus: - - - -. Similar points on the first, second, and fourth dashes fix particle locations at one-third second intervals, while the additional dash in the third position establishes the direction in which the particle is moving. A cam on the disc's driving shaft trips a microswitch which operates the camera through a double push-pull solenoid arrangement, moving the next frame of film into place during one of the longer, ($1/4$ sec) closed intervals of the rotating shutter.

Figure 48 is a rotating-shutter test photograph taken in the lee of the $3/8$ -inch bar, 2-inch mesh grid moving at 25.7 cm/sec. The longer streak groups represent particle velocities of the order of one or two cm/sec. The field shown is in the region of transition between wake-flow and homogeneous, decaying turbulence.

Figure 49 shows four-second trajectories traced from Fig. 48 and the three photos immediately following it. Note that the sampling of particle positions at one-third second intervals is continuous from frame to frame. Due to the width of the illuminating light beam, trajectories seen crossing during the same time interval in this two dimensional plot (x,z) may be separated by as much as several inches in the y direction.

Experimental Program

Continuous sampling of particle positions has been set up to determine Lagrangian correlation functions, the correlation of the velocity of a particle at an initial instant with its velocity at successively later instants. It is generally agreed that this is the fundamental statistical measurement

Fig. 48. Test photograph taken with the rotating shutter. Time interval between corresponding points of first and second streaks is $1/3$ second and between points on succeeding streaks, $1/6$ second.

Fig. 49

Four-second trajectories traced from Fig. 48 and the three succeeding photographs. Time intervals between positions are $1/3$ of a second, the numbers indicating elapsed time in whole seconds.

appropriate to the study of diffusion. Indeed, G. I. Taylor, in his classical study of "Diffusion by Continuous Movements,"⁽⁶⁾ developed the remarkable expression below, showing that the mean dispersion of particles ($\overline{x^2}$) is a function only of the intensity of turbulence ($\overline{u^2}$) and the Lagrangian correlation function ($R(t)$).

$$\overline{x^2} = 2 \overline{u^2} \int_0^T \int_0^t R(t) dT dt .$$

This equation has been the starting point of many subsequent treatments of diffusion, their individual contributions chiefly being ways of getting around the correlation function, since it presents evaluation difficulties in both the laboratory and in the atmosphere. Yet it remains the heart of the matter.

For example, Taylor points out two limiting solutions of the equation. The first, when T is so short that $R(t)$ does not vary significantly from unity (i.e., when the turbulent components essentially conserve themselves), readily leads to the result that the mean dispersion is directly proportional to the elapsed time, this implying that a cloud of particles emitted from a fixed point in a mean current will fill a cone. The second case, when T is so long that $R(t)$ falls to zero (i.e., when the final velocity of the particles has nothing at all to do with its initial velocity), gives the result that the dispersion is proportional to the square root of the time; here the cloud of particles in a mean current fill a paraboloid.

The program of experiments with the towing channel will investigate this transition from conic to paraboloidal dispersion as a function of the stability of the medium. First, direct measurements of particle trajectories will be used to evaluate the Lagrangian correlation functions in stable, neutral, and, if possible, unstable stratifications, comparing their rates of fall-off from unity in the several cases. The occurrence and significance of negative correlations after longer time intervals as reported in some atmospheric observations will be investigated. Then comparisons of these effects on model plumes will be studied, seeking models of the atmospheric stack effluent cases known as "coning" and "fanning." And finally, in this series of studies, a comparison of the model results will be made with the limited amount of data that have been obtained in the atmosphere in neutral balloon experiments.

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IV. Climatology

During this period the five-year climatological report ANL-5592 was published. This extensive report summarizes meteorological measurements made both at Argonne National Laboratory and the U. S. Weather Bureau station at the Chicago Midway Airport during the period July 1949 - June 1954.

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5. Marinelli, L. D. Estimates of Bone Pathology to be Expected from Sr^{90} . ANL-5716 (Preliminary draft, April 22, 1957).
6. Marinelli, L. D. The Use of Low Level Gamma Spectrometry in the Field of Nuclear Medicine. (International Symposium on Nuclear Medicine, Turin, Italy, June 5-7, 1957).
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8. Miller, C. E. and L. D. Marinelli. Cs^{137} Fallout. A Summary of Observations in Humans (Radiation Research). (Abstract)
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10. Rowland, R. E., J. Jowsey and J. H. Marshall. The Microradiographic Appearance of Radium Damaged Bone (Radiation Research) (Abstract)
11. Shonka, F. R. and J. E. Rose. Conducting Plastic, Tissue Equivalent to Neutrons and Photons of Widely Different Energies. (Radiation Research) (Abstract)

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STAFF ACTIVITIES

American Meteorological Society (Chanute Branch), Rantoul, Illinois, February 28.

"Challenges to the Meteorologist in the Atomic Age." H. Moses.

Radiation Research Society, Rochester, New York, May 13-15.

"The Use of NaI Scintillation Spectroscopy in the Determination of γ -Ray Emitters in Bulk Material at Natural Levels." P. F. Gustafson and L. D. Marinelli.

"The Microradiographic Appearance of Radium Damaged Bones." R. E. Rowland, J. Jowsey, and J. H. Marshall.

"Autoradiographic Analysis of Non-Uniform Concentrations of β Radioactivity in Bone." J. H. Marshall, R. E. Rowland, and J. Jowsey.

"Cs¹³⁷ Fallout: A Summary of Observations in Humans." C. E. Miller and L. D. Marinelli.

"Continuous Sampling of Air for Radon Content." H. F. Lucas, A. F. Stehney and F. H. Ilcewicz.

"Conducting Plastic, Tissue Equivalent to Neutron and Photons of Widely Different Energies." F. R. Shonka* and J. E. Rose.

Naval Research Training Seminar on "Life Sciences and Atomic Energy." Brookhaven National Laboratory, New York, June 3.

"The Detection of Gamma Ray Emitting Isotopes in the Human Body." C. E. Miller.

III International Meeting on Nuclear Medicine. Turin, Italy, June 5-7.

"The Use of Low Level Gamma-Ray Spectrometry in the Field of Nuclear Medicine." L. D. Marinelli.

Second Technical Meeting on Nuclear Energy, Rome, Italy, June 24-28.

"La Radioattivit Gamma Del Corpo Umano." L. D. Marinelli.

AAAS Gordon Research Conferences. Bones and Teeth. Meriden, New Hampshire. July 14-19.

"Quantitative Autoradiography of Ca⁴⁵ and Sr⁹⁰ in Cortical Bone." J. H. Marshall.

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