ANL-6104
Biology and Medicine
(TID-4500, 15th Ed.)
AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

RADIOLOGICAL PHYSICS DIVISION
SEMIANNUAL REPORT

July through December, 1959

J. E. Rose, Division Director
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Preceding Report
ANL-6049 - January through June, 1959

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

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AN EMISSION SPECTRUM FROM RADIATION-DAMAGED XYLENE

I. B. Berlman

Emission spectra from radiation-damaged organic solid scintillators have been published,⁽¹⁾ whereas, to the author's knowledge, no emission spectrum from a radiation-damaged organic solvent has as yet been published. A possible explanation for this omission is that whereas the fluorescent yield from an organic solid is large because of the negligible quenching of the fluorescent light by the radiation products immobilized in the medium, in the case of a radiation-damaged organic solvent, the fluorescent yield is likely to be low because of the enhanced quenching engendered by the diffusion of the radiation-produced quenching molecules.

To investigate the possibility of a measurable emission yield from the radiation products of radiation-damaged xylene, 10cc of doubly-distilled and nitrogenated p-xylene, in a stoppered vial, were exposed to approximately 1.6×10^7 r from a Co^{60} gamma-ray source. The irradiated xylene, placed in a 1-cm quartz cell with an 8-mm quartz spacer, was excited with 3130 Å light from a Bausch and Lomb Monochromator. The choice of this wavelength was based on previous observations of the appearance of new long-wave absorption bands in the spectrum of heavily irradiated xylene.⁽²⁾ The choice of a sample thickness of 2mm was predicated on the results of an independent absorption measurement of the irradiated xylene which disclosed that in such a path length most of the 3130 Å light was effectively absorbed without undue absorption of the emitted light.

The emitted light from the radiation-damaged xylene was recorded by means of a Beckman DK-2 Spectrophotometer containing a 1P28 photomultiplier tube calibrated by means of a tungsten-in-quartz lamp.* The emission curve shown in Fig. 1 has been corrected for the wavelength response of the recording instrument.

Since 3130 Å light is not absorbed by nonirradiated xylene, it is concluded that the emitted light must originate from molecules produced by radiation.

In order to obtain a rough estimate of the fluorescent quantum yield involved, a comparison has been made between the yield of diphenyloxazole (PPO) molecules (in proportion of 1 g/l of xylene) and

*This calibration was performed by R. W. Anderson, a summer employee, in 1959.

the yield of the heavily irradiated (1.6×10^7 r) xylene under identical conditions of 3130 Å illumination. From a direct comparison of the area under the two emission curves, a fluorescent quantum yield of approximately 4% relative to that of PPO was estimated for the radiation-produced molecules. It is believed that no gross corrections are involved.

Fig. 1

Light emitted by irradiated xylene.

Efforts are being made to identify the molecules which are emitting the light.

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NONRADIOACTIVE ENERGY-TRANSFER MEASUREMENTS IN AN ORGANIC SCINTILLATION SOLUTION*

I. B. Berlman

The efficiency of nonradioactive energy transfer of an organic scintillating solution as a function of the solute concentration has been determined and substantiated by three independent methods. These methods include a pulsed electron-beam time-decay method, and two different ultraviolet stimulated light-output experiments. The solution studied was a p-xylene and diphenyloxazole (PPO) system. In Table 1 are shown some of the representative values of energy-transfer efficiency, in per cent, as a function of the PPO concentration.

Table 1

Conc. PPO, g/l.	Method I U-V, %	Method II U-V, %	Method III τ , %
0.02	14 $\pm$ 2	12 $\pm$ 2	14 $\pm$ 3
0.04	23 $\pm$ 2	25 $\pm$ 2	
0.1	41 $\pm$ 2	39 $\pm$ 2	40 $\pm$ 3
0.2	56 $\pm$ 2	59 $\pm$ 2	55 $\pm$ 3
0.25	62 $\pm$ 2	62 $\pm$ 2	59 $\pm$ 3
0.5	77 $\pm$ 2	80 $\pm$ 2	75 $\pm$ 3
1.0	85 $\pm$ 2	88 $\pm$ 2	84 $\pm$ 3
2.0	92 $\pm$ 2		90 $\pm$ 3

*Abstract of a paper which is being submitted to the Journal of Chemical Physics.

MEASUREMENT OF THE AMOUNT OF LIGHT EMERGING FROM A RIGHT CYLINDER CELL CONTAINING A SCINTILLATING SOLUTION ON VARYING THE TYPE OF REFLECTING SURFACE

I. B. Berlman

Introduction

A quantitative analysis has been made in an earlier report of the portion of light emerging from a scintillating solution housed in a right cylindrical cell with totally absorbing walls.⁽¹⁾ This calculated value, is 0.145. It is obvious that the fraction of light emanating from the window of a geometrically identical cell but having various reflecting surfaces can be calculated simply by a comparison of the emergent light.

Measurements have been made on cells having the following types of reflecting surfaces: 1) polished aluminum, 2) aluminum evaporated on aluminum, 3) aluminum evaporated on glass, 4) aluminum oxide, and 5) Teflon. The choice of these types of reflecting surfaces was dictated mainly by their availability.

Equipment

The description of the cell which is used for containing the scintillating solution is described in Reference 2. A cylindrical insert possessing one of the several types of reflecting surfaces under investigation is placed in this cell. After the solution is poured into the insert, a quartz window is placed over the opening of the cell and is kept in place by a retaining ring.

A 1-mc Co^{60} source, positioned at 20 cm from the center of the cell was used to excite the fluorescence of the solution. The emergent light was measured by means of a photomultiplier-electrometer dc light-output tester designed by R. K. Swank.*

The inserts containing the various types of reflecting surfaces were made as follows: Insert 1 with totally absorbing surfaces was made of brass which was cleaned, activated, and blackened in Ebonol "C" Special, according to the instructions from the Enthone Corporation.** Insert 2 was machined from solid aluminum stock with a highly reflective surface made by taking a fine cut on a lathe and polishing with Putz Pomade.†

*Physics Division

**Enthone Corp., Inc. New Haven, Conn.

† Manufactured by J. C. Paul and Company, Chicago, Illinois

Insert 3 was another polished aluminum insert on which aluminum was vacuum evaporated. The purpose of this insert was to detect any improvement provided by a freshly deposited aluminum surface. Insert 4 was made of glass, the inner surface of which was coated with a layer of evaporated aluminum. Insert 5 was brass with the inside surface sprayed with a solution of sodium silicate and aluminum oxide. Insert 6 was machined from a solid rod of Teflon. Since Teflon is somewhat transparent, greater reflectivity was expected on eliminating the Teflon insert and machining an entire cell from a solid rod of Teflon, with the proviso, that the internal diameter of the unitized cell be of the same dimensions as the internal diameter of the insert. This was made and tested.

Two scintillating solutions were used in making the light output tests: Solution I was xylene and 1 g/l. of diphenyloxazole (PPO), and solution II was xylene plus 2 g/l. of PPO and $\frac{1}{4}$ g/l. of p-bis [2-(5-phenyloxazolyl)]-benzene (POPOP). The mean wavelength of the emission spectrum from solution I was approximately 3800 Å; from solution II, 4200 Å.

Before the addition of the solute, p-xylene was twice distilled and nitrogenated. Each time the cell was filled, the operation was performed in an enclosed glove box with nitrogen flowing.

Discussion of the Results

The results obtained, which are tabulated in Table 2, demonstrate that Teflon is much better than any of the other surfaces. However, since Teflon stock material is not always uniformly white and is not always of the same degree of whiteness, the results will necessarily be dependent on the quality of the stock.

Table 2
Light output as a function reflecting surface

Surface	Solution I, %	Solution II, %
Black	14.5	14.5
Aluminum	25 ± 1	27 ± 1
Aluminum on aluminum	42 ± 1	42 ± 1
Aluminum on glass	47 ± 1	52 ± 1
Aluminum oxide	58 ± 1	60 ± 1
Teflon 1*	70 ± 1	70 ± 1
Teflon 2**	75 ± 1	75 ± 1

*Solid cell

**Insert

The amount of light emerging from the solid Teflon cell (Teflon 2) as shown in Table 2 is slightly greater than that from a Teflon insert (Teflon 1). This is due to the greater reflectivity resulting from the use of a thicker reflecting wall and possibly due to the greater reflectivity resulting from the use of whiter Teflon in the cell construction.

Aluminum surfaces are definitely inferior to Teflon, with the added disadvantage that aluminum oxidizes very readily in air.

No corrections have been made in the computations of the emissivity of the standard absorbing cell to take into account the processes of absorption and re-emission, but as a precaution the concentrations of the solutes in the solutions have been so chosen so as to minimize these effects. The measurements were made with two solutions with different self-absorption properties and different emission spectra. The relatively close agreement between the results in Column I and Column II indicates that there were no inherent gross errors. In Column II some of the values for aluminum are slightly larger than the corresponding values in Column I. These are explained by the greater reflectivity of aluminum at longer wavelengths.

The author wishes to thank Richard F. Selman for the vacuum evaporations, Edwin J. Fudala for machining the various cells and inserts, and Richard K. Langs for building the electrometer circuit.

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ENERGY RESOLUTION OF THE "TWIN" SCINTILLATION FAST NEUTRON SPECTROMETER

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

The "twin" scintillation fast neutron spectrometer,⁽¹⁾ which consists of a gamma-radiation-matched pair of liquid scintillating fluids with widely differing proton concentrations, is being used by the present authors to measure the cosmic-ray-produced neutron background as well as the output of various neutron sources. As has been described in a previous report,⁽²⁾ this spectrometer is currently in the process of being calibrated in terms of neutron energy by exposing it to monoenergetic neutrons of various energies produced with the 250-keV Cockcroft-Walton linear accelerator. A preliminary calibration curve has been plotted⁽²⁾ which shows the variation with incident neutron energy of the pulse height at the point of inflection in the forward edge of the proton recoil spectrum obtained from the spectrometer. With this preliminary calibration curve, the various proton recoil pulse height spectra obtained in the energy calibration runs were converted into neutron flux spectra by means of the approximate relationship

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

where $n(E)$ is the integrated neutron flux per unit energy interval of neutron energy E falling on the counter, $N(E)$ is the number of proton recoils detected per unit energy interval at proton energy E , $\sigma(E)$ is the neutron-proton scattering cross section at E , and k is a constant depending upon the properties of the spectrometer. From each of these converted spectra one can deduce the correction to the energy calibration curve which is necessary at the given incident neutron energy as a consequence of the fact that the peak of the neutron spectrum does not correspond exactly to the point of inflection in the resulting proton recoil spectrum (see Equation (1)).

The results of converting one of the proton recoil spectra to a neutron spectrum are shown in Fig. 2, which represents the data from a run in which a thick deuterium-in-zirconium target was bombarded with 225-keV deuterons. The scintillation cells of the spectrometer, which are of the order of 3.5 cm in diameter by 3 cm long, were placed at a distance of 79 cm from the 0.8 cm diameter neutron-producing target for this run. Hence, the data were taken under "good geometry" conditions. The error bars attached to the points in the Fig. 2 spectrum represent the resultants of the uncertainties introduced by the various processes used in converting from a proton recoil pulse height spectrum to a neutron flux spectrum. The greatest source of uncertainty, and the one which largely determines the size of the error bars, is the amount by which the preliminary energy calibration curve is uncertain. It is interesting to note that Fig. 2 indicates that the

Fig. 2. Spectrum of thick-target D-D neutrons obtained with the "Twin" Scintillation Spectrometer

preliminary energy calibration curve needs little correction at the neutron energy corresponding to the peak in this run since computation yields 2.18 ± 0.03 Mev for the conditions of the run.

The full width at one-half maximum amplitude of the neutron peak of Fig. 2 is 0.51 Mev, which represents an energy resolution of 23 per cent at 2.19 Mev. The energy resolution of the "twin" scintillation spectrometer at 2.19 kev must be at least a little better than this, however, because the neutron flux itself must have had an energy distribution whose width was appreciable in comparison with 0.51 Mev owing to the fact that the bombarding deuteron energy was 0.225 Mev and the deuterium target was thick. In conclusion, it might be noted for comparison purposes that the neutron energy resolution of the liquid nitrogen-cooled $\text{Li}^6\text{I}(\text{Eu})$ crystal spectrometer system reported by R. B. Murray is 18 per cent for 5.3 Mev neutrons.⁽³⁾

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DRIFT VELOCITY OF ELECTRONS IN NITROGEN, HELIUM,
NEON, ARGON, KRYPTON, AND XENON*

J. C. Bowe

Accurate values of electron drift velocity in purified noble gases and nitrogen have been obtained by measuring the transit time of photoelectrons across the gap of a parallel-plate condenser. Drift velocities in pure krypton and xenon have not been measured before. The values obtained for the other gases are invariably lower than those reported by Nielsen, the discrepancy for neon being large. The sharp maximum found by English and Hanna for argon at $E/p_0 < 0.1$ and tentatively ascribed by them to the Ramsauer effect was not observed in the purified gas.

*Abstract of paper accepted for publication in Physical Review.

TRANSPORT COLLISION CROSS SECTIONS FROM ELECTRON DRIFT-VELOCITY DATA*

J. C. Bowe

Information concerning the transport collision cross section $\sigma_t(\epsilon)$ of low-energy electrons in the noble gases was obtained from drift-velocity data. The results (with ϵ in ev) are He: $\sigma_t(\epsilon) = (28 \pm 1) \text{ cm}^{-1}$ for $0.13 \leq \epsilon \leq 4$; Ne: $\sigma_t(\epsilon) = (6.05) \epsilon^{0.157} \text{ cm}^{-1}$ for $0.38 \leq \epsilon \leq 8$; Ar: $\sigma_t(\epsilon) = (6.3 \pm 0.6) \epsilon \text{ cm}^{-1}$ for $1.6 \leq \epsilon \leq 11$; Kr: $\sigma_t(\epsilon) = 14 \epsilon \text{ cm}^{-1}$ for $1.6 \leq \epsilon \leq 3$; Xe: $\sigma_t(\epsilon) = 26 \epsilon \text{ cm}^{-1}$ for $1 \leq \epsilon \leq 2.4$. All σ_t values are for 0°C and 1 mm Hg pressure. These results are compared with other experimental and theoretical data. For nitrogen, the drift-velocity measurements indicate an average fractional energy loss per collision of 20 (2m/M)** in the energy range $\epsilon = 0.07$ to 0.15 ev.

*Abstract of paper accepted for publication in Physical Review.

**Electron mass/mass of gas atom or molecule

POSITIVE ION EMISSION: HYDROGEN EFFECTS

H. A. Schultz

Introduction

Any technique of chemical analysis must avoid interference between possible components and maintain a stable calibration. Water vapor is one example of a component of the atmosphere which interferes with the determination of traces of Freon-12 by positive ion emission techniques. In field tests that were run, this particular interference was eliminated satisfactorily by the use of a cold trap to freeze out the water vapor.⁽¹⁾ In these same tests, the calibration of the device was stabilized by the addition of hydrogen to the sample. Hydrogen accelerated the decrease in ion current following reductions in Freon concentration. A similar effect has been obtained with the present laboratory apparatus.

Hydrogen in Argon

Recent experiments, which will be discussed below, indicate that hydrogen as an additive to argon affects the ion current through two independent mechanisms. The desirable speed of response to halogen reduction is an effect associated with the boat (source of ionizable material), while the variations observed in background ion current are primarily a filament effect. In order to take advantage of the halogen effect, the background effect must be controlled. "Background" here means that the filament and boat are both heated, but the gas does not contain chlorine. In argon plus 0.33% hydrogen, the background ion currents are time dependent.⁽²⁾ The background decreases during the day, provided the gaseous mixture is changed periodically (about once an hour) but increases overnight, when the gas is not changed. The background ion current may be depressed much further, to as low as 1 or 2% of the morning value, by flashing the filament at 1530°K in a mixture of argon plus hydrogen (0.33%), dropping the filament temperature to normal (1300°K) and changing the gaseous mixture.

The high thermal conductivity of the hydrogen does not seem to be the property of prime importance. Starting with the high morning background, helium was substituted for hydrogen in a succession of refills. The helium increased the thermal conductivity of the gas, as hydrogen would have, but it had practically no effect upon the ion current, in contrast to the marked decrease that hydrogen would have produced.

The following experiments indicate that the effect of hydrogen upon the background is associated with the operation of the filament. The flashing process is effective in decreasing the background current whether the

boat is operated during the flashing or not. Also, in a series of tests, the low level background was obtained by flashing; then the hydrogen was omitted in the last refill of the day. The ion current increased overnight. Ion current vs time patterns were obtained under three different conditions. First, in the control tests, both the filament and the boat were operated continually. In the second case, the filament was operated all the time but the boat was operated only one-fourth of the time ($\frac{1}{2}$ hr on, $1\frac{1}{2}$ hrs off; or 1 hr on, 3 hrs off). This interruption of the boat did not affect the ion current vs time pattern. In the third case, both the filament and the boat were operated only one-fourth the time. The ion current vs "on" time was quite similar to the first two patterns, while ion current vs total elapsed time was very much different.

The heating of the filament is accompanied by a variety of secondary effects, any of which may conceivably contribute to the observed effect of hydrogen upon the background ion current. Since the container and the contents are all heated, desorption, leakage, or diffusion effects might be at work. Previous tests indicated that the introduction of a small amount of oxygen produces a rise of background similar to the rise which is observed when a filament is prepared by flashing in argon plus hydrogen and then operated overnight in pure argon. To check on the possibility of leakage or diffusion of air through the seals of the bell, a series of tests was run with these seals surrounded by argon at atmospheric pressure, and another series was run using an atmosphere of oxygen. No significant differences were observed. In a number of other tests, the glass bell was heated externally to desorb gases from its walls. The results were negative but were not considered to be conclusive since the temperature and the extent of the heated area were not well controlled.

The problem of possible change in the composition of the gaseous mixture was then attacked more directly. Provisions were made to withdraw samples of the gas for mass spectrometer analysis in the Chemistry Division. Samples have been taken from the argon tank, from the end of the processing train, and from the bell at various steps in ionization runs. No satisfactory correlations have been found between the analyses and the ionization results.

The impurities found have been: hydrogen (0.07 to 0.17%, when none was added); nitrogen (0.04 to 0.40%); oxygen (0.01 to 0.19%) and, in one sample, water vapor (0.01%). Methane, carbon monoxide and carbon dioxide have been sought and not found ($<0.01\%$). The samples were 99.5 to 99.9% argon, when no additives were used. One striking correlation has been noted: the nitrogen concentration of the samples is very close to 0.02% plus 2 times the oxygen concentration in all cases. However, samples taken under supposedly identical conditions show O_2 and N_2 variations of nearly 10 to 1. It is suspected that there is an error in the sampling and handling technique. It does not seem to be due to leakage of air, as air is $4(N_2):1(O_2)$, not 2:1.

The evidence that the quick termination of halogen response is a boat effect is as follows: when argon is used alone or with added oxygen, the enhancement of the ion current due to chlorine persists even after the gaseous chlorine has been removed from the bell. However, the ion currents obtained with the boat off are not affected, provided adequate collecting potential has been maintained continually. This indicates that the residual chlorine effect is carried over by the boat. With hydrogen added, there is much less carry-over.

Nitrogen

Nitrogen has also been used as principal gas, replacing argon, and hydrogen and oxygen have been used as additives to it. The nitrogen was shunted around the furnace containing hot magnesium since magnesium and nitrogen combine at high temperatures.

The background ion currents obtained in nitrogen depended upon filament temperature, hydrogen and oxygen concentrations, and time in a pattern which was qualitatively similar to that obtained in argon. However, there were differences in detail. For example, filament flashing in hydrogen gave a temporary low-level background ion current in nitrogen, as in argon, but more hydrogen was required in nitrogen to obtain the same effect. Continuation of nitrogen studies has been delayed while further efforts have been made to find explanations for the results of the argon studies.

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LATE OBSERVATIONS OF THE DISTRIBUTION OF RADIUM IN THE HUMAN SKELETON*

R. E. Rowland

The deposition of the alkaline earth elements in bone is a subject which has changed from one of purely academic interest to one of wide-spread concern today. This concern is the result of the presence of one particular radioisotope in the biosphere, strontium-90. What effects will the radiations from this isotope have upon us after it is deposited within our bones?

In order to understand clearly the over-all problem of strontium-90 toxicity, there are many areas that need investigation. For example, we may start with the metabolism of the alkaline earth elements in bone: how well do they substitute, at trace concentrations, for calcium in the mineral of bone? Once a radioactive isotope is deposited within bone, it continually irradiates the surrounding tissues. What dose rates actually exist within bone, and what tissues actually receive this irradiation? What is the effect of this dose on these tissues? After an isotope is built into bone, how long does it remain? What natural processes work to remove it and at what rates? If these questions can be answered, do they lead us to a method of counteracting the deleterious effects of irradiation, or to a method of removing the radioactive isotope?

In an attempt to answer some of these questions, we have turned our attention to those human beings who have carried another radioactive alkaline earth, radium, in their bones for many years. Here, we feel, is information of extreme value in the assessment of the hazards of Sr^{90} , for although radium and strontium are not chemically identical, they do behave with remarkable similarity in bone. Furthermore, the information available describes the uptake and distribution of this element in human bone, and, after all, it is in the human being that our primary interest lies.

Most of us are familiar with the fact that many individuals acquired a burden of radium through their contact with the radium watch dial industry. However, in addition to these now famous dial painters, there were probably many times as many people who acquired a burden of this radioactive element through its use as a therapeutic agent in the 1920's. Through these two sources a very large number of people have been exposed to a bone-seeking radioisotope and have by this date carried it for 30 odd years. What has happened to them?

*Presented at Univ. Minn. Symposium on "Radioisotopes in the Biosphere," Oct. 19-23, 1959, and to be published in the Proceedings of the Symposium.

I have had the opportunity of examining bone samples from 19 individuals who have carried this element from a minimum of 10 years to a maximum of 40 years. Their body burdens ranged from a high of $20 \mu\text{c}$ down to about $1 \mu\text{c}$ of Ra^{226} . (For reference, the industrial maximum permissible burden is $0.1 \mu\text{c}$.) Some of these individuals received relatively pure Ra^{226} , others received both Ra^{226} and Ra^{228} . The latter, mesothorium 1, is thought to be considerably more toxic than Ra^{226} .

These individuals, for the most part, received their radium as adults, although several of the women who painted watch dials started at age 15. The typical exposure period ranged from a few months to a few years; the routes of entry were both intravenous (for some medical cases), and oral (for dial painters and those who drank radium water).

When the bones of the dial painters were first examined by the autoradiographic technique, a spotty distribution of the activity was apparent. The term "hot spot" was coined to describe these radium concentrations. We now know that these concentrations exist in regions where new bone mineral was forming at the time the radium was acquired, and that in this mineral the original concentration of radium, expressed as the ratio of radium to calcium, was essentially the same as the radium to calcium ratio that existed in the blood plasma at the time the mineral was formed.⁽¹⁾

When we examine the bone sections in greater detail a second type of distribution of radium becomes evident, the so-called diffuse distribution. By exposing an autoradiographic plate to a bone section for a considerably longer period of time than is needed to show the hot spots, this rather uniform distribution is made visible (Figs. 3 and 4).

Figure 3 is an autoradiograph produced by exposing a plate to a piece of cortical bone from the femur of a woman who died with a body burden of $2.7 \mu\text{c}$ Ra^{226} . This activity was obtained at age 20 through exposure as a radium dial painter for about one year. The autoradiograph shows the distribution of radium 33 years later, at age 53. The number of hot spots is unusually large.

Figure 4 is an autoradiograph of a piece of cortical bone from the tibia of a woman who died with a burden of $3.8 \mu\text{c}$ Ra^{226} . In this case the activity is reported to have been acquired during a two-month period at age 15 when the subject painted watch dials. Here we can see the diffuse distribution quite clearly, with a few superimposed hot spots. This bone sample obtained at death 32 years after the activity was acquired.

We believe that this diffuse distribution is the visible result of an exchange process which continually transfers calcium atoms from blood to bone and back again, and which is characterized by an unusually long time constant. In this process the alkaline earth elements can substitute

Fig. 3. An autoradiograph of a segment of cortical bone from the femur of case RB, exposed for 2 months on an Eastman Kodak No Screen autoradiographic plate. There are an unusual number of hot spots in this segment of bone.

Fig. 4. An autoradiograph of a segment of cortical bone from the tibia of case 302, exposed one month on an Eastman Kodak No Screen⁽²⁾ autoradiographic plate. Several hot spots are seen superimposed on the diffuse distribution.

remarkably well for calcium, at least at very low concentrations. Thus, the diffuse distribution we see today is the result of this exchange which labeled the bone mineral existing at the time these individuals were exposed to radium. This activity is not to be considered as permanently fixed within the bone mineral, however, for this same exchange process is slowly removing it from the bone.

The hot spots are not nearly as large in area as one would be led to believe from their appearance in these illustrations. When we employ a medium of considerably higher resolution, a thin emulsion which stays in contact with the bone section during development and fixation, such as illustrated in Figs. 5 and 6, we see that the hot spots in cortical bone are usually no larger than a single haversian system.

Figure 5 contains a microradiograph of a 100-micron thick bone section, and a stripping film autoradiograph of the area outlined in the microradiograph. The entire large haversian system in the center of the figure contains radium, but in order to show clearly the alpha tracks, we have greatly enlarged the autoradiograph. Note the scarcity of tracks over the adjacent bone and over the canal itself as compared to the number over the haversian system.

Figure 6 contains a similar microradiograph and autoradiograph pair; again the autoradiograph is shown at considerably higher magnification to make visible the individual alpha tracks. Both Figs. 5 and 6 illustrate hot spots found in the femur of a woman who, at age 32, received 41 weekly injections of $10\ \mu\text{c}\ \text{Ra}^{226}$ each, and died 24 years later with a terminal burden of $3.6\ \mu\text{c}$. This type of repeated injection produced haversian systems completely labeled from outer cement line to the inner canal, for the blood contained this radioelement longer than the complete period (about three months) required for the formation of a human osteon.

An alpha-track autoradiograph, because of its property of resolving the short tracks of individual alpha particles, provides an excellent method, first suggested by Hoecker and Roofe,⁽²⁾ of measuring the radium concentrations which exist in microscopic volumes of bone. A count of the number of alpha tracks per unit area in a suitable autoradiograph plate, exposed for a known time, can be employed to determine the concentration of radium per unit volume of bone if the range of the alpha particles in bone and the fraction of the gaseous daughter radon retained in bone are known.

The nineteen cases which have been analyzed thus far by this technique are listed in Table 3, and include data from 12 cases reported in a previous study.⁽³⁾ They are arranged in three groups according to the length of time that the radium was carried before the bone sample was obtained.

Fig. 5. A microradiograph and a stripping film autoradiograph of a 100-micron thick bone section from the femur of a woman who died with $3.6 \mu\text{c Ra}^{226}$ 24 years after receiving this isotope medically at age 32, (case R). The autoradiograph covers the outlined area of the microradiograph, and illustrates the intensity of the hot spot as compared to the adjacent diffuse radium distribution.

Fig. 6. A microradiograph and a stripping film autoradiograph of a 100-micron thick bone section from the femur of the same case illustrated in Fig. 5. The autoradiograph covers the outlined area of the microradiograph.

Table 3

Radium patients

Patient	Terminal radium burden, μc	Sex	Age radium acquired	Age at death	Years radium carried	Method of acquisition	Refer- ence
EE	50.?	F	21-24	32	11	Dial painter	4
KS	19.?	F	15-20	31	16	Dial painter	4
IL	8.?	F	16-20	30	14	Dial painter	4
FR	7.	F	21-22	48	27	Dial painter	5
103	6.8	F	36	64	28	I. V. medically	6,7
R	3.6	F	32	56	24	I. V. medically	6,7
Q	1.2	M	33	59	26	I. V. medically	7
BM	1.2	M	21-23?	51	28-30?	I. V. medically	
112	1.2	F	25	48	23	Oral: medically	6,7
I	0.8	F	28	53	26	I. V. medically	7
118	10.5	F	48	84	36	Oral? medically	6,7
LS	8.	F	17	57	41	Dial painter	5
302	3.8	F	15	47	32	Dial painter	7
HG	3.	F	16-19	48	32	Dial painter	5
RB	2.7	F	20	53	33	Dial painter	
313	1.3	F	18-47	48	30	Dial painter	7
IJ	1.2	F	18-21		31	Dial painter	
03473	1.2	F	19-21		36	Dial painter	8
ML	~1.	F	16-17		39	Dial painter	

The three cases that carried radium between 10 and 20 years all acquired it from the painting of watch dials. Question marks are placed with the reported body burdens because these estimates were made many years ago (in the 1920's and 1930's). If these cases had contained Ra^{228} , as is highly probable, most of it would have decayed as a consequence of its 6.7-year half-life, leaving only the 1620-year Ra^{226} present today.

Of the second group, those surviving between 20 and 30 years, all but one happened to have acquired radium medically. Those acquiring this isotope medically generally received it at a more mature age than the dial painters, a good number of whom started painting in their late teens.

The third group contains those that carried radium 30 or more years; it consists almost entirely of former dial painters.

In Table 4 are listed the measured radium concentrations obtained by means of the alpha-track counts. For each case we have attempted to find the most intense hot spot and a representative average value of the over-all diffuse distribution. These values are expressed in terms of $\mu\mu\text{c Ra}^{226}$ per mg of bone. For comparative purposes there is included a calculated "uniform label," the body burden of radium divided by the bone

Table 4

Measured hot spot and diffuse concentrations

Patient	Terminal radium burden, μc	Maximum hot spot, $\mu\mu\text{c, mg}$	Average diffuse, $\mu\mu\text{c, mg}$	Calculated uniform label, $\mu\mu\text{c, mg}$	Ratio: hotspot to diffuse	Ratio: diffuse to uniform label
EE	50.7	19.	1.2		16	
KS	19.1		1.4			
IL	8.1	9.0	0.28		32	
FR	7.	30.	0.32	1.0	94	0.32
103	6.8	38.	0.30	1.2	127	0.25
R	3.6	16.	0.15	0.66	107	0.23
Q	1.2	12.	0.080	0.17	150	0.47
BM	1.2	6.0	0.043	0.17	140	0.25
112	1.2	1.4	0.075	0.20	19	0.38
I	0.8	13.	0.060	0.15	218	0.40
118	10.5	41.	0.50	2.0	82	0.25
LS	8.	16.	0.54	1.3	30	0.42
302	3.8	20.	0.54	0.67	37	0.81
HG	3.	9.3	0.39	0.50	24	0.78
RB	2.7	15.	0.38	0.49	40	0.78
313	1.3	2.8	0.15	0.30	19	0.50
IJ	1.2	4.7	0.046	0.20	102	0.23
03473	1.2	3.3	0.18	0.20	18	0.90
ML	~1.	6.0	0.10	0.17	60	0.59

mass. This represents the radium concentration per mass of bone that would exist if this radioelement were uniformly distributed throughout the skeleton. Bone mass was assumed to be 10% of body weight, while the body weight, if not known, was assumed to be 70 kg for males and 60 kg for females.

The data in this table can be employed in various ways to characterize the distribution of radium in human bone. The ratio of the specific activities of the hot spots to the adjacent diffuse distributions is a measure of the actual degree of nonuniformity existing in these cases. The average value of this ratio for 18 of these cases is 73. It will be noted that this ratio is usually higher in the bones of those who acquired their radium medically; this may reflect the fact that exposure to radium medically usually was for a shorter period than exposure via the dial industry.

When radium, or any alkaline earth radioisotope, is present in the blood for only a short time, the resulting hot spot to diffuse ratio is very high. With longer exposure to this radioisotope, the ratio will be lowered, for the diffuse deposition will continue to increase in magnitude, whereas the hot spots, formed at the specific activity of the blood, cannot increase in activity. This is illustrated by the Elgin State Hospital patients, R, O,

and I, (Table 4) who received equal weekly injections of radium; R and Q received 41 and 45 injections, respectively, and we found hot spot to diffuse ratios of 107 and 150, while in case I, who received only 18 injections, we found a ratio of 218.

When we turn our attention to the diffuse distribution, a rather intriguing result is evident. The ratio of the measured diffuse concentration to the hypothetical uniform label ranges from a low of 0.23 to a high of 0.90, with an average value of 0.47. That is, for these 16 cases for which we can calculate a meaningful value for the uniform label, the average ratio of the measured diffuse to the uniform label is about one-half. Thus it is evident that this diffuse distribution is not an inconsequential fraction of the total body burden, but rather, a significant fraction of it.

The uniformity of this distribution is also quite remarkable. To be sure, variations do occur and are observed, and the value reported is the average value. However, the range of an average alpha particle in bone is 32 microns, so that, when we count the tracks over 1 mm^2 (the typical area surveyed for a single diffuse analysis), we are actually determining the bone radium content in a volume of $3 \times 10^{-5} \text{ cm}^3$, containing $60 \mu\text{g}$ of bone. Yet, from this small volume we obtain values of the diffuse specific activity that vary by less than a factor of two from region to region.

Thus, it is convenient to think of the distribution of radium in the skeleton as occurring in two components. First of all, about half of the activity may be considered to be uniformly distributed throughout the skeleton. This, of course, is just the type of distribution of an isotope we would expect if it were continually present as a contaminant in our food throughout our lifetimes, such as may be the case with strontium-90. Our radium cases then have, superimposed on this uniform distribution, the intense hot spots. It would be most illuminating if we could determine which of these two types of distribution was the most likely to produce visible evidence of radiation-induced damage.

The data compiled in Table 4 can be employed in many ways. For example, if I may speculate for a moment, let us use these data to see if we can determine a bone turnover rate. Let us restrict the meaning of bone turnover to new bone formation, such as the replacement of a resorption cavity within compact bone with a new osteon, or to the deposit of a layer of trabecular bone balanced by the removal of an equal volume elsewhere. That is, in the adult, let us consider a bone turnover occurring which does not increase the total volume of bone present.

Next, let us design a typical case in which the hot spot to diffuse ratio is 100, and the diffuse to uniform label is one half; further, let us specify that this is the situation 30 years after the exposure to radium, and that the exposure lasted for one year. Can we determine from these data the bone turnover rate?

From these figures it is apparent that, terminally, the hot spots must occupy 1% of the total bone volume (and contain 50% of the total body radium), whereas the diffuse label occupies the other 99% of the bone. If we make two assumptions, first, that bone resorption is not inhibited by radiation dose, and second, that bone turnover does not change with age, we can calculate a bone turnover rate. The calculations are shown in an appendix to this paper; the result shows that any turnover rate between 2 and 5 per cent would be expected to leave 1% of the bone labeled with hot spots after 30 years.* Note that a 5% rate would lead to a labeling of 5% of the skeleton after 1 year of exposure, while a 2% rate would yield a 2% label; however, after 30 years, only 22% of the original bone would be left if bone turnover were 5% per year, while at the 2% rate 55% of the original bone would still be left. These considerations tend to make the lower rate look more reasonable.** When we realize that radiation dose often does inhibit resorption, and that it seems reasonable to expect bone turnover to decrease with age in the normal human, then a yearly rate of 1% or 2% bone turnover might seem quite reasonable on the basis of these data.

We can go one step further in our analysis of the radium distribution data available from these human cases; we can compute the terminal dose rates that existed within the bone at the time of biopsy. For the calculation from bone radium concentrations to dose we follow the method of Spiers.⁽⁹⁾ We have chosen to express the dose to a cell within bone, rather than to bone itself. For this cell we consider an osteocyte within a 10-micron spherical hole in bone, the lacuna. If, instead of this cell, we choose to consider the dose received by a 10-micron thick layer of endosteum, periosteum, or lining of the haversian canal, the value will be about one-half that listed for this lacuna in Table 5.

In this table we have listed the terminal dose rate to a lacuna existing in a hot spot and to one receiving radiation from the average diffuse radium distribution. The unit employed is rad per day, so that these data express the rate at which energy is delivered to the cell by the alpha particles. It can be seen from the table that rather sizable dose rates occur within the hot spots in some cases, and that even the least intense spots delivered over a rad per day to the osteocytes.

* We have been unable to report a meaningful value of the fraction of the bone volume occupied by hot spots because of the small fraction of the total skeleton that is available for study. Local variations in bone turnover may result in one sample having several times as many hot spots as an adjacent sample of bone.

** With a large volume of bone turned over, the diffuse distribution would be expected to appear quite spotty on an autoradiograph. The newly-formed mineral, while still containing radium, would contain the same radium to calcium ratio as existed in the blood plasma at the time of its formation. The blood plasma contains a lower radium to calcium ratio than the diffusely labeled bone a few months after the last exposure to radium, and this ratio continues to fall with time. Thus, the uniformity of the diffuse distribution seems to imply that the total bone turnover is not great.

Table 5

Terminal dose rates

Patient	Terminal body burden, μc	Terminal dose rate to a 10- μ lacuna		Minimum lifetime dose to a 10- μ lacuna	
		Hot spot, rads/day	Diffuse, rads/day	Hot spot, rads	Diffuse, rads
EE	50. ?	15.	0.95	60,000	3,800
KS	19. ?		1.1		6,400
IL	8. ?	7.2	0.22	37,000	1,100
FR	7.	24.	0.25	240,000	2,500
103	6.8	30.	0.24	310,000	2,500
R	3.6	13.	0.12	110,000	1,100
Q	1.2	9.5	0.064	90,000	610
BM	1.2	4.8	0.034	49,000	350
112	1.2	1.1	0.060	9,200	500
I	0.8	10.	0.048	95,000	460
118	10.5	33.	0.40	430,000	5,200
LS	8.	13.	0.43	190,000	6,400
302	3.8	16.	0.43	190,000	5,000
HG	3.	7.4	0.31	86,000	3,600
RB	2.7	12.	0.30	150,000	3,700
313	1.3	2.2	0.12	24,000	1,300
IJ	1.2	3.7	0.037	42,000	420
03473	1.2	2.6	0.14	34,000	1,800
ML	$\sim 1.$	4.8	0.080	68,000	1,100

The terminal dose rates, multiplied by the number of days the activity was carried, have been listed here as the minimum lifetime doses. It is evident that tremendous doses have been received by these cells over a 20-, 30-, or 40-year period. Even in the diffuse distribution, these lifetime doses became quite respectable, and it must be recalled that almost every cell in bone receives at least this diffuse dose.

You will note the use of the term "minimum lifetime dose." Evidence is accumulating⁽³⁾ that activity is slowly lost from both the diffuse distribution and the hot spots in bone by the same mechanism, which we term a "long-term" exchange process, that deposited the diffuse originally. Hence, earlier in the lifetimes of these individuals, greater radium concentrations may have existed in both the hot spots and the diffuse distributions, and hence, greater doses than we measure terminally may have been delivered. In addition, if any of these cases contained significant fractions of Ra^{228} , which by now has decayed considerably as a result of its 6.7-year half-life, this contribution to the total dose is also missing for our evaluation.

When we examine this list of 19 cases and their associated bone doses it is natural to ask, "What effect did this dose have on this individual?" These cases, with one or two exceptions, were discovered to be radium cases as a result of their symptoms, so in this respect they are not representative of all radium cases. Only four of the list were without what may be considered to be radiation-induced tumors; these four are Q, I, 118, and 03473. It should be mentioned in this respect that in a current study at Argonne National Laboratory⁽⁸⁾ some 180 former dial painters, now living, have been located, and their body burdens measured. Studies of this nature, covering radium-burdened subjects located on the basis of industrial exposure, in contrast to these who were located as a result of their symptoms, may give a clearer indication of the probability that a given dose and/or dose rate, will induce recognizable radiation damage to the human being.

In summary, the following facts have been observed from a study of these 19 individuals who carried radium for many years:

1. Two distinctly different isotope distributions exist in each of these cases who were exposed for, at most, only a few years: an intense hot spot distribution, and a remarkably uniform diffuse distribution.
2. The magnitude of the diffuse distribution is, on the average, about one-half as great as that of the uniform label, the distribution that would exist if all the activity in the bone were uniformly distributed.
3. The hot spots in these cases contain about 75 times as much activity per unit bone mass as the diffuse distribution; (the values actually range from a low of 16 to a high of 218).
4. The lifetime doses delivered to osteocytes in their lacuna reach tremendous levels in the hot spots, in some cases several hundreds of thousands of rads, while all the osteocytes in bone receive doses ranging from a few hundred to a few thousands rads. It is our hope that from these human data we can eventually pick out the significant factors and then employ them to evaluate the hazards of strontium-90 to mankind on a firm, realistic, basis.

I wish to acknowledge the support of Dr. R. J. Hasterlik of the Argonne Cancer Research Hospital and Dr. A. J. Finkel of the Argonne National Laboratory, who have contributed most of the specimens used in this study. This work has been undertaken at the suggestion of L. D. Marinelli; to him and Dr. J. H. Marshall I am indebted for continual advice and criticism.

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Appendix

Bone Turnover from Terminal Hot-Spot Data

Let us define the turnover of bone by an exponential function

$$N = xt_1 e^{-xt_2}$$

Here N is the fraction of the total bone volume occupied by hot spots at the time of examination, t_2 years after exposure to radium; x is the yearly bone turnover rate; t_1 is the time, in years, of the exposure to radium, and $t_1 \ll t_2$, so $xt_1\%$ is the fraction of the labeled bone at the end of the exposure period.

The fraction of the original bone remaining after a given turnover rate has been operative for t_2 years is given by:

$$N_1 = e^{-xt_2}$$

From these two expressions we can calculate the fraction of the original bone remaining and the fraction of the total bone occupied by hot-spot deposits. The following table is based on an exposure (t_1) of one year and examination at the end of 30 years (t_2).

Table 6

Rate, % per year	Fraction (N) of bone that is hot spot after 30 years, %	Fraction (N_1) of original bone remaining after 30 years, %
10	0.5	5
5	1.1	22
4	1.2	30
3	1.2	40
2	1.1	55
1	0.74	74

THE RADIUM DISTRIBUTION IN THE TOOTH OF A DIAL PAINTER

R. E. Rowland

From a 55-year old dial painter (code 03473) who had been previously measured in the human body counter by C. E. Miller and assigned a body burden of $1.2\mu\text{c Ra}^{226}$, we have obtained a single tooth.

This tooth was removed on January 26, 1959, shipped, air dried, to this laboratory and identified by the dentist as an impacted mandibular third molar (wisdom tooth). Since this woman painted dials for a two-year period, from age 19 to 21, there appeared to be a good chance that this tooth was growing during this period, and thus may have acquired considerable radium during its formation.

The radium content of this tooth was evaluated by gamma-ray analysis, using an 8" x 4" NaI(Tl) crystal in the steel room. The tooth was sealed in glass and the total gamma-ray count evaluated after the radon daughters had equilibrated with the parent Ra^{226} .

The total Ra^{226} content of this tooth was found to be $2.1 \times 10^{-9}\text{c}$, or about 0.2 per cent of the total body radium content. The calcium content of this tooth was not measured, but if we assume a value of 0.4 g (35 per cent of 1.2 g total weight), then the specific activity is about $5 \times 10^{-9}\text{c/g Ca}$. This is somewhat greater than the value for the average body specific activity, $1.2 \times 10^{-9}\text{c/g Ca}$.

Since the work of Mays *et al.*⁽¹⁾ had shown that the radon retention of beagle teeth was quite low (9 to 13 per cent), the radon retention of this tooth was measured to see if the retention in human teeth was also low. On February 6, 1959, the tooth was sealed in a glass vial and a gamma-ray spectrum obtained. Subsequently, 83 days later, after equilibrium had been attained, the spectrum was remeasured and the retention found to be 76 per cent. This value was considered to be too high, probably due to a radon leak, and the entire procedure repeated. The second evaluation, between May 6, 1959, and June 18, 1959, (a period of 43 days) yielded a value of 73 per cent.

It thus appears that the radon retention in this particular tooth, 36 years after the radium was deposited, is close to 75 per cent. Whether this is a typical value for human teeth or not, we cannot judge at the present time. It is, however, very great compared to the low retention (9 to 13 per cent) Mays measured in the beagle teeth.

In order to study the distribution of radium within this tooth, it was embedded in methyl methacrylate and sectioned longitudinally with a high-speed, water-cooled rotary saw into slabs 0.010 inches thick. These

sections were first microradiographed, and subsequently several types of autoradiographs were made from each. Satisfactory autoradiographs were obtained by using Eastman Kodak autoradiographic plates, type A, for periods from one to three months. Stripping film was also placed on some sections; a two-month exposure produced autoradiographs of very high resolution which served to localize the radium deposits. From NTA* plates on which sections had been placed for periods of three to twenty-six days, quantitative alpha-track counts were made to evaluate the radium concentrations.

A very intense line of radium is visible in the dentine, Fig. 7, and adjacent to this line is a broad area of rather uniform deposition. A low diffuse level of activity exists in the dentine between this intense line and the enamel. The enamel itself contains very little activity, yet study of the stripping film shows a thin line of alpha tracks immediately below the surface of the enamel. Such a deposition has been shown by Sognaes⁽²⁾ to be due to salivary secretion; whether such a process takes place in an impacted tooth we cannot judge.

As can be seen from the microradiograph some bone is adjacent to the tooth, but, as indicated by the autoradiograph, this bone contains very little radium. A sequestrum had been removed from this patient's left mandible during July 1958, and has been quantitatively analyzed for radium.⁽³⁾ In Table 7 are listed the observed radium concentrations found in the tooth and bony sequestrum.

Table 7

Measured radium concentrations

Deposition	Bone	Tooth
Hot-spot	3.3 $\mu\mu\text{c}/\text{mg}$	
Diffuse	0.18 $\mu\mu\text{c}/\text{mg}$	
Dentine hot line		21.1 $\frac{\mu\mu\text{c}}{\text{mg}}$
Dentine diffuse		0.088 - 0.15 $\frac{\mu\mu\text{c}}{\text{mg}}$
Dentine uniform deposit		5.6 $\frac{\mu\mu\text{c}}{\text{mg}}$
Enamel diffuse		5 x 10 ⁻³ $\frac{\mu\mu\text{c}}{\text{mg}}$
Enamel line		0.3 $\frac{\mu\mu\text{c}}{\text{mg}}$
Ratio: $\frac{\text{hot spot}}{\text{diffuse}}$	18	176
Ratio: $\frac{\text{diffuse}}{\text{uniform label}}$	0.9	0.6

*Eastman Kodak Co.

Fig. 7 A microradiograph (a) and an autoradiograph (b) of a 200-micron thick tooth section. Five locations where alpha-track counts have been made are indicated. These locations are identified in Table 7 as follows: 1. Dentine hot line. 2. Dentine uniform deposit. 3. Dentine diffuse. 4. Enamel diffuse. 5. Enamel line. Regions 4 and 5 are identified on the microradiograph because the low level of activity in these regions left no visible darkening on this reproduction of the original autoradiograph.

The radium concentrations in the tooth were calculated from the alpha-track counts assuming 75 per cent radon retention and an average alpha-particle range of 6.3 mg/cm^2 in both the dentine and enamel. In Table 7 are listed concentration values for five locations in the tooth; these locations are indicated in Fig. 7. The ratios of hot-spot to diffuse and diffuse to uniform label for the tooth were calculated using an average value of the observed dentine diffuse, $0.12 \mu\mu\text{c/mg}$. The uniform label is defined as the concentration that would exist if all the radium in the body were uniformly distributed; in this case the value is $0.20 \mu\mu\text{c/mg}$.

It is evident that the concentration found in the hot line in the tooth is considerably greater than that found in the bony sequestrum. However, it may well be that more intense radium deposits do exist in the remaining bone of this patient; the sample available for study was very small. Nevertheless, it appears that the concentration in the tooth is probably greater than any concentration in bone, for no dial painter in our previous survey had a hot-spot to diffuse ratio greater than 102,⁽³⁾ the average value of this ratio for eleven dial painters being 43. (In contrast, the highest value for this ratio in patients who received radium intravenously was 218, the average value for five patients being 148.)

This high value is interpreted as follows: At the time of deposition, 36 years ago, radium substituted for calcium remarkably well, so that new mineral contained the same radium to calcium ratio as the interstitial fluid with which it was in contact. Thus both in bone and teeth one initially might expect the same radium to calcium ratio. In bone, however, a slow exchange of calcium takes place between the interstitial fluid and bone, which in time lowers the concentration of radium in both the hot spots and the diffuse distribution. It is this exchange, in fact, which is originally responsible for the diffuse distribution.

In the teeth, however, if this exchange took place at a very much lower rate, little loss of activity would be expected from the original concentrations. Thus, terminally, high concentrations should be found in the tooth and much lower concentrations in bone. Some exchange probably does take place within the dentine of the tooth since a diffuse distribution is apparent. If little loss has taken place from the dentine, then the diffuse distribution existing terminally is probably not greatly different in concentration from the original distribution.

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RETENTION AND PLASMA CLEARANCE OF THE ALKALINE EARTH ELEMENTS

I. Introduction

R. E. Rowland

The retention, R , of either calcium, strontium, or radium at any time, t , after administration to a dog may be described by a power function of the form

$$R = a t^{-b}$$

where a and b are constants. It is significant that the exponent, b , has a value close to 0.2 for each of these elements when radioactive tracers are intravenously administered to dogs a year or so of age.

Directing our attention to the human being, we find the little information available seems to indicate that, although the retention of these elements may still be described by a power function, the value of the exponent, b , may vary with the element studied.

In order to summarize present knowledge of the retention of the alkaline earth elements, information available from rather extensive studies in dogs is reviewed and presented in Part III of this report. Data describing the plasma clearance rates of these tracers are included to show the relationship between the retention and the plasma clearance.

To overcome the lack of information found in regard to the retention and plasma clearance of barium in the dog, new data obtained from studies in this laboratory are presented in Part II; they are also included in the summary in Part III.

Finally, in Part IV, the available data describing retention and plasma clearance of these elements in the human being are discussed.

II. The Retention and Plasma Clearance of Barium in the Beagle

R. E. Rowland and C. H. Williams

In order to ascertain whether barium behaves in a manner similar to the other alkaline earth elements following intravenous injection into the dog, we measured the plasma clearance and retention of barium in two beagles. Ba^{133} was employed as the tracer. This isotope has a half-life of 7.2 years, decays by electron capture, and emits several gamma rays. Thus its

half-life is excellently suited for long retention studies, the radiation dose is minimal, for no beta particles are emitted, and the body burden is easily determined by means of scintillation counters. On the other hand, barium is a rather toxic material, and since Ba^{133} is not available carrier free, stable barium is injected with the tracer.

The injection material was in the form of BaCl_2 in 0.075 N HCl, at a concentration of $10\mu\text{c}/\text{ml}$, with a stable barium level of 10 mg/ml of solution.

To test the toxicity of barium, a 4.75-kg mongrel puppy was injected with 5 mg of BaCl_2 in 0.1 N HCl, which was neutralized immediately before injection with 0.1 N NaOH. A total volume of 1 ml was injected into the external jugular vein over a period of 3 minutes, while the animal was anesthetized with Nembutal. This animal seemed to tolerate this level of barium quite nicely, and it appeared that we could safely inject quantities of stable barium of the order of 1 mg/kg body weight.

Two beagles were used in the study. The first, No. A22B, was 8 months old (245 days), and weighed 7.2 kg at time of injection. This dog received $6.1\mu\text{c Ba}^{133}$, as described above, neutralized with NaOH immediately before injection. The second dog, No. 576, weighed 10.1 kg

and was of unknown age. This dog had been acquired $2\frac{1}{2}$ years earlier by the Laboratory, and was thought to be over a year old at that time, making him about 4 years old at the time of injection. This animal received $10\mu\text{c Ba}^{133}$ as previously described.

Fig. 8 Observed values of the concentration of Ba^{133} in the plasma of two beagles following intravenous injection of this isotope.

The plasma clearance studies were performed by measuring the Ba^{133} content of whole blood and assuming that the plasma content of the blood was constant. At each sampling of the blood, about 4 ml were obtained in a heparinized syringe from which 3 ml were removed, sealed in a glass vial and counted in a NaI(Tl) well counter. The observed values, plotted in Fig. 8, were calculated by comparison with two standards made from the injection solution.

The body burdens were measured by placing a 5" x 4" NaI(Tl) crystal, connected with a

256-channel analyzer in a reproducible position with respect to the dog in the steel room of the Division of Biology and Medicine. Immediately after the injection, each animal was counted, so that the original count included all of the injected activity. All subsequent counts were expressed as a fraction of the original count, and are so plotted in Fig. 9.

Fig. 9 Measured retention of Ba^{133} in two beagles following intravenous injection of this isotope.

Ba^{133} emits two strong gamma rays, one at 81 kev and one at 360 kev, as well as other less intense gamma rays. It was of interest to note that the shape of the spectrum, as recorded by this particular NaI(Tl) crystal, remained unchanged throughout the period of observation, namely minutes to months after the injection. This indicates that the effective absorption and scattering of the gamma radiation was essentially the same after the isotope was deposited within the bone as it was when the isotope was in the blood. Thus, in this case, the technique of recording an original burden measurement immediately after the injection, before any excretion has taken place, is valid as the 100% point, and the spectrum thus obtained may be compared directly to later spectra, even though the location of the isotope within the dog is not the same at later times.

Due to scheduling of the steel room, we were unable to count the dogs again until the 20th day after injection. After that time we were able to get weekly body burdens as long as it was necessary. This did mean, unfortunately, that we were unable to obtain the retention at one day, and thus obtain directly the value of the term, a , in the power function retention formula.

The measured values of the fractional retention are plotted for each dog in Fig. 9. It can be seen that the younger of the two animals retained more of the tracer than the elder, an observation in agreement with observations on strontium retention in dogs of various ages.⁽¹⁾

The retention and the plasma clearance are expressed in Table 8 as power functions, where the time (t) is in days. The function for the retention, in terms of fractional retention, is based on the observations from 20 to 188 days. It will be noted from the value of the a term for the younger

dog, 1.36, that the data cannot be extended back to the first day as a straight line on the log-log plot. The function for plasma clearance expresses the concentration in per cent of injected dose per ml plasma after the first day. Actually the plasma data do not fit a power function well until after the second day; the value at $t = 1$ day was obtained by extending the straight line which fits the subsequent data back to this time.

Table 8

Retention and plasma clearance of Ba^{133} in two beagles

Dog	Age	Retention, fraction of injected dose	Plasma concentration, % of injected dose/ml plasma
A22B	245 days	$1.36 t^{-0.24}$	$1.72 \times 10^{-3} t^{-1.27}$
576	~4 yr	$0.96 t^{-0.23}$	$5.15 \times 10^{-3} t^{-1.28}$

The significance of these results will be discussed in Part III, where we compare the behavior of the other members of the alkaline earth family.

We wish to acknowledge the cooperation of P. F. Gustafson and S. Brar in the measurement of the body burdens of these animals. We are also extremely grateful for the assistance given by R. J. Flynn and D. D. Banister during the injections and subsequent care of the animals.

III. Retention and Plasma Clearance of the Alkaline Earth Elements in Dogs

R. E. Rowland

Excellent data on the behavior of radium and strontium in beagles are available from the studies conducted at the Radiological Laboratories at the University of Utah. In Tables 9 and 10 are listed the published power functions which describe the retention and plasma concentrations following intravenous injection into adult animals.

Table 9

Retention of the alkaline earth radioelements in dogs

Isotope	Breed	Age	Retention	Reference
Ca^{45}	Mixed	Adult	$0.76 t^{-0.2}$	2
Sr^{90}	Beagles	17.7 mo	$0.68 t^{-0.21}$	4
Ra^{226}	Beagles	Av = 1.3 yr	$0.787 t^{-0.198}$	5
Ba^{133}	Beagle	245 days	$1.36 t^{-0.24}$	This report
Ba^{133}	Beagle	~4 yr	$0.96 t^{-0.23}$	This report

Table 10

Plasma concentrations of the alkaline earths in dogs

Isotope	Breed	Age	Plasma concentration	Reference
Ca ⁴⁵	Basenji and cocker	2 yr	$14.5 \times 10^{-3} t^{-1.24}$	3
Sr ⁹⁰	Beagles	16-17 mo	$5.03 \times 10^{-3} t^{-1.25}$	7
Ra ²²⁶	Beagles	1.3 yr	$2.64 \times 10^{-3} t^{-1.24}$	5
Ba ¹³³	Beagle	245 days	$1.72 \times 10^{-3} t^{-1.27}$	This report
Ba ¹³³	Beagle	~4 yr	$5.15 \times 10^{-3} t^{-1.28}$	This report

The thesis of Maletskos⁽²⁾ contains data describing the retention of Ca⁴⁵ in dogs in which his results are described as sums of exponential functions from which is calculated a power function from the data for the retention between 20 and 100 days.

The plasma concentrations for Ca⁴⁵ following intravenous injection into dogs were obtained from unpublished observations of Jowsey *et al.*⁽³⁾

As can be seen from Table 9 the retention of all of these alkaline earths may be described by power functions, and in each case the exponent is very close to 0.2. The only significant change is in the value of the a term; the variations are clearly shown in Fig. 10.

Fig. 10 Retention of four alkaline earth tracers in adult dogs. The strontium and radium curves are the published power functions of the Utah group (see text); the calcium curve is our power function fit to the data of Maletskos;⁽²⁾ and the barium data are from the four-year-old beagle, No. 576.

The fact that this term is age-dependent has been previously mentioned;⁽¹⁾ in addition, Kornberg⁽⁶⁾ has described an experiment with rats in which the value of this term, describing the retention of Sr^{90} after interperitoneal injection, was dependent upon the calcium content of the diet. Hence, these two variables, and perhaps others, can change the value of the constant term, a .

Kornberg also states that the retention of Sr^{90} by the rats may be described by a power function with exponent 0.2, and that this exponent was unaffected by the calcium content of the diet. This was not the case when the retention of Ca^{45} was studied, for the value of the exponent in the power function was found to be dependent on the calcium content of the diet, decreasing in magnitude with decreasing diet calcium. This is a significant difference in behavior between these two elements which has not been investigated in the dog.

When attention is given to the power function description of the plasma concentrations after the first day for these radioelements, another marked similarity is evident. In each case, as tabulated in Table 10, the plasma concentration may be described by a power function, and the exponents found for each alkaline earth isotope are very nearly equal. These data are presented in graphical form in Fig. 11 for adult dogs.

Fig. 11

Summary of the clearance of four alkaline earth tracers from the plasma of adult dogs following intravenous injection. Strontium and radium curves are those published by the Utah group (see text); the calcium curve is taken from unpublished data from our Laboratory;⁽³⁾ and the barium data are from our four-year old beagle, No. 576.

Thus it is apparent that both the retention of these alkaline earth elements, when present in tracer quantities, and their clearance from the plasma, show little change from element to element. Even more impressive is the fact that the shape of the plasma clearance curve is predictable from the retention curve; that is, the value of the exponent in the power function

describing the plasma concentration is very nearly equal to the value of the exponent in the time derivative of the expression for the retention of these elements.

IV. The Retention and Plasma Clearance of the Alkaline Earths by Human Beings

R. E. Rowland

Having noted that the first derivatives of the equations describing the retention of alkaline earths in dogs are very closely related to the functions describing the plasma clearance of these elements, we inquire whether this relationship might also hold for the human being. The literature was searched for data describing the retention and plasma clearance in human beings of intravenously injected alkaline earth isotopes. These data are discussed below, grouped according to the element studied.

Calcium-45

Several authors have published data describing the removal of Ca^{45} from human plasma following intravenous injection.⁽⁸⁻¹¹⁾ These data are

Fig. 12 A summary of the plasma clearance of Ca^{45} for five human cases, following intravenous injection of this isotope.

plotted in Fig. 12 expressed in units of per cent dose per mg Ca . The data of Bronner and Harris⁽¹¹⁾ were plotted in their paper transformed to a body weight of 35.6 kg. These have been transformed back to the original weight of the 21-year old male subject, 51.4 kg, by multiplying their data by the ratio of weights, 35.6/51.4. Also included in Fig. 12 are unpublished data from our Laboratory⁽¹³⁾ describing the clearance of Ca^{45} from a 51-year old male radium patient, (Case BM).

It will be noted from this figure that these plasma clearance curves do not become definite straight lines, indicative of a power function, during the time over which observations have been made. It appears as if plasma clearance would have to be

followed out to about 2000 hours (80 days) to prove definitely that such a behavior did or did not exist. However, if one defines the best straight line between 220 hours and 672 hours for the combined data from references 11 and 13, it appears to have a slope approximately equal to $t^{-1.2}$.

The best data found for the retention of Ca^{45} in a human subject are those of Bronner *et al.*,⁽¹²⁾ who calculated the retention in a 21-year old male (same case as reference above) for 60 days from excreta studies. These are plotted in Fig. 13 and may be described by a power function

$$R = 0.96 t^{-0.12}$$

Fig. 13

The retention of calcium, strontium, and radium in human patients. The calcium data are from Bronner *et al.*,⁽¹²⁾ the strontium data from Van Dilla *et al.*,⁽¹⁶⁾ the two radium cases from Seil *et al.*,⁽¹⁸⁾ In all of these cases the retention was calculated from excreta measurements. Also plotted is the power function for the retention of radium from the observations of the Elgin State Hospital cases.⁽¹⁹⁾

From the derivative of this expression, one expects a plasma clearance described by $t^{-1.12}$, a value perhaps not greatly different from the value mentioned above, $t^{-1.2}$. While these seem to be in fair agreement, they certainly cannot be used to illustrate the equality of the first derivative of the retention equation to the slope of the plasma clearance equation.

Strontium

The removal of strontium from the plasma of human patients has been described by several authors. In Fig. 14 average values for these different groups given intravenous Sr^{85} are plotted. Two of these sets of data were kindly supplied by Dr. H. Spencer;^(14,15) the third is from the work of Van Dilla *et al.*⁽¹⁶⁾

Also plotted in Fig. 14 is the disappearance of stable strontium from the plasma of a 40-year old, 85-kg male subject, following the injection of

100 mg Sr as SrCl. This study by Harrison *et al.*⁽¹⁷⁾ indicates quite effectively that this larger quantity of stable strontium is cleared from the plasma at the same rate as the much smaller isotopic tracers.

Fig. 14 Plasma concentrations following intravenous injection of strontium in human patients.

The data of Van Dilla *et al.*,⁽¹⁶⁾ which extend out to 600 hours, seem to be approaching a straight line of slope $t^{-1.5}$, but data are needed for longer periods to verify this.

The retention of Sr^{85} in these five cases was measured during the period from 5 to 15 days, and in one case, 30 days after injection by measurement of total Sr^{85} excreted. In Fig. 13 the retention for each of these cases is plotted; each case seems to be described by a straight line over this interval, but the slopes differ from $t^{-0.16}$ to $t^{-0.44}$.

A study of the terminal distributions of Ca^{45} and Sr^{85} , as described by Schulert *et al.*⁽²⁰⁾ in a series of ten terminal patients who survived $\frac{1}{8}$ to 124 days after administration of the tracers, shows that a considerable fraction of the injected dose remains in the soft tissues many weeks after injection. Table 11, based on their published data, expresses the fraction of the retained dose found in the soft tissues of each patient.

Table 11

Percentage of retained calcium and strontium tracers in the soft tissues of terminal patients after intravenous administration

Patient	H.H.	S.K.	S.J.	C.C.	J.Z.	G.B.	S.G.	T.M.	A.W.	M.S.
Survival, days,	$\frac{1}{8}$	2	3	3	6	18	30	39	65	124
Ca^{45}	52	40	34	42	35	28	7.9	7.2	1.5	4.5
Sr^{85}	63	40	55	41	38	35	5.7	5.2	5	0.6

This high tissue fraction of the retained burden in man is in marked contrast to the situation in the dog. Glad *et al.*⁽¹⁾ described a double tracer

experiment with Sr^{85} and Ra^{226} in which a beagle was sacrificed 8 days after an injection of the two isotopes. At that time only 2.7% of the retained Sr^{85} and 7% of the retained Ra^{226} was in the soft tissues. This might account for some of the observed differences between the two species. Perhaps one should not look for power-law behavior in the plasma clearance rate until almost all of the body burden is deposited within the skeleton. If this were the case, then the expected relationship between the retention and plasma clearance rate would only be valid for a skeletally-bound isotope.

Thus it appears that the most interesting data describing the retention and plasma clearance of these isotopes will be found after the first 100 days. No such data are available for humans.

Radium

Radium is the isotope with which human beings have had the most experience, and it is on this experience that we base a good part of our knowledge of the effects of bone-seeking radioisotopes on man. Yet, nowhere in the literature are there any data describing the clearance of this element from human plasma.*

The retention of radium in human beings has been based on the measurements of the Elgin State Hospital patients;⁽¹⁹⁾ from them has been obtained the well-known expression for their retention of radium,

$$R = 0.54 t^{-0.52} ,$$

which is plotted in Fig. 13.

Recently, Marinelli et al.⁽²²⁾ have described the retention and excretion of Ra^{226} from a young adult male who accidentally inhaled RaSO_4 . Their retention data, after the first 100 days, are described by a power function with $t^{-0.78}$, while the excretion is found to follow a power function with $t^{-1.78}$. These data, covering a period of 6.5 years, show definitely the relationship between the retention function and its differential, the excretion function, but only after the isotope had transferred from the lung to the skeleton.

* Aub et al.⁽²¹⁾ measured the radium in the blood of a dial painter who was receiving medication in attempt to release radium from the skeleton. The woman had painted watch dials for a seven-year period, and at the time of treatment, seven years later, retained $18 \mu\text{c}$ of radium. If this had been a case of intravenous injections, the power function $R = 0.54 t^{-0.52}$ would predict a retention of three quarters of one per cent of the injected dose ten years later. Assuming the mid-point of the exposure period as the equivalent time of injection the injected dose would have been $2400 \mu\text{c}$. If radium is cleared from the plasma in a manner similar to Sr^{90} , the plasma values will be described by the equation $P_t = 38 t^{-1.5}$ per cent of dose/g Ca. Using this function the plasma activity is calculated to be 0.21×10^{-12} c/cc of blood ten years after injection. The measured values were about 1×10^{-12} c/cc blood, a factor of five greater than the predicted value. Since the medication was shown to increase the daily elimination of radium by factors of 3 to 6, this measured value may be interpreted as being remarkably close to the value predicted by a power-function behavior of the plasma concentration.

It should be noted that Marinelli *et al.*⁽²³⁾ in a description of the elimination of radium from several patients following accidental inhalation, pointed out that not all of their measured body burdens could be assigned to the skeleton or lung, so that a significant fraction of the measured radium might be in the soft tissues. This was based on the assumption that only skeletal radium released radon, and that this release was 70% of the total found. Later work of Mays *et al.*⁽²⁴⁾ has shown that shortly after acquisition, about 90% of canine skeletal radon is released. If this is also true for the human being, the increased radon release implies even more soft tissue radium than Marinelli had estimated and lends considerable weight to his suggestion.

One set of data is available in the literature describing the early retention of Ra^{226} following intravenous injection of this element. In 1915 Seil, Viol, and Gordon⁽¹⁸⁾ described the retention of radium in a 23-year old male, following intravenous administration of $100 \mu\text{c Ra}^{226}$. They measured the total excreta (and incidentally found a fecal to urinary ratio of about 10:1) and based their retention on this. They repeated this study on the same individual two months later, and two sets of retention data are plotted in Fig. 13.

The curves plotted from their data can be seen to be similar in slope to that obtained for Ca^{45} ; the data of Seil actually fit a power function with $t^{-0.11}$ in one case, and $t^{-0.16}$ in the other, while the Ca^{45} data are described by $t^{-0.12}$. These data contradict the

Elgin patients' power function, as can be seen in Fig. 13, yet we must recall that this power function was essentially determined by the retention at 20 years and the known dose, but not on the basis of any data within the first few weeks.

Fig. 15 Plasma concentrations following intravenous injection of Ba^{140} in human patients.

Barium

Here the only published study discovered is that of Bauer,⁽²⁵⁾ which describes the plasma clearance of Ba^{140} following an intravenous injection of BaCl_2 in a 25-year old woman. Bauer also kindly supplied the plasma values for a normal 32-year old male (Case 18 in the above reference). These are plotted in Fig. 15. It can be seen

that plasma measurements would have to be taken for a much longer time in order to determine whether or not a power law fits.

Summary

From this review of the retention and plasma clearance of intravenously injected tracers of the alkaline earth family, it becomes apparent that the power law formulation for the retention of these elements is valid for the dog, and that the differential of this expression has the same slope as the power law describing the plasma concentration of the tracer. In the human, however, the data seem to indicate that these relationships do not hold during the first few weeks following intravenous administration of an alkaline earth tracer. It is suggested that after the first three or four months a power law probably will describe the retention, and that its derivative, also a power law, will describe the plasma concentration or excretion function.

The lack of agreement at early times may, in part, be due to the fact that the body burden is not entirely a skeletal burden. A power law should probably be expected to describe only the loss of activity from the skeleton. Yet to be resolved is the basic question: can the retention of all of these alkaline earth elements in the human being be described by power functions with constant slope, (i.e., constant value of the exponent, b), such as is the case for the dog, or is b a function of the isotope employed?

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AUTORADIOGRAPHIC DOSIMETRY OF MOUSE BONES CONTAINING Ca^{45} , Sr^{90} , AND Ra^{226}

J. H. Marshall and M. P. Finkel

The tumor incidence in CF No. 1 female mice injected with Ca^{45} , Sr^{90} , or Ra^{226} has previously been reported in terms of injected radioactivity per unit of body weight.^(1, 2) In order to interpret these data in terms of energy absorbed rather than microcuries injected, it is necessary to know the microscopic dose distributions at various times after injection. For this purpose quantitative autoradiographic measurements of microscopic dose rate were made in the femur, rib, lumbar spine, skull, ilium, and humerus in animals killed from 15 minutes to 18 months after they had received a single intravenous injection of one of these nuclides. It was anticipated that differences in the locations of the resulting osteogenic sarcomas might be explained, at least in part, by such detailed dosimetry. Also, it seemed that comparisons of the patterns of distribution of the doses delivered by these nuclides, which have similar biochemical properties but very dissimilar radiations, might provide information on the site of tumor induction. The present report describes the progress of these studies.

Methods

Eighty CF No. 1 female mice, which were comparable to those used in the long-term comparative toxicity experiments,^(1, 2) were injected intravenously with Ca^{45} , Sr^{90} , or Ra^{226} when they were approximately 70 days old. The injection levels were selected on the basis of their carcinogenicity, as determined in these earlier experiments. In the Ra^{226} series approximately $0.03 \mu\text{c/g}$ body weight had resulted in the greatest incidence of osteogenic sarcomas. It was estimated from the dose-response curves that a similar incidence would be expected after the injection of $0.4 \mu\text{c Sr}^{90}/\text{g}$ and $6.0 \mu\text{c Ca}^{45}/\text{g}$. Twelve mice were injected with each nuclide and were sacrificed in groups of three, 4 hours, 3 days, 30 days, and 59 days later. It was necessary that animals scheduled for later sacrifice receive lower doses because these highly carcinogenic levels are associated with a marked decrease in life expectancy. Consequently, one-tenth of these dose levels were given to 30 mice scheduled for sacrifice in 3 months, 12 months, and 18 months. In addition, in order to measure more accurately the removal of activity from the growing distal end of the femur, 14 mice were injected with $0.25 \mu\text{c Ca}^{45}/\text{g}$, and these were sacrificed 15 minutes, 1 hour, 4 hours, 23 hours, 3 days, 6 days, 10 days, 15 days, and 30 days later. Since the measured dose rates were found to be proportional to the amount of activity injected, the data for each nuclide have been normalized to a single level.

The fresh bones were ground on sand paper to an approximate plane of symmetry so that subsequent autoradiography could properly take into account the radiation from all parts of the bone. This was especially necessary in the case of Sr^{90} - Y^{90} . The bones were then dehydrated in acetone and embedded in Scotchcast* which has a density of 1.34 g/cm^3 , and which thus served not only to support the bone but also to fill the empty spaces with a material not too unlike soft tissue. The Scotchcast blocks were milled down until the proper plane of the bone was exposed. This was the mid-sagittal plane of the femur, humerus, and skull, the sagittal or frontal plane of the lumbar spine, the frontal plane of the ilium, and a cross section approximately 2 mm thick of the rib.

The embedded bones were lightly clamped to emulsion-coated microscope slides supplied by Eastman Kodak Company. Autoradiographic type A plates with T coat were used for Sr^{90} and Ca^{45} . NTA plates with an emulsion thickness of 25μ were used for Ra^{226} . Several different exposures were used for each block, ranging from $\frac{1}{2}$ hour for the highest levels of Ca^{45} and Sr^{90} to 3 months, which was necessary to produce sufficient alpha-track darkening on NTA film for densitometer measurements, for some of the Ra^{226} exposures. Films exposed for more than a few days were kept in a freezer, and calibration exposures before and after each experimental exposure showed that fading was usually negligible. Radium exposures short enough for alpha-track counting were also made in order to check the densitometer measurements. Similar plates were exposed to plaster-of-Paris radiators containing known concentrations of the three radioelements, as described by Dudley,⁽³⁾ and these were developed along with plates that had been exposed to bone.

Densitometry was done with a microscope-densitometer-Brown recorder combination. The effective aperture was 30μ . Plots of autoradiographic darkening versus exposure were constructed for Sr^{90} and for Ca^{45} so that the data could be corrected for any nonlinearity with exposure. The Ra^{226} data were calibrated and checked for linearity by counting alpha tracks at both high and low levels of exposure. The following alpha-particle ranges were used for the calibration calculations: 28.1μ for bone with 30 per cent Rn retention and $\rho = 2 \text{ g/cm}^3$; 28.0μ for plaster of Paris with 100 per cent Rn retention (which had been measured) and $\rho = 2.32 \text{ g/cm}^3$.

The relation between autoradiographic darkening and dose rate for Ca^{45} and Sr^{90} was obtained by exposing films to plaster-of-Paris radiators containing these isotopes, as described above. The activity per gram in bone from a thick, uniform source can be derived directly by comparing the film darkening produced by the activity in the bone with that produced by the radiator.⁽⁴⁾ The relation between dose rate and microcuries per gram in such a source follows directly from the conservation of energy.⁽⁵⁾

* Scotchcast Resin No. 2, Minnesota Mining and Manufacturing Co., St. Paul, Minnesota.

In principle, a given beta-particle flux will deliver different doses to soft tissue and to bone because their effective atomic numbers are different. However the difference is small (10 per cent or less) and depends in a complex way upon the size of the soft tissue cavity. Therefore, no correction has been made for the effect of atomic number on beta-particle dose, and the dose to soft tissue for a given film darkening is considered to be the same as that to bone.

In the procedure described above for determining dose rate from autoradiographic darkening, the assumption is made that the angular distribution of beta rays from a non-uniform source is the same as that from a uniform source. Dudley⁽³⁾ has shown that the sensitivity of type A film is not the same for perpendicularly incident and diffusely incident beta particles. For Sr^{90} - Y^{90} the difference is small, but for Ca^{45} , beta particles that approach the film perpendicularly are about twice as effective in darkening it as beta particles that approach from all angles. For this reason, the measured peak Ca^{45} dose rates may be somewhat exaggerated. However, this exaggeration should not be large because the peak dose rates correspond not to point sources but to surfaces of Ca^{45} activity from which the beta particles approach the film at many angles. Dudley's dosage-sensitivity curves for type A film also indicate that Sr^{90} - Y^{90} should produce about twice as much darkening per rad as Ca^{45} . Similarly, we find that our type A plates are 2.2 times as sensitive to Sr^{90} as to Ca^{45} . However, no adjustment for this fact is required with the calibration method we have used.

Corrections for the radioactive decay of Ca^{45} and Sr^{90} have been made so that the reported values of dose rate correspond to the time of sacrifice of the animal. However, no correction has been made for the early growth of Y^{90} or for the fact that the Y^{90} originally contained in the injection solution was deposited in an yttrium-like pattern. While the initial Y^{90} decayed with a 64-hour half-life, new Y^{90} , with a strontium-like deposition pattern, grew into the bone. Since the range of Y^{90} beta rays is large, this difference in deposition pattern should not affect the dosage pattern substantially; the reported values correspond to complete retention and build-up of Y^{90} at Sr^{90} sites.

The dosimetry problems with Ra^{226} are somewhat different from those with the beta emitters. Spiers has calculated the dose from the alpha particles emitted by radium and its daughters delivered to a small tissue cavity surrounded by bone ⁽⁶⁾ He found that the ratio of the dose in a small soft-tissue cavity to that in the surrounding bone is 1.36. In addition to this factor, we have used a cavity size correction of 0.85 calculated by Kononenko,⁽⁷⁾ so that the reported dose rates for Ra^{226} refer to the average dose rate in a $10\text{-}\mu$ lacuna within bone. Values for radon retention and radon buildup as a function of time after injection have been taken from the compilation of Mays.⁽⁸⁾ Since these values fit the known data for man, dog,

and rat, they probably describe the situation in mice as well. These values are 1.5 per cent at one day, 9.5 per cent at 10 days, 18 per cent at 100 days, and 22 per cent at 1000 days. Since radon retention in embedded bone is somewhat greater than in living bone,⁽⁹⁾ 30 per cent retention has been used for the embedded specimens during autoradiography.

Results

A gross measure of the retention of Ca^{45} , Sr^{90} , and Ra^{226} in the femur as a function of time after injection was obtained by counting all of the embedded femurs with a thin end-window Geiger tube. The slopes of the three retention curves (which were arbitrarily normalized) are compared in Fig. 16 with the whole-body retention of Sr^{90} in a similar group of mice.⁽¹⁰⁾ The slopes of Sr^{90} retention by the whole body and by the femur alone are very similar. Ra^{226} retention in the femur followed the same slope, but the slope for Ca^{45} retention was somewhat less. For comparison with the autoradiographic data, these curves have been replotted in Figs. 17, 18 and 19 (corrected for radioactive decay or for radon buildup *in vivo*, as necessary). The dose rates were normalized by calculating the average energy emitted per gram of bone, with the assumption that skeletal weight was 10 per cent of body weight. The retention of Sr^{90} had been found to be $0.77 t^{-0.31}$, as shown in Fig. 16. A value of $0.8 t^{-0.13}$ was used for Ca^{45} . This was based on the slope shown in Fig. 16 and an assumed retention at one day slightly greater than that of Sr^{90} . For radium a retention of $0.7 t^{-0.3}$ was used, based on the slope in Fig. 16 and an assumed retention at one day slightly less than that of Sr^{90} .

Autoradiographic dosimetry of Ca^{45} , Sr^{90} , and Ra^{226} in the femur as a function of time after injection is summarized in Figs. 17, 18, and 19. A few of the autoradiographs upon which these curves are based are shown in Fig. 20. Each point on the curves is the average from two different exposures of the femurs of two to three animals. The curve labelled "Distal Plate" gives the maximum dose rate just below the distal epiphyseal plate. The curve labelled "Trabecular Max" gives the maximum dose rate in either the proximal or distal ends, which were comparable in all cases. The curve labelled "Trabecular Average" gives the average dose rate in the ends of the bone except for the distal epiphyseal plate; these values were obtained by scanning the area and taking the average from the tracing on the Brown recorder. "Metaphysis Max" and "Shaft Max" are self-explanatory. The curve labelled "Diffuse" gives the dose rate from the diffuse component which was measured in the shaft well away from any periosteal or endosteal deposition. For Sr^{90} , cross radiation prevented such a diffuse measurement, so the curve for this nuclide represents the minimum dose rate measured at the center of the cortical bone somewhere along the length of the shaft. The dashed curve, "Calculated Average," gives the average skeletal dose (without allowance for escape) based on the retention values described above.

Fig. 16. Relative retention of femur determined by thick source beta counting and whole body retention of Sr^{90} in mice. Upper Sr^{90} curve, whole-body retention; lower three curves, relative retention of femur.

Fig. 17. Cell dose rate in femur - Ca^{45}

Fig. 18. Cell dose rate in femur - Sr^{90}

Fig. 19. Cell dose rate in femur - Ra^{226}

Fig. 20
Autoradiographs of femurs of
mice that received Ra^{226} ,
 Sr^{90} , or Ca^{45} .

There is no evidence of nonproportionality between dose rates as shown in these figures and the injected activity per unit body weight; (remember that the curves for Sr^{90} and Ra^{226} include points determined from animals with injections differing by a factor of 10, and those for Ca^{45} by factors of 10 and 24). Therefore, values for the dose rates at lower injection levels can be obtained as simple ratios from the given values.

The most striking feature of the femur dosimetry is the intense deposition of the three radioelements just below the distal epiphyseal plate, where calcification accompanying longitudinal growth was taking place. The rate of growth of the femur during this period (most of this growth took place at the distal end) was $33 \pm 6 \mu$ per day. The average femur length at the time of injection was 14.6 mm; thirty days later it was 15.6 mm; and by 100 days it had almost reached its final length of 16.3 mm. Since the width of the calcifying zone was roughly 300μ , by the tenth day after injection, when growth of about 330μ had occurred, much of the originally-deposited activity beneath the plate had been resorbed. The loss of activity in this region is most clearly demonstrated by curve "P" in Fig. 17. Although the total dose to the epiphyseal area was decreasing rapidly during the first 10 days the maximum dose rate did not fall off until the entire plate had been replaced. By 50 days the maximum dose rate in this area had fallen below the diffuse dose rate in the shaft because the specific activity of the newly calcified bone depended upon the specific activity of the blood, which was constantly decreasing during this time. Although removal of the three radioelements was controlled primarily by the same resorption process, the maximum dose rate from Sr^{90} began to fall earlier than the maximum dose rates from Ca^{45} and from Ra^{226} . This was due to the fact that every portion of the epiphyseal plate received cross radiation from Sr^{90} located in all other parts of the plate, so resorption from any portion of the plate decreased the maximum dose rate day by day. The dose rates from Ca^{45} and Ra^{226} followed local specific activity much more closely and were not influenced by resorption occurring in more distant parts of the calcified layer. Whether exchangeable activity also influenced the dose rates could not be determined with certainty.

Throughout the femur the dose rate from Sr^{90} fell steadily during the 18 months of the experiment, largely because of the general decrease in cross radiation as described above. On the other hand, the maximum dose rates from Ca^{45} and Ra^{226} remained rather constant after the first few hours. Although resorption reduced the average dose rate by reducing the amount of bone containing the maximum deposits, it did not reduce the maximum dose rate. Some of the dose rates given for radium increase with time, but this increase is largely due to the increasing values of in vivo radon retention used in the dose calculation. The general fall of the Ca^{45} dose rates is due to radioactive decay (half-life 163 days).

Two general features of the three sets of curves should be pointed out. First, the ratio of the maximum to the minimum dose rate at any given time after injection is a measure of the degree of non-uniformity of the spatial distribution of the dose rate. This non-uniformity factor is smallest for Sr^{90} and greatest for Ra^{226} . Second, the position of the calculated average dose rate relative to the dose rates measured autoradiographically is quite different for Sr^{90} from that for Ra^{226} and for Ca^{45} . The autoradiographically determined curves for the latter two isotopes are quite close to the calculated curves, but in the case of Sr^{90} they are 2 or 3 times lower. This is a direct effect of the escape of Sr^{90} - Y^{90} beta particles from the bone. As Lamerton⁽¹¹⁾ has pointed out, the maximum Sr^{90} dose rate in the mouse skeleton is comparable to this calculated average (which neglects escape). For Ca^{45} the maximum dose rates are 5-7 times and for Ra^{226} 10-20 times the calculated average.

The dose rates from the three nuclides three days after injection are shown along the longitudinal axis of the femur in Fig. 21 and along a cross section through the mid-shaft in Fig. 22. The dose rates 59 days after injection in a cross section through the metaphysis 3 mm from the distal end of the femur, are illustrated in Fig. 23. The low-level alpha activity in the marrow cavity and outside the shaft in Figs. 22 and 23 is probably due to diffusion of radon in the Scotchcast and would not be expected in vivo.

The maximum dose rates measured in some other parts of the skeleton are shown in Fig. 24, where, for comparison, one curve from Figs. 17, 18, and 19 has been included. In general, the peak dose rates from Ca^{45} and Ra^{226} in other bones were comparable to those in the femur. For example, the maximum dose rate from Ca^{45} in the humerus, just below the proximal epiphyseal plate was very close to that in the femur. In the case of Sr^{90} , however, the smaller bones had lower peak doses. For example, the maximum dose rate in the rib corresponded to the minimum dose rate in the femur shaft. Autoradiographs of the lumbar spine, rib, skull, and ilium appear in Figs. 25 through 28. The dose rates in the rib and lumbar spine 3 days after injection are illustrated in Figs. 29 and 30. (This particular profile of a Ca^{45} lumbar spine did not include the maximum dose rate that had been observed, but it was typical in other respects.)

Discussion

The locations of the tumors that were observed in the comparative toxicity experiments are listed in Table 12. A number of marked differences were noted, some of which may have been due to variations in sampling, but several of which appeared to be significant. The femur, for example, contained 35% of the Sr^{90} tumors but only 19% and 8% of the Ca^{45} and Ra^{226} tumors, respectively. In contrast, the lumbar spine contained similar proportions of the total tumors induced by the three isotopes, while the rib was the site of

Fig. 21
Femur axis at 3 days

Fig. 22
Femur midshaft
profile at 3 days

Fig. 23
Femoral metaphysis
at 59 days section
3 mm from distal tip

Table 12

Anatomical distribution of osteogenic sarcomas⁽²⁾

Location	Percentage distribution by isotope		
	Radium-226 (% of 154*)	Strontium-90 (% of 352)	Calcium-45 (% of 182)
Femur	8.4	35.4	19.2
Tibia**	5.8	19.6	8.8
Humerus	4.5	3.4	1.6
Radius-ulna	0.0	0.3	0.5
Foot	0.6	0.0	0.0
Cervical spine	0.6	0.3	0.5
Thoracic spine**	16.2	1.7	8.8
Lumbar spine	18.2	19.3	22.0
Sacral spine	12.3	7.9	10.4
Caudal spine	5.8	3.1	2.2
Maxilla	4.5	1.1	1.1
Mandible	3.2	1.7	1.1
Skull - other	6.5	0.3	1.6
Pelvis	3.9	4.8	14.3
Scapula	1.3	0.0	1.1
Rib	5.8	0.9	5.5
Sternum	1.9	0.3	1.1
Extremities	19.4	58.8	30.2
Spine	53.2	32.4	44.0
Skull	14.3	3.1	3.9
Miscellaneous	12.9	6.0	22.0

*Undoubtedly many tumors were missed because they were too small to be detected radiographically and they occurred in areas not routinely taken for histopathologic study. This defect applies in larger degree to the radium experiment because the quality of the radiograms in the strontium and calcium experiments was improved.

**The right tibia was routinely sectioned in the strontium and calcium experiments but not in the radium experiment. The reverse was true of the thoracic spine. A small proportion of the total number of tumors was found only in the histologic material.

Fig. 24. Cell dose rate

a smaller proportion of the Sr^{90} tumors than of the Ca^{45} and Ra^{226} tumors. These differences may be associated with the size of the bone and the corresponding differences in dose rate. In the relatively large femur, the local Sr^{90} dose is augmented by contributions from neighboring deposits, whereas in the tiny rib this cannot happen, and much of the energy is dissipated beyond the bone. The Ra^{226} and Ca^{45} doses, on the other hand, depend primarily upon the specific activity of the local deposit and not upon cross radiation from adjacent deposits. The lumbar spine is probably an example of the average situation occurring in a bone of intermediate size. The low incidence of tumors in the humerus is puzzling in view of the fact that the dose-rate picture in this bone appears to be very similar to that in the femur. The explanation is probably physiological rather than physical.

Figure 31 shows the accumulated cell dose in rads for the femur obtained by integrating the indicated curves in Figs. 17, 18 and 19. The frequency distributions of time to death with osteogenic sarcomas in the comparative toxicity experiments are given in Fig. 32. With the combined information from Figs. 31 and 32, the total accumulated dose received at the time an animal died with an osteogenic sarcoma can be estimated. The most accurate estimate of the accumulated dose necessary for tumor production under the conditions of this experiment is that for Sr^{90} because the dose effective in producing the tumors in the femur probably lies in the shaded region between the metaphyseal maximum and the shaft minimum curves no matter where the sensitive volume lies. It is possible, of course, that the early high dose rate just below the epiphyseal plate is also responsible for tumor induction. However, it has not been included in Fig. 31 because its contribution to the total accumulated dose after 100 days is small.

Fig. 25
Autoradiographs of
lumbar spine

Fig. 26. Autoradiographs of rib

Fig. 27. Autoradiographs of skull

Fig. 28. Autoradiographs of ilium

Fig. 29. Rib profile 3 days after injection

Fig. 30. Lumbar spine profile at 3 days

Fig. 31 Accumulated cell dose in rads for the femur.

Fig. 32 Frequency distributions of time to death with osteogenic sarcomas

The earliest tumor death among the animals that received $0.44 \mu\text{c/g}$ Sr^{90} occurred at about 250 days, at which time the femur had received between 4,600 and 12,000 rads. On the other hand, the earliest tumor death among the animals that had received $2.20 \mu\text{c/g}$ occurred at about 160 days, when the femur had received 15,000 to 40,000 rads. At the other extreme, $0.044 \mu\text{c/g}$ was associated with one tumor death at 320 days and several others between 560 and 700 days. The minimum and maximum total accumulated doses for the femurs of these animals ranged from 580 to 2,500 rads. Until we have some sound information on the duration of the induction period and the time required for the neoplastic change to be manifest, we do not know the proper limit to put on the time scale for integrating the absorbed dose.

It is interesting to speculate on the location of the radiation-sensitive volume for the production of osteogenic sarcoma. There are fewer Ca^{45} tumors than Sr^{90} tumors in the femur, despite the fact that within the bone volume the accumulated dose and dose rate for Ca^{45} are almost everywhere greater than the corresponding Sr^{90} values. This is probably not due to a sharply decreased effectiveness for Ca^{45} at high dose rates because the Ca^{45} and Sr^{90} dose-response curves are similar in shape. Therefore, if a sensitive volume common to Ca^{45} and Sr^{90} exists, it would seem to lie outside the bone volume and thus at or near bone surfaces. The hypothesis that the sensitive volume lies at or near bone surfaces can, therefore, provide an explanation for the Ca^{45} and Sr^{90} results. The same hypothesis also helps to explain the factor of almost 10 between the carcinogenicity of injected Pu^{239} and Ra^{226} (12) since the former is deposited primarily on endosteal and periosteal surfaces rather than in the calcified matrix.

Fig. 33

Calculated dose profile for a thin plane source in bone (average over 10μ)

If bone surfaces are the sensitive regions for the induction of osteogenic sarcomas, the short-range particles from Ca^{45} and Ra^{226} should be more efficient than the long range particles from Sr^{90} in the small bones that have a relatively large surface-to-volume ratio. Accurate surface dose measurements after a single injection of Ca^{45} or Ra^{226} , however, are exceedingly difficult because the intense depositions occur on growing bone surfaces, and this growth rapidly separates the plane of maximum dose rate from the sensitive endosteal and periosteal cells. The complexity of accurate surface dose calculations for Ca^{45} and Ra^{226} can be appreciated by reference to Fig. 33, where the theoretical distributions of dose arising from a perpendicular plane source of radioactivity

within bone having a density of 2 g/cm^3 have been plotted. The radium curve was calculated by direct integration, and the Ca^{45} and $\text{Sr}^{90}\text{-Y}^{90}$ curves were calculated from Loevinger's point source function.⁽¹³⁾ The curves for Sr^{90} and Y^{90} were computed separately and then combined. Since at the exact position of the radioactive surface the dose is infinite, the average doses over a ten-micron interval have been plotted throughout. The dosage distribution at a growing surface is not quite this simple, however, because new bone laid down adjacent to this radioactive plane will also contain activity. This tends to skew the curves slightly in the direction of growth. However, the main point is that as growth proceeds the sensitive layer of cells moves farther and farther from the original site of deposition, and the proportion of the total available dose this sensitive layer will receive will depend upon the range of the emitted particles. Whereas at the source itself identical integral doses of Ra^{226} , Ca^{45} , and Sr^{90} will deliver dose rates in that decreasing order, 25μ from the source the order will be $\text{Ca} > \text{Ra} > \text{Sr}$, at 35μ it will be $\text{Ca} > \text{Sr} > \text{Ra}$, and at 100μ it will be $\text{Sr} > \text{Ca} > \text{Ra}$. In view of the fact that surface growth proceeds at the order of several microns per day, perhaps these shifting ratios will explain some of the differences in tumor location observed in the comparative toxicity experiments. However, comparison of the calculated with the observed widths of the dose distributions for a growing surface (Fig. 33 as compared with Fig. 22, for instance) shows that much better autoradiographic resolution would be required for accurate surface dose measurements for Ca^{45} and Ra^{226} .

Summary

The microscopic distribution of the dose in the mouse skeleton arising from deposited Ca^{45} , Sr^{90} , and Ra^{226} has been measured autoradiographically, and these measurements are being correlated with the data on tumor incidence and location that were obtained previously.

Because the range of the particles emitted by $\text{Sr}^{90}\text{-Y}^{90}$ is so much greater than that of the particles coming from Ca^{45} and Ra^{226} , the differences between maximum and minimum dose rates are much less for the first than for the last two nuclides. In addition, any local area of bone containing Sr^{90} receives dose contributions from adjacent areas. As a consequence, the smaller the bone, the lower its maximum dose rate from Sr^{90} , but not from Ca^{45} or Ra^{226} .

The Sr^{90} beta-particle doses accumulated by the time of death with an osteogenic sarcoma are the order of 10,000 rads for maximum incidence and the order of 1,000 rads for an incidence of a few percent. These are merely order of magnitude estimates, however. Further refinement must await a better understanding of the temporal distribution of tumor appearance and the effect of dose rate.

Comparison of the Sr^{90} with the Ca^{45} data suggests that the radiation-sensitive region for production of osteogenic sarcoma lies at or near microscopic bone surfaces rather than within the mineral volume. If this hypothesis is correct, then hot spots following single injections of Ca^{45} or Ra^{226} will not coincide with the sensitive region for very long after injection, and the effective dose rate will be more nearly that of the diffuse component.

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SOME INDIVIDUAL ALPHA-ACTIVITY COUNTS IN MIXTURES

J. Sadauskis

Thorium in Tooth Sample

A count of thorium activities isolated from a tooth from a Thorotrast patient has been reported.⁽¹⁾ Since a 10-channel analyzer was used, which limited the total number of counts taken, the same sample containing the isolated thorium was remeasured with a 100-channel analyzer. Table 13 shows a comparison of runs with both analyzers and Fig. 34 shows a plot of the data from the 100-channel analyzer.

Table 13

Comparison of results from 10-channel and
100-channel analyzers with tooth sample

	10-Channel analyzer (Run started Oct. 15, 1958) dpm	100-Channel analyzer (Run started Aug. 25, 1959) dpm
Th ²³²	0.6 ± 0.2	0.58 ± 0.05
Th ²²⁸	2.0 ± 0.4	1.64 ± 0.10
	1.5 ± 0.4 (calc. for Aug. 25, 1959)	
Th ²³⁰	not detected	small amount present

Th²³⁰ Impurity in Commercial Thorium

Two thorium solutions, used as standards for Th²²⁸, were found to contain Th²³⁰ as an impurity. The ratio of Th²³⁰ to Th²³² activities in the older solution, the thorium of which was purified between 1906 and 1917, was found to be 0.21 ± 0.01, while for the newer solution with thorium purified in 1946 it was 0.12 ± 0.01.

Percent of Ra²²⁶ Decay to Excited State

The percent of Ra²²⁶ atoms that decay to the excited state was determined. The source used was Ra²²⁶ adsorbed to fused quartz, and two mil Lektromesh was used for collimation to avoid trouble from conversion electrons.⁽²⁾ The resolution of the excited state peak is seen in Fig. 35.

Fig. 34

Thorium isotopes present in tooth sample. For each channel, instrument background was subtracted from the total channel count. Some channels have a negative count because of statistical fluctuation.

Fig. 35

Alpha particle spectrum of Ra²²⁶. Source used was Ra²²⁶ adsorbed to fused quartz with 2-mil Lektromesh for collimation.

The excited state count and total count were obtained by a method already described.⁽³⁾ Results of four runs are given in Table 14, and the average of the four runs, each weighted according to the total number of counts taken, is 5.7% which is the same value as that obtained by Asaro and Perlman.⁽⁴⁾

Table 14

Runs with Ra^{226} for determination of extent of decay to the excited state

Run	Time for run	Excited state count	Total count	Percent decay to excited state
1	1486 min	590	10,200	5.8
2	1487 min	590	10,400	5.7
3	1486 min	620	10,500	5.9
4	6832 min	2700	47,400	5.7
		weighted average		5.7

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GAMMA-RAY STUDIES ON RADIOACTIVE FALLOUT

P. F. Gustafson,[†] S. S. Brar[†] and M. A. Kerrigan[†]Part I. Radioactivity Measurements and Dose Calculations of
Soil Collected at Argonne

The gross fission product radioactivity in soils collected from August through November, 1959, has shown a continuing decrease from the values observed at the time of the last report.⁽¹⁾ Sampling, as in the past, was done in Argonne Park and west of D-202, two samples from each location taken to a depth of 6 inches.

A rather serious source of variation in the determination of the activity per soil sample was introduced during the period under consideration. This has been the effect of unsupervised cutting of grass in the areas where collections were made, with the uncertainty as to how much cut grass should be included with each sample. This is most important when it is realized that a large fraction of the fission activity, particularly the short-lived members, is adsorbed or otherwise associated with the vegetative cover. This uncertainty has resulted in rather large fluctuations in activity values during August and September as illustrated in Table 15, where the activities in mc/mi² for the six gamma-emitting fission products under investigation are presented.

Table 15

Gamma-ray emitting fallout radioactivity on the ground at
Argonne National Laboratory in mc/mi²

Isotope	1959				
	July	August	September	October	November
Zr ⁹⁵ -Nb ⁹⁵ *	765	592	444	366	264
Cs ¹³⁷	152	159	172	201	186
Ru ¹⁰⁶	1202	855	1003	969	996
Ru ¹⁰³	88	53	30	-	-
Ce ¹⁴¹	113	74	48	-	-
Ce ¹⁴⁴	1812	1706	1774	1924	1846
Totals**	4879	4031	3915	3826	3556

*Assuming Zr⁹⁵-Nb⁹⁵ to be in equilibrium

**Totals include activity due to both Zr⁹⁵ and Nb⁹⁵.

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During the latter half of 1959, Ce^{141} and Ru^{103} activity decreased below the level at which meaningful assay could be made. The Zr^{95} is decreasing at a rate which closely parallels the radioactive decay of this radionuclide, thus indicating that scant amounts of fresh activity are coming down. The Cs^{137} activity, still on the increase in soils, exhibits a much slower rate of accumulation than occurred during the spring months. This activity reached a value of 186 mc/mi^2 in November, representing an increase of some 20 percent over the July concentration. The corresponding Sr^{90} concentration in November, using the $\text{Cs}^{137}/\text{Sr}^{90}$ activity ratio of 1.6, was 116 mc/mi^2 for this area.

The dose rates accompanying such fission product deposition, computed for a point one meter above an infinite plane, are shown in Table 16. In May, 1959 (the time of maximum dose rate) the dose rate accruing from all six fission products was $10.06 \text{ } \mu\text{r/hr.}^{(1)}$ By November the dose rate had fallen to a value of $3.96 \text{ } \mu\text{r/hr.}$, a decrease to slightly under 40 percent of the peak value. The dose from $\text{Zr}^{95}\text{-Nb}^{95}$ continues to be the largest contributor to the total dose, but if it diminishes at the present rate, the dose from it will soon be surpassed by that from Ru^{106} .

Table 16

Dose rate due to fallout at Argonne National Laboratory in $\mu\text{r/hour}$

Isotope	1959				
	July	August	September	October	November
$\text{Zr}^{95}\text{-Nb}^{95}$	5.71	4.47	3.35	2.76	2.00
Cs^{137}	0.45	0.48	0.51	0.59	0.55
Ru^{106}	1.22	0.87	1.01	0.99	1.01
Ru^{103}	0.21	0.13	0.07	-	-
Ce^{141}	0.03	0.02	0.01	-	-
Ce^{144}	0.39	0.37	0.38	0.42	0.40
Totals	8.01	6.34	5.33	4.76	3.96
Natural Background = $13.23 \text{ } \mu\text{r/hour}$					

Part II. Fission Product Radioactivity in Air Filters

Weekly air filters collected by the Industrial Hygiene and Safety Division, provided through the courtesy of Dr. J. Sedlet, have been analyzed by gamma-ray spectrometry for gamma-emitting fission products. The period for which analyses were made extends from January 1958 through December 1959. Air filter samples taken at the Argonne site and from the surrounding territory were included in this study.

The weekly filters were grouped by months; a known portion of each batch was then ashed to secure a sample of smaller mass, thus making for improved counting geometry. Ashing, done in a muffle furnace at 325 to 400°C in covered vessels, resulted in no detectible loss of activity. Since the filters have an asbestos base, a fairly sizeable residue remained (20 percent of initial weight). This residual material was then placed in plastic Petri dishes and counted on the BIM 5" x 4" NaI crystal in the BIM steel room.

Considerable time elapsed between the time of collection of the filters and the time of counting, and still more, of course, since the last nuclear detonation. Thus the short-lived isotopes, including Ru^{103} and Ce^{141} , had for all intents and purposes disappeared. Analyses, using appropriate standards, and hand calculation of data using a method of successive approximations, were made for Cs^{137} , Zr^{95} - Nb^{95} , Ce^{144} , Ru^{106} , and Sb^{125} , all five of which are fission products. In addition, analyses for W^{185} and Rh^{102} were also carried out by gamma-ray spectrometry and in the subsequent calculations; of these two, the former radioisotope was produced during the U.S. "Hardtack Series" (April-August, 1958) conducted at the Pacific Proving Ground. The Rh^{102} was produced in the course of high-altitude missile detonations carried out in early August 1958 over Johnston Island in the Pacific. These activation, or "tag" radionuclides may thus serve to identify both the source and time of production of certain portions of the fallout.

These data overlap those of Dr. C. E. Miller⁽²⁾ as regards the Cs^{137} concentration in air as January-March 1958 filters were included in both series of measurements. The activity pertaining to each radionuclide examined is expressed in counts per 100 minutes per air filter. Each filter was run for approximately one week at a flow rate of 25 to 35 cubic meters of air per hour. Thus the volume of air passing through such a filter in one week is on the order of 5000 m^3 . Each monthly group of filters contained 20 to 30 weekly samples, but it was deemed wise to relate activity in terms of that present per filter. The activity has not been expressed in curies due to the fact that the standardization of all isotopes involved (namely Sb^{125} and Rh^{102}) has not been completed at time of writing.

Figure 36 shows the total gamma activity per month per air filter and represents the arithmetic sum of the activities due to each isotope considered. During the two-year interval, the gamma activity concentration in air varied by roughly a factor of 100. Distinct maxima appear during the spring months, due presumably to the break in the tropopause occurring at this season of the year. Figure 37 shows Cs^{137} , Ru^{106} , and Sb^{125} activity corrected for decay to the middle of month of collection as a function of time. The activity of both Ru^{106} and Sb^{125} relative to that of Cs^{137} is higher than expected on the basis of thermal fission yield. The explanation

Fig. 36 Gamma activity of air filters.

Fig. 37 Cs^{137} , Ru^{106} , and Sb^{125} activity corrected for decay to the middle of the month of collection as a function of time.

for this apparent discrepancy centers about the fact that thermal fission is not the primary source of radioactivity in a thermonuclear device detonated by a fission reaction. Granted that the initial impetus is provided by thermal fission, the resultant thermonuclear reaction, however, produces fast neutrons in copious abundance which in turn cause fast fissioning in the remaining trigger material. The net result is that radioactivity produced by fast fission outweighs that coming from the initial thermal fission phase.

The rapid decrease in total air activity during the spring and summer of 1959 is due in part to the radioactive decay of 65-day Zr^{95} . The Zr^{95} activity as a function of time is shown in Fig. 38, with Cs^{137} included for comparison. Figure 39 indicates temporal variation of W^{185} , Rh^{102} , and Cs^{137} activity. Since W^{185} was introduced during the Hardtack Series, it is not present (at least in amounts detectable by these means) prior to April 1958. Although a slight amount of activity due to this isotope was observed in the June air filters, July marked the entry of relatively large concentrations of W^{185} into surface air at Argonne. Actually the highest concentration occurred in July with secondary peaks present in October 1958 and January 1959. If the W^{185} concentrations in air are corrected for radioactive decay back to mid-June 1958 (the assumed mid-time of production of this radioisotope), one may then obtain an idea of the relative amount of Cs^{137} (and other fission products) present in the air at any given time which was produced concurrently with the W^{185} . On this basis it was concluded that 25 to 40 percent of the fallout observed during the spring months of 1959 arose from the Hardtack Series, with the remainder coming primarily for the Russian series in October 1958.

Fig. 38 Zr^{95} activity as a function of time. Cs^{137} is included as comparison.

Fig. 39 Temporal variation of W^{185} , Rh^{102} , and Cs^{137} activity.

The presence of Rh^{102} in surface air was first noted in September 1959 (see Fig. 39) and rose rather sharply throughout the remainder of the year. Careful examination of air filters collected prior to September failed to show any positive indication of this radioisotope. It will be noted that all the radioelements under investigation have shown a tendency to remain relatively constant in air collected from October through December 1959, or that they have decreased at a rate much slower than that prevailing during the spring and summer months.

Part III. Discussion of Soil and Air Fallout Data

The collection and analysis of monthly soil samples has made it possible to discern more clearly the rates at which fallout is accumulating. The air-filter data provide a means whereby the activity intermediate between stratospheric retention and ground deposition may be measured. Rainfall constitutes a third readily accessible medium of interest and will be spectrometrically examined in the very near future. A fourth, and perhaps the most crucial member of this progression, to be examined would be the stratospheric reservoir itself. Such a survey, however, is not feasible within the framework of the limited survey presently being conducted.

By using the fallout rates deduced from the soil data and the apportionment between U.S.S.R. and U.S. tests provided by the air-filter investigation, it is possible to compute the amount of any isotope in the stratospheric reservoir at any given time if certain assumptions are made (vide infra). Consider Cs^{137} for example: the fallout rate from soil data for this isotope in the period from March through July 1959 was $6 \text{ mc/mi}^2/\text{mo}$; on the basis of a 60-40 percent proportionation, we compute $4 \text{ mc/mi}^2/\text{mo}$ as the rate from the Russian tests and the other $2 \text{ mc/mi}^2/\text{mo}$ from Pacific tests. In November 1958, with 6-month and 4-year mean storage times for Russian and Pacific debris respectively, the stratospheric reservoir at 40° North latitude contained 117 mc/mi^2 . The region 35° to 45° North comprises an area of $1.5 \times 10^7 \text{ mi}^2$, thus the total stratospheric reservoir in this belt contained $1.5 \times 10^7 \text{ mi}^2 \times 117 \text{ mc/mi}^2 = 1.75 \text{ megacuries of } \text{Cs}^{137}$. Since approximately 25 percent of the total world-wide deposition of Sr^{90} ,⁽³⁾ and presumably therefore also Cs^{137} , occurs in this latitude band, the entire reservoir contained 4 times this amount, or 7 megacuries of Cs^{137} . This figure is in fair agreement with the data reported by Libby,⁽⁴⁾ which indicated that, on the basis of extrapolation of air and rain sampling in November 1958 there was on the order of 4 megacuries of Sr^{90} in the entire stratospheric reservoir, or 6.4 megacuries of Cs^{137} through the use of the Cs/Sr activity ratio of 1.6.

In the period March to July 1959, the ratios of $\text{Ce}^{141}/\text{Zr}^{95}$ and $\text{Ru}^{103}/\text{Zr}^{95}$ activities in new fallout indicated that they were produced during October 1958. On the other hand, the relative amounts of longer-lived Ce^{144} , Ru^{106} , and Cs^{137} coming down during the same time interval showed that a significant fraction (25 to 40 percent) of these radionuclides arose from tests conducted prior to the fall of 1958.

The fallout rate and proportionation factors make it possible to also predict future fallout deposition, and attendant dose rates. Figure 40 illustrates the gamma dose rate in $\mu\text{rad/hr}$ at a height of one meter above an infinite plane for $\text{Ce}^{141,144}$, $\text{Ru}^{103,106}$, $\text{Zr}^{95}\text{-Nb}^{95}$, and Cs^{137} contrasted with the dose due to Cs^{137} alone. The dose rate was expressed in $\mu\text{rad/hr}$ in order to compare more easily with dose due to internal emitters. The empirical data extend from May 1957 (the time at which soil sampling and dose computation were initiated at the Argonne site) up through the end of 1959. Interpolated values derived from the mathematical model were used for the interval October 1958 through February 1959. Since there is good agreement between experimental and calculated dose rate values, extrapolation into the future has been made on the basis of no further testing of nuclear weapons. As expected, Cs^{137} becomes the dominant factor as time goes by, and after about 1965 the external dose is due almost exclusively to this radioelement.

Fig. 40 Gamma dose rate one meter above an infinite plane.

Moving on to the matter of stratospheric storage time, one is immediately struck by the rapid decrease in the concentration of activity in air during the latter half of 1959. In fact, from June through September the concentration of the longer-lived fission products decreases by a factor of two every month. This half-time is essentially the same as that assumed for the half-time for tropospheric clearance. There are at least two plausible explanations for this phenomenon. First, the stratospheric storage times generally used, namely 6 to 12 months for MT explosions carried out in the far north, and 3 to 4 years for similar detonations occurring near the equator, are incorrect, and are in fact much shorter. Secondly, the activity observed at ground level may not be of recent stratospheric origin, but may have entered the troposphere at the time of the most recent break in the tropopause. This effect might be termed the "stratospheric faucet effect," being analogous to turning on a water faucet for a limited time and then shutting it off. Although this effect is most pronounced in the spring, there is an indication that a similar faucet action of lesser magnitude occurred in the fall of 1958. However, one may argue that this is merely tropospheric fallout from the Russian series and our own limited efforts in Nevada in the fall of 1958. There is also a rise, or at least a tendency to remain constant, during the fall of 1959. Here again, one may be inclined to assign this added activity to the missile shots, as it coincides with the appearance of the Rh^{102} tag. The critical period will be during the spring months in 1960 for seeing whether or not the faucet effect repeats with a markedly increased concentration of fission activity in air as a result.

In any event, it appears that fallout does not come down at a constant rate, nor at a rate which is a simple combination of two or more rates. This fact may lead to serious errors in predictions of future fallout levels arising from tests already concluded. A further point of interest is shown by the air-filter data, namely the appearance of Rh^{102} in surface air after a holdup time of 14 months. This indicates that there may well be quite different rates due to debris coming from very high altitudes. This latter point may have considerable bearing on the future testing and/or detection of nuclear detonations in the outer atmosphere.

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THE CESIUM-POTASSIUM TREND IN HUMANS AS A FUNCTION OF TIME

C. E. Miller

The total-body gamma-ray spectra of 13 unexposed employees of the Laboratory chosen to be typical of the civilian population of the Chicago area have been measured periodically for Cs^{137} since March 1955.⁽¹⁾

Their Cs^{137} body contents have been determined and are reported in units of $\mu\mu\text{c Cs}^{137}$ per gram of natural body potassium. The Cs/K ratios of eight subjects are presented in Table 17 for six dates during the past 18 months. It will be noted that the average value rose from 50 $\mu\mu\text{c Cs}^{137}/\text{g}$ of K in July 31, 1958 to a high of 64.4 $\mu\mu\text{c Cs}^{137}/\text{g}$ of K in June, 1959, and fell to 55 $\mu\mu\text{c Cs}^{137}/\text{g}$ K in December 1959. The data for the other five subjects are not included since they were not counted on one of the six dates. This omission does not alter the average ratio by more than 3 per cent.

Table 17

Cesium-137 Potassium-40 ratios in eight control subjects
over a span of 18 months

Subject	7/31/58	11/20/58	3/19/59	6/17/59	9/14/59	12/8/59
C.L.	57	48.5	59	68.5	60	56.2
I.S.	51.5	52	59	54.6	54.5	47.4
H.M.	71.0	68.5	75.2	83.2	68.2	69.5
C.M.	43	58	76	81	64.2	62
B.O.	47	47.8	54.5	57.2	45.5	42.2
B.P.	43.5	46.2	53.5	59	54.7	53.5
R.R.	39	39.5	47	46	45.6	45.2
O.S.	48.4	51	61.8	65.5	64	63.1
Average	50	51.4	60.7	64.4	57.1	55

Mr. Ingvar Andersson, Atomic Energy Company, Stockholm, Sweden, measured 19 females during May and 27 males during June of 1959, finding a Cs^{137}/K ratio of 57 $\mu\mu\text{c Cs}^{137}/\text{g}$ K for the females and 73.5 $\mu\mu\text{c Cs}^{137}/\text{g}$ K for the males.⁽²⁾ He also measured the author's content on June 1, 1959, and found a value in excellent agreement with the Argonne value. Thus, the results from these two laboratories can be compared directly.

Each of the above subjects was showered thoroughly before being counted and each wore special low-background coveralls during the measurements. A number of control subjects, measured while attired in their normal

street clothes, apparently contained from 5 to 10 per cent more Cs^{137} . Mr. Andersson likewise counted subjects while they were wearing first street clothes and then, special low-background clothing. He reported that subjects yielded a total counting rate 30 per cent higher when dressed in street clothing.

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MEASUREMENT OF NATURAL URANIUM IN THE LUNGS OF A HUMAN

C. E. Miller

A sample of natural uranium was measured in the lung-equivalent phantom to determine the feasibility of routinely monitoring employees for the presence of accumulated uranium in their lungs. The physical details of the lung-equivalent phantom, and the reliability of the data obtained therewith, have been published elsewhere.^(1,2)

An 8" x 4" NaI(Tl) crystal was placed 1 cm from the surface of the lung-equivalent phantom and the gamma-ray spectra obtained of background and of 2 g normal uranium in the phantom. A spectrum was also obtained with this crystal 1 cm from the surface of an uncontaminated subject's chest. The net spectra of the uncontaminated human, of background, and of 0.02 g of natural uranium are given in Fig. 41.

Fig. 41

The data necessary to determine the counting time needed to realize a desired accuracy, namely, the counting rates of background, of the typical human, and of 0.02 g uranium in two energy bands are given in Table 18. In calculating the standard deviations, the assumption was made that background and the unexposed human were counted for 50 minutes each. The standard deviations that would result if the subject under study were counted for 10 minutes, 50 minutes and 100 minutes were calculated from the data given in the third column. From these standard deviations and the data of the fourth column, the uncertainties of the measurements - expressed in terms of grams of natural uranium - are given in the last three columns.

A ten-minute measurement with one 8" x 4" NaI(Tl) crystal against the chest would give an uncertainty of 0.0015 g or 10 percent of maximum permissible level (MPL). Either the uncertainty could be reduced 30 percent, or the counting time could be reduced to five minutes if a second crystal were used simultaneously against the subject's back.

Table 18

Energy band	Background, c/m	Net unexposed human, c/m	Total, c/m	0.02 g U ₃ O ₈ , c/m	U ₃ O ₈ , g		
					Counting Period		
					10 min	50 min	100 min
73 kev to 127 kev	110.8	118.9	229.7	81	0.0015	0.0008	0.0007
155 kev to 195 kev	82.4	68.7	151.1	22.8	0.004	0.0022	0.0019

If the gamma-ray background of the steel room is absolutely reproducible day after day, then the subject's gross spectrum can be compared to his pre-employment gross spectrum. If the backgrounds do vary slightly, the background would have to be run each day and the subject's net spectrum compared to his pre-employment net spectrum.

The counting time necessary to determine the lung content within 10 percent of MPL is not so unreasonably long as to exclude the use of chest measurements on a semiroutine basis in feed material plants. Such measurements would be valuable, particularly in those instances where an employee has submitted a contaminated bioassay specimen. The largest expenditure of time will undoubtedly be involved in showering (decontaminating) the subject before the measurement.

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STATUS OF THE RADIUM DIAL PROJECT

C. E. Miller

The majority of the former radium dial painters that have been studied at Argonne National Laboratory have been employees of one of the "Radium Art Studios," which were owned by the Radium Chemical Company of Pittsburgh, Pa. This organization operated art studios in several towns in the Middle West. In order to protect the anonymity of the subjects but still permit a discussion of the status of this study, the towns where these studios were located will be designated by letters.

A "Radium Art Studio" was established in Town A in 1917 or 1918. Radium dials for watches and other instruments were painted in the A studios until late in 1921 at which time the studio moved to Town B. The studio moved from Town B to Town C in September, 1922 where it remained until 1935 at which time it was moved to New York City. The Radium Dial Company simultaneously operated another studio for about eight months during 1925 in a town which will be designated as Town D. Although the company may have owned other studios, these are the only ones pertinent to our discussion.

No employment records or lists of employees who painted radium dials have ever been available and furthermore, probably do not exist for this period of interest (1918-1930). In the past the names of former dial painters have been obtained from other former dial painters who were subjects for the ANL study. The memories of these persons were aided by group photographs taken at the studios or at social gatherings in the 1920's and which have recently come into our possession.

In June 1959, sets of city directories were finally located for three of the towns (B, C, and D) in which studios were located. Since the city directories gave the occupations of the persons listed, employment lists have been reconstructed for the period during which the canvasses for the directories were made for Towns B and C. Unfortunately, city directories were only compiled on even-numbered years; nevertheless, it is felt that the lists are very complete for a particular date during the even year. Evidently the canvassers obtained the employment lists directly from the studios since individuals are listed in the directory who worked at the studio but lived in an outlying town.

Town B

A total of 48 individuals were listed in the B City Directory of 1922 as being employees of the Radium Art Studio. Of these, 23 have been studied, 13 are deceased, 2 are waiting to be scheduled, 6 have not been cooperative, and 4 have not been contacted. The 23 studied subjects have supplied the

names of 6 additional individuals who painted for a short period of time but were not listed in the directory.

It is of interest to note that although the current body contents of all individuals thus far counted vary over a wide range, ($0.005 \mu\text{c}$ to $1.61 \mu\text{c}$), most of the subjects who worked at studio B have a body content of from 0.1 to $0.2 \mu\text{c}$.

Town C

The occupation of each person listed in the 1924, 1926, 1928, and 1930 City Directories for Town C was checked and lists of the employees of the Radium Art Studio in each of these years compiled. These names obtained from the city directories, as well as the names obtained by identification of individuals in group pictures that had been taken at the studio at known times, have been integrated into an alphabetical master list of 191 different names. Of these 191 individuals, 87 have been measured, 40 are deceased, 38 have been contacted but are unwilling to cooperate, 10 are waiting to be scheduled, 10 have been located but not contacted, and the whereabouts of 8 are unknown.

This compilation has become valuable as a guide in selecting additional patients. The dates of employment that the subjects gave when interviewed at ANL, as well as the years in which their names appeared in the directories have been included in the master list. The subject's body content of radium also has been recorded. It was observed that those employees who did not work before 1926 did not have an appreciable body content of radium. As a general rule, an individual who started working after July 1925 contains at the present time less than 0.05 if not less than 0.01 microcurie of radium.

Not all individuals who worked during the period from 1922 through 1924 contain appreciable quantities of radium. However, all of those employees of Studio C who do contain from $0.1 \mu\text{c}$ to $2.4 \mu\text{c}$ did work some time during the 1922 to 1925 period. Furthermore, every subject from this studio who contains over one microcurie, as well as the majority of those who are deceased, worked during the year 1924. Unfortunately, most of the uncooperative ones worked during 1924 so a major effort will be made to obtain their assistance since they may be expected to contain at least $0.1 \mu\text{c}$.

No list or picture has been found which can be used to identify those individuals who worked at Studio C between September 1922 (when the studio opened) until the first city directory canvass in April 1924. The five measured individuals who worked during this period contain from $0.1 \mu\text{c}$ to $0.6 \mu\text{c}$. An effort will be made to determine whether there are other employees of this period who have not been located.

Town D

A "Radium Art Studio" was located in Town D for about eight months during 1925. Since the directories were compiled on even years, the employees were not listed in either the 1924 or 1926 directories. These employees are being located through the cooperation of the ones that have been to the Laboratory. Most of the subjects who worked in Town D and who did not transfer to Town C, when Studio D closed down, have a current body content that is less than $0.01 \mu\text{c}$.

Town A

Recently the names of fourteen individuals who painted watches in Studio A have been obtained. Although information has been obtained relating to the home towns and relatives of some of these painters, no effort has been made to trace these subjects to date. Information that is available would indicate that individuals from this group would probably contain from $0.1 \mu\text{c}$ to $2 \mu\text{c}$.

Conclusion

When the radium body content of the subjects is correlated with the period of employment, it is found that, as a general rule, those individuals who worked after July 1925 contain less than 10^{-8} c while those employed before this date contain $0.1 \mu\text{c}$ or more. Consequently, only those women who painted watches before July 1925 are being traced and asked to cooperate.

There are about sixteen former dial painters from Studio B and about forty from Studio C who, if possible, will be persuaded to participate. It is the goal of the Argonne group to study these 56 subjects before July 1, 1960 and thus to complete this phase of the project.

RETENTION OF RADIUM IN HUMANS FROM 21 TO 29 YEARS AFTER INTRAVENOUS ADMINISTRATION

C. E. Miller

The most reliable data on Ra^{226} excretion at long intervals following I. V. excretion are those of Norris, Speckman and Gustafson.⁽¹⁾ They were obtained from mental patients injected in 1931 at the rate of $10 \mu\mu\text{c}$ per week, measured first 1 to 7 months after the last injection and 6, 233, and 258 months thereafter. Although the rates of excretion, as calculated by the retention curves of 1951-1953, were in agreement with actual measurements of fecal excretion, there have been some doubts cast on the validity of the "power law of retention" based on some shorter-term Sr^{90} studies on primates and accidentally exposed men.^(2,3)

Because of the importance of the issues involved and the unique opportunity of repeating the measurements (in vivo on these patients) six years later, nine of them were measured again for $\text{Ra}(\text{B} + \text{C})$ with the low-level crystal spectrometer.

Owing to difficulties involved in the management of these mental cases, their measurements may not yet be precise. They are of sufficient accuracy to be compared as shown in Table 19. In the last column the 1959 $\text{Ra}(\text{B} + \text{C})$ estimate is computed as percent of the average of the May 1951 and July 1953 Ra values.

Table 19

Patient	May 1951		July 1953		July 1959	
	$\text{RaB} + \text{RaC}$ μc	Radon μc	$\text{RaB} + \text{RaC}$ μc	Radon μc	$\text{RaB} + \text{RaC}$ μc	% Retained
A	0.57	1.03	0.6	1.15	0.49	84
J	0.32	0.66	0.24	0.66	0.22	79
K	0.22	0.70	0.22	0.70	0.36	(163)
N	0.57	1.15	0.5	1.20	0.37	69
O	0.39	0.55	0.34	0.68	0.23	62
S	0.19	0.35	0.2	0.42	0.322	(165)
T	0.27	0.71	0.32	0.77	0.29	100
U	0.19	0.39	0.22	0.36	0.17	81
Paul N	0.077	0.39	0.1	0.42	0.156	(175)
Total	2.8		2.74		2.61	94% (78%)

It is worthy of note that the average content has diminished to only 94% if all patients are included. Three of them - K, S and Paul N, reveal, however, abominably high values, well off the estimated combined error of 25%

in the 1951-1953 values and 10% or better in the 1959 estimates. If these patients are excluded - and there are good reasons to conclude that movements during measurement produced serious errors - then average decrease for the group is 78%, well within the experimental error of the value of 84% expected by the law

$$\text{Retention} = A t^{-b}$$

where $b \simeq 0.52$.

A final conclusion awaits exhaled radon measurements and remeasurement of the γ -ray emission from the whole body.

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STATUS REPORT OF STUDY OF LONG-TERM EFFECTS OF RADIUM IN HUMANS

A. J. Finkel[†], C. E. Miller and R. J. Hasterlik^{††}

During the past three years, approximately 200 persons suspected of having an appreciable body content of radium have been studied by means of the Argonne human gamma-ray spectrometer. Of these persons, seven received radium therapeutically, and the remainder were exposed to radium in the luminous dial industry or in radium chemical laboratories. Clinical studies have been completed for 175 persons and have included skeletal radiographic surveys and complete blood counts, as well as a recording of the occupational and medical history.

Systematic criteria for grading the severity of radiographic lesions have been developed, and the degree of radiographic change has been correlated with the current body-content of radium. Radiographic changes range from those that are barely detectable, such as slight coarsening of trabeculation, to those of advanced severity, such as aseptic necrosis, pathological fracture, and malignancy. The determination of a point at which this succession of changes becomes clinically significant represents an important problem.

The radiographic changes in the skeleton that can be specifically attributed to radium deposition can conveniently be put into categories such as advanced, moderate, mild, and minimal. The correlation of these changes with the body radium content is given in table 20.

Table 20
Radium in humans

Body content μc	Number measured	Number x-rayed	Radiographic changes in skeleton attributable to radium deposition					
			None	Minimal	Mild	Moderate	Advanced	(Malignant)
<0.001	22	17	14	3	0	0	0	
0.001 - 0.0031	6	5	5	0	0	0	0	
0.0032 - 0.0099	26	19	19	0	0	0	0	
0.01 - 0.031	59	51	48	2	1	0	0	
0.032 - 0.099	28	25	21	3	0	1**	0	
0.10 - 0.316	24	24	17	2	2	2*	1**	
0.32 - 0.99	16	15	1	4	5	4	1	(1)
1.0 - 3.16	19	18	0	1	4	3	10	(4)
3.2 - 4.2	2	1	0	0	0	0	2***	(0)
	202	175						

*MsTh present in each case

**Final diagnosis deferred; further studies pending

***Based on films taken elsewhere
in one case

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The body content of radium-226 has been divided into categories each of which spans a square root of an order of magnitude. In general, mild changes, which may not be clinically significant, are not seen until a level greater than 0.01 microcuries is reached. Unequivocal changes of a moderate degree, and which may be of some clinical significance, are not seen until the body burden is at least 0.30 microcuries. In the three cases where moderate changes have been seen in lower body radium levels, mesothorium has also been detected in two cases, and the study of the third case is incomplete. On the other hand, when the body content is in excess of 1.0 microcuries, the skeletal x-rays uniformly show some degree of change that can be specifically associated with radium although in 28% of these cases ($\frac{5}{18}$), the changes are not more severe than mild.

The number of cases collected is beginning to approach a magnitude that will make possible quantitative evaluation of the long-term effects of radium deposition in humans. It is anticipated that another 50 cases can be added within the next six months.

RADIOACTIVITY OF COMMERCIAL PHOTOMULTIPLIER TUBES

H. A. May

Introduction

The limitations in low-level scintillation counting imposed by the background radioactivity of the photomultiplier tubes seem to have been first recognized by Audric and Long.⁽¹⁾ Hester⁽²⁾ reported differences in backgrounds, as correlated with potassium content of the Kodial (lead glass) and Pyrex used in certain EMI tubes; his limited crystal size and lack of pulse height analysis apparently prevented detection of radium as well. Spectra of typical DuMont tubes were given by Miller *et al.*,⁽³⁾ permitting a more quantitative knowledge of the background contribution in any specified energy region. More extensive measurements of potassium and radium content of some American glasses and source materials were reported by members of P. R. Bell's group at Oak Ridge,⁽⁴⁾ and by Miller and Steingraber in a previous report of this series.⁽⁵⁾

We have shown⁽⁶⁾ that counting of activity distributed throughout the human body, or other extended sources, is facilitated by the use of large crystals, 8-to 9-inch diameters being typical of present production capabilities. In order to obtain the best resolution of which these crystals are inherently capable, an array of several 3-inch diameter phototubes is preferred over the use of a single large tube. As we shall show, low-background tubes now available have most of the remaining activity in their stem. Hence the contribution goes up linearly with the number of tubes used: therefore, one must consider the relative importance of high resolution and low background in practical applications.

A useful figure of merit for a photomultiplier is the background contribution per unit photocathode area. Since neither the photocathode uniformity nor the collection efficiency is the same for 3-inch and 5-inch tubes, resolution is not directly proportional to total cathode area. In general, in an array of smaller tubes, the number of internal reflections experienced by the scintillation quanta before collection is reduced, the total light collected is increased, and, hence, the resolution improves more than would be expected from consideration of photocathode area alone. The proposed figure of merit must be considered only as a general indicator of the value of a particular tube type.

Present Measurements

Measurement of naturally-occurring radioelements is essential to the success of an important portion of the work carried on by this Division. The presence of these same elements in the detector is undesirable, and their elimination is more advantageous than statistical analysis of the overall background reduction might indicate, hence, our continued efforts to measure the activity of commercially-available tubes and to foster development of low-level types. Many measurements have been made during the past year, utilizing a $7 \times 3\frac{1}{2}$ -inch crystal of exceptionally low potassium content; these are summarized here.

These data may be expressed in two ways, each having certain merits and limitations. In the first, the experimental results, consisting of the spectral distribution and net increase in total counts over a given energy band for a given geometry and crystal size, may be stated directly. By these means, the relative activity of various similar samples is apparent; the data may be used to estimate directly the performance of crystals of other sizes or different geometry; the precision achieved depends upon factors which usually are not known exactly.

Alternatively, one may state the data in terms of specific activity of each sample. Since the geometry of an intact phototube is difficult to reproduce with a source of known activity, the conversion factors employed are of unknown accuracy. If destructive testing is permissible, samples of the phototube or its component parts may be crushed or compressed into a more reproducible geometry and counted. The data thus expressed will have a reassuring appearance of accuracy, without conveying much information about what it really means in actual counting.

All data include the range from 35 kev to 1.575 Mev. Tubes were counted with their faces resting upon a $\frac{1}{4}$ -inch quartz plate in contact with the 0.010-inch stainless steel covering the crystal face. The purpose of this plate was to simulate the geometry which would exist if the tube being counted were actually coupled to the crystal. In some cases, a $\frac{3}{8}$ -inch Lucite plate was used instead. Any differences so introduced were less than the statistical uncertainties. The Bakelite tube bases were always removed.

Every effort was made to secure a low and constant background. Measurements were made inside an 8-inch walled steel cave with additional shielding in the form of 2-inch bismuth bricks arranged about the crystal or $\frac{1}{2}$ inch of mercury in the crystal housing - the exact details depending upon other experiments being carried on concurrently. Total backgrounds under these conditions average about 400 ± 20 c/m, the fluctuations from day to day being, in large part, due to varying radon levels in the laboratory air. In some instances, aged air was introduced into the cave at the rate of

a few cfm, noticeably stabilizing the background. Sample and background counts were made alternately for at least 100 minutes each. The standard deviation in percent, due to counting statistics of a single determination, as a function of net sample counting rate is given by Fig. 42. When possible, successive counts were made of different tubes of the same type; the resultant improvement in statistics may be readily calculated. Since at most only six tubes of one type were tested, no estimate has been made of their distribution about the mean.

Fig. 42. % Standard deviation = $\frac{100 \sqrt{t(S + 2B)}}{tS}$
 $t = 100 \text{ min}$

Table 21 summarizes most of the determinations made on intact phototubes. The first part gives results of "tube face down" geometry, the second part with the stem closest to the crystal. In the latter, the tube faces were partially shielded from the crystal by a $\frac{1}{4}$ -inch Pb plate with a hole in the center through which the stem projected. Figures 43 and 44 compare spectra of the low background tubes. It is apparent that the major portion of activity remaining in the quartz or Vycor faceplate types, is in the stem. In order to separate completely the stem vs faceplate contributions and reduce the data to amount of activity per gram of glass, the tubes would have to be broken apart, crushed, and counted. This we have been unwilling to do. However, similar conclusions for standard DuMont types were reported by Lazar⁽⁷⁾ and are borne out by limited measurements here. All samples were placed directly on the crystal face, so that counting geometry was approximately 2π . Results are tabulated in Table 22. The ratio of stem to faceplate activities is in general about the same as those calculated from Table 21.

Table 21

Phototubes

Mfg	Type	Nomi- nal size	No. of tubes counted		Net counts/min (per tube)	Activity, * curies		Mass, gms (glass only per tube)	c/m/gm of glass	c/m/cm ² of photo- cathode
			Individ- ually	Simultane- ously		K ⁴⁰	Ra			
<u>Part I - Face down</u>										
DuMont	6363	3"		3 (face- plates only)	18					
DuMont	6363			3 (entire tubes)	39					
			6		40.6			112	.35	1.13
DuMont	K1785 (Vycor face)	3"		3	25					
				3	27					
			1 (face- plate only)		13.2					
			1		27.7					
			6		24.4					
			mean		26.0 ± 1.5	1.2 x 10 ⁻⁹	5 x 10 ⁻¹¹	175	.15	.67
EMI	9531B (Pyrex face)	3½"	2 (avg of 8 runs)		59			180	.33	1.18
EMI	9531B (Quartz face)	3½"		3	16					
			4		25 ± 5			205	.12	0.50
DuMont	6364	5"	5		110 ± 5			493	.215	1.16
			1		205					
EMI	9530B (Pyrex)	5"	3		184					1.9
EMI	9530B (Quartz)	5"	2 (avg of 6 runs)		18.3 ± 3.2			400	.046	.19
<u>Part II - Stem down</u>										
DuMont	K1785	3"	3		60	4.2 x 10 ⁻¹⁰	3.8 x 10 ⁻¹¹			
EMI	9531B (Pyrex)	3½"	2		85					
EMI	9531B (Quartz)	3½"	4		59	1.0 x 10 ⁻⁹	1.0 x 10 ⁻¹¹			
				3	70	1.25 x 10 ⁻⁹	1.6 x 10 ⁻¹¹			
EMI	9530B (Pyrex)	5"	2		75	9.4 x 10 ⁻¹⁰	1.9 x 10 ⁻¹¹			

*Assuming all activity located in stem.

Fig. 43. Activity in EMI Type 9531 B quartz face tubes

Fig. 44. Activity in DuMont K 1785 3-in. PM tubes

Conclusion

The values in the last column of Table 21 are seen to fall roughly into three groups, with the rather substantial superiority of the 5-inch EMI quartz tube quite evident. Given a large crystal (8 x 4 in.), this tube would yield a resolution of 10 or 12% (for Cs^{137}). Three or four smaller tubes would certainly improve this figure; however, the activity of these must be less by a factor of four to six times than that of the currently-available ones if the total background contribution is to remain the same as that of a single 5-in. EMI tube.

It is apparent that glasses should be developed having reduced activity and still possessing the desired thermal expansion and electrical

properties. A more radical approach may be necessary, involving an all-metal body fusion-sealed or hard soldered to a transparent window of synthetic sapphire or quartz, with high purity alumina insulating inserts at the stem.

Table 22
PM tube components

Part	Mass (gms)	Net counts/min	c/m/gm	Activity curies/gm
1. Glass, 6363 faceplate	46	17.4	0.38	1.25×10^{-10} c/gm K^{40}
2. Glass, 6363 sides	53	30	0.56	
3. Glass, 6363 press & stem	14	100.3	7	
Glass, 6363 press & stem	40	239	6	
4. Glass, 6364 faceplate	200	32	0.16	Radium
5. Glass, 6364 sides	140	44	0.30	2.3×10^{-13} c/gm Ra
6.* Glass, K 1785 press & metal leads	18 35	30 51	1.66 1.45	1.4×10^{-12} c/gm Ra
7.* Glass, tubulation (part of unused stem, item #6)	28.3	96	3.4	2.9×10^{-12} c/gm Ra
8. Multiplier structures (3)	142	26.5 ± 2	0.19	
9.* Alumina insulators (4) (as used in item #8)	4.45	47 ± 2	1.1	1.2×10^{-11} c/gm K^{40} 8×10^{-13} c/gm Ra
10.* Metal parts from 2 multiplier structures	52	4 ± 3	.08	

*Furnished by Mr. William Hyde of DuMont.

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ACTIVITY INDUCED IN MEAT BY HIGH-ENERGY ELECTRON BOMBARDMENT

H. A. May

The Food and Container Institute of the Quartermaster Corps, U. S. Army, sponsors a large amount of research on radiation sterilization and preservation of foodstuffs. One of the most economical methods is irradiation by high-energy electrons. Some induced radioactivity is produced - largely as the result of photonuclear (principally γ, n) reactions; measurements of thick-target cross sections have been reported for 24-Mev electrons incident on aqueous solutions enriched in those elements most prevalent in foodstuffs. From such cross sections, which are of the order of millibarns, quantitative estimates of the most important long-lived activities expected in food were made.⁽¹⁾ One of the participants in this program, the Stanford Research Institute, requested some measurement of several samples of ground beef, as a check on their calculations.

The samples were packaged and irradiated in commercial Number 10 tin cans, 15.5 cm in diameter by 17 cm long, each sample weighing approximately 3 kg. The electron beam impinged on the ends of the container; an exposure to 24-Mev electrons of 75 microampere-minutes, on each end, resulted in a dose, averaged throughout the volume, of the order of 10 megarads.* Samples were counted 43 days after irradiation. Because of the relatively large amount of activity in the metal can itself, irradiated samples were transferred to clean, new containers. Control samples were also evaluated for initial activity. The low-background 7" x 3½" NaI(Tl) crystal was employed for all runs with the sample resting directly on a ¾" Lucite beta shield, approximately ½" from the crystal face. Counting times of 100 minutes were usually used.

Figure 45 is the net spectrum of a control sample, revealing gamma rays from K^{40} , Cs^{137} , and Zn^{65} , in that order of relative activity. The net increase due to an irradiation of 12.1 megarads is shown in Fig. 46; Na^{22} contributes most of the observed counts. Subtraction of a normalized Na^{22} spectrum reveals more clearly the peaks at 880 kev, thought to be due to 33-day Rb^{84} , and those of Zn^{65} at 1.11 Mev. In addition, the figure suggests other lines in the 100-to 400-kev region, but these are poorly resolved and identification has not been attempted.

* Samples were irradiated at the John Jay Hopkins Laboratory for Pure and Applied Science, General Dynamics Corporation, San Diego, and supplied by Richard Glass and Herbert Smith of Stanford Research Institute. The dose figures are theirs.

Calibration samples were prepared by incorporating known amounts of the identified isotopes, uniformly distributed, into 10 cans of agar media or meat. Since a standardized Zn^{65} source in solution was not readily available, a point source suspended at the geometrical center of a can filled with scattering media, was employed. A correction factor for the different geometry and collection efficiency based on the Monte Carlo methods of Miller and Snow⁽²⁾ was applied. Table 23 summarizes the pertinent data on the samples counted.

Table 23

Sample	Type	Wt, g	Irradiation level, megarads	K^{40} , g natural K	Cs^{137} , μc	Cs/K $\mu\mu\text{c}/\text{g K}$	Zn^{65} , μc
C_1^*	Control	2708	0	7.85 $= 6.4 \times 10^{-9}\text{c}$	0.94×10^{-3}	120	1×10^{-5}
C_2^*	Control	2695	0	8.07 $= 6.6 \times 10^{-9}\text{c}$	1.0×10^{-3}	124	1×10^{-5}
				Na^{22}		Zn^{65}	
I_1^{**}	Irradiated	2716	12.1	$7.9 \times 10^{-4} \mu\text{c}$ $= 2.4 \times 10^{-8} \mu\text{c}/\text{g}/\text{megarad}$		$\sim 2.0 \times 10^{-5} \mu\text{c}$	
I_2^{***}	Irradiated	2680	8.85	$5.6 \times 10^{-4} \mu\text{c}$ $= 2.4 \times 10^{-8} \mu\text{c}/\text{g}/\text{megarad}$		$\sim 1.5 \times 10^{-5} \mu\text{c}$	

* C_1 & C_2 : Total average activity (including K^{40}) = 7.3×10^{-9} curie

** I_1 : Total increase in activity = 8.1×10^{-10} curie

*** I_2 : Total increase in activity = 5.7×10^{-10} curie

The radioactivity induced in the metal can was also investigated. Figure 47 is a spectrum of the can in which sample I_2 was irradiated, 45 days after irradiation. Only one line is readily identified, that of Mn^{54} , estimated to be equivalent to 1×10^{-8} curies. The prominent peak at 160 kev may be due to 14-day $\text{Sn}^{117\text{m}}$; positive identification, requiring additional counts or chemical extraction procedures, has not been attempted. Total activity at this time is of the order of 2×10^{-8} curies. These container values are only approximate since they are based on comparison with the standardized sources used to measure the radioactivity of the contents. They do indicate, however, the correct order of magnitude and because of their low value, suggest that the activity induced by electron irradiation in the can plays no part in the preservation of the food or its suitability for human consumption.

Fig. 45. Net spectrum of unirradiated meat

Fig. 46. Net activity from irradiated meat

Fig. 47. Net spectrum of irradiated can

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THE ARGONNE METEOROLOGICAL MODEL TOWING TANK

Paul Frenzen

Introduction

With the completion of the trial runs including all three conditions of fluid stability described in previous reports of this series,⁽¹⁻⁵⁾ efforts were directed toward the revision of apparatus and technique in order to reduce areas of uncertainty found in the preliminary results. Improved photographic equipment was installed, and a superior method of obtaining the vertical temperature gradient within the undisturbed fluid of the tank prior to each run was devised. Further, an appreciable gain in the overall accuracy of particle displacement measurements and a considerable saving in the total time required for completion of a set of experiments has been effected by contracting with an outside organization to carry out the routine data-reduction phase of the program.

During the latter part of the period covered by this report, the experimental program was resumed with a series of runs under conditions of neutral stability.

Photography

Trajectory photographs obtained during the earlier runs contained inherent sources of uncertainty when it came to measuring particle positions, largely because of two factors. The first, an uncertainty as to just where the boundaries of a particular particle's image were for a given time on the photographic print, resulted principally from the use of a continuously rotating shutter to obtain consecutive images of the moving particles at one-second intervals. The shutter did not open and close cleanly; further, it necessarily remained open an appreciable period of time (approx 0.05 sec) in order to obtain sufficient light on the negative to record an image. Since the tracer particles traveled significant fractions of their diameters during this time, the images recorded were blurred.

The second factor contributing to the uncertain particle positions arose from the necessary employment of enlarged photographic prints as an intermediate step in the data-reduction process since the available digitized measuring equipment (Benson-Lehner OSCAR) was not of the type designed to evaluate projected images of negatives, but rather was one requiring opaque materials on the order of roll charts. Emulsion grain in the print paper, therefore, contributed further to an operator's indecision when attempting to center cross hairs upon the blurred image

of a tracer particle. An attempt was made to circumvent the tendency of paper prints to shrink nonuniformly after the wetting and drying stages of their development by using shrink-resistant, plasticized enlarging paper; but there is little doubt that some systematic distortion remained.

Steps designed to minimize these difficulties in recording and measuring the position of the tracer particles began with the construction of a high-speed, electronic flash-lighting system (Electronics Division) along the lines of similar equipment which had been successfully employed for tracer particle photography in fluid models by Prof. D. Fultz's laboratory at the University of Chicago. The unit built at Argonne supplies 30 to 300 watt-seconds of energy to from one to four xenon-filled, quartz flash tubes. Duration of illumination is of the order of one millisecond or less, and the power supply is large enough to maintain a repetition rate of several flashes per second even at the maximum power setting.

With four flash tubes installed directly in front of the two continuous mercury-arc light sources mounted above the test section of the towing tank, sufficient refracted light is seen by the camera in each tracer oil bubble to record a sharp image on medium-speed film (Kodak Plus X Pan, thin emulsion, fine grain) through an aperture of f-22. Not only does the short duration of the electronic flash improve the image delineation, but also the ample amount of illumination supplied makes it possible to use both a reduced lens aperture and fine-grain film.

The photographic-printing phase of the data-reduction process has been entirely eliminated by contracting with an outside organization equipped with projection film readers to measure particle displacements directly from the experimental negatives. The details are briefly described in a later section of this report.

Temperature Measurement

Vertical temperature gradients in the earlier, trial runs were measured by mercury-in-glass thermometers which were mounted along a vertical line very near the glass wall of the test section, a position required by the passage of the turbulence-producing grid. In order to obtain a measure of the gradient horizontally averaged through the undisturbed fluid just before a run, an optical temperature gradient measuring system was installed.

Systems of this type are based upon the fact that the index of refraction of water varies with its temperature in a manner that is known to a high degree of accuracy. An initially horizontal beam of light traversing a volume of water remains undeflected if the vertical gradient of temperature $d\theta/dz = 0$ but is curved downward if $d\theta/dz > 0$ (stable stratification) or upward if $d\theta/dz < 0$ (unstable stratification). For

small deflections, the angle of departure of the beam from the horizontal $\alpha = (x/\mu)(d\mu/dz)$ where μ is the index of refraction and x is the horizontal distance the ray has traveled in the fluid; $d\mu/dz$ may be readily converted to $d\theta/dz$ since $d\mu/d\theta$ is known.

For application of the method to the towing tank experiments three achromatic lenses (twelve-inch focal length) were mounted in such a way that they projected the images of miniature, uncoiled incandescent lamp filaments through the fluid of the test section at three different depths, the images appearing as bright horizontal indices on a graduated screen 6.8 meters from the tank. A one-millimeter deflection detected at the screen indicates a change in the average vertical temperature gradient of a little more than 0.02°C per cm. Measurements are made at three depths, one across the center of the test section and the others five centimeters above and below, in order to evaluate departures of the gradient from linearity.

Some dispersion of the filament image occurs because of the finite width of the pencil of rays projected and because of the polychromatic nature of the light source. When large temperature gradients are present, as there will be during the extremely stable fluid experiments, these dispersion effects will be more pronounced; it may be necessary to correct these two factors.

Analysis

Arrangements have been made with the Data Processing Division of the University of Dayton Research Institute to measure particle displacements directly from the 35-mm negatives produced here in the towing tank experiments by means of a digitized, projection film reader (Telereadex Corporation's Dilog 316). With this equipment, displacements on the negative are enlarged more than thirty times, this corresponding to a magnification of about five times the original displacements in the experiment itself. Tracer particle positions, located on the projected image by intersecting crosswires, are determined within a few hundredths of a millimeter, and the information is automatically punched on cards. After verifying the trajectory measurements by replotting them point by point with an automatic, digital plotter, the Dayton group transfers the data from the cards to paper tape in a format which can be fed directly into Argonne's digital computer GEORGE.

Further refinements made in the autocorrelation computation program have made this last step feasible, for initially the tape code employed at Dayton was incompatible with the input requirements of GEORGE. The program now also carries out the "block averaging" process of collecting correlation estimates and produces an output tape suitable for immediate use in the automatic plotter without the intermediate steps previously required.

With the procedures of data reduction and analysis outlined above, it will be possible to obtain finished autocorrelation function curves in from ten days to two weeks after the completion of the experimental runs, the postal service between Chicago and Dayton accounting for a large fraction of the delay. What is more important, it will no longer be necessary to suspend the experimental program for several weeks after each series of runs in order to reduce data it had taken only as many days to produce.

Mean Flow Studies

As has been described in previous reports, one of the problems associated with the generation of turbulence fields in the manner of this experiment is the simultaneous creation of undesired mean flow components. These are, in general, of two types, the one a steady drift in the form of an elongated toroidal circulation flowing along the tank axis and returning near the boundaries, while the other is an oscillatory surge associated with a standing wave. As a necessary prelude to attempts to decrease their magnitude and/or compensate for their effects, a series of experiments designed to measure these flow components was carried out.

With the camera circuits arranged to expose one negative for a second every two seconds and the electronic flash unit set to trigger in a pattern of three flashes marking both the speed and direction of each tracer particle in view, a series of runs otherwise identical with the neutrally-stratified fluid, turbulent trajectory experiments was carried out. Enlarged prints of the negatives obtained were analyzed in detail; between twenty and forty individual velocity samples distributed over the field were abstracted from each photo with the aid of Argonne's digitized reader, OSCAR. Averages of these data constituted measures of the total mean flow (drift plus surge) since the turbulent velocity components were largely removed by the space-averaging over the field.

The results of four such runs (twelve photos per run) were compared. They appeared to indicate that the drift component was not as constant from one run to the next as had been believed from the results of earlier runs. Values obtained ranged from -0.1 mm/sec to -1.9 mm/sec. However, it is probable that not enough time was allowed to elapse between successive runs; the remnants of large-scale, secondary circulations excited in the previous experiment were contributing to the mean flows observed. Additional mean flow runs are planned to investigate this point. In a series of turbulent trajectory experiments in which a minimum of thirty minutes was allowed between runs, the average drift of twenty-seven trajectories from as many runs was -1.3 mm/sec, a value lying in the same range as that obtained from the mean flow experiments.

The buoyancy of the tracer particles contributes an average vertical component to the trajectories that is analagous to the longitudinal drift in the horizontal. But since the vertical velocities which result vary with the particles' size and with the age and exact formulation of the tracer batch from which the particles came, this component can not be treated as a constant for all experiments. It must be determined from the average vertical displacement of each trajectory in the series.

The longitudinal surge component of the mean flow was found to be highly reproducible from one run to the next. Velocity averages obtained in the mean flow experiments indicated nearly identical surge amplitudes of ± 1.2 mm/sec in all four runs. However, the surge phase (relative to the time of grid passage) and period did not seem to be quite so uniform in the data. Therefore, at the suggestion of Prof. D. Fultz, a check was made of the phase and period of the varying pressure field coupled with the oscillatory motions.

A large cork constrained to move only up and down by a lightweight lever was floated on the free surface of the water above the test section in the opening provided at that point for the insertion of tracer. The float was connected to a small, pivoted mirror mounted overhead. Vertical motions of the water surface were thus converted into angular displacements of the mirror. Because the mirror pivot could not be based upon the tank lid which itself moves slightly in response to the surge component, the connection between the float and the mirror could not be completed until after the carriage had passed between them. This restricted the recording of the pressure waves to times later than fifteen seconds after the passage of the grid.

Small motions of the float and mirror were magnified by a 3.7-meter long optical lever. A narrow slit, illuminated once each second by one of the electronic flash tubes, was projected through the pivoted mirror to a recording point on the wall. With the aid of the eye's persistence of vision and a little practice it was possible to mark positions of the light slit flashed upon a piece of paper at intervals of a second with surprising accuracy.

The amplitude of the pressure wave appeared to be as constant as the surge in the velocity field. Its magnitude in terms of the movement of the free water surface was of the order of ± 1 mm; but since the output of the make-shift float and mirror system is dependent upon its unknown response characteristics, the velocity measurements are the more accurate estimate of the surge amplitude.

The average period of the oscillation was 6.7 sec; in thirty-nine measurements of the surge period obtained with the float and mirror in nine consecutive runs, twenty-seven of these were within ± 0.2 sec of the

mean while thirty-three were within ± 0.5 sec. As to the phase, the average arrival time of one of the troughs in the free surface over the test section was 24.9 sec after the passage of the grid, all nine estimates lying within ± 0.3 sec of this mean. Again, however, the probability that a phase shift was present in the response of the recording system cannot be ignored; the measurements verify that the phase of the surge is constant from one run to the next but do not specify just what the phase is relative to the time of grid passage.

Thus it has been shown that the surge component of the mean flow is essentially constant in amplitude, period, and phase in consecutive runs in neutrally-stratified fluid with a grid velocity of 25 cm/sec; therefore, a constant surge component can be subtracted from turbulent trajectory data obtained in a series of experiments of that same kind.

Turbulent Trajectory Experiments

The experimental program was resumed with a rerun of the neutrally-stratified series using the same grid driven at the same velocity ($\frac{3}{8}$ -inch bar, 2-inch mesh, 25 cm per sec). In addition to the improvements in experimental apparatus and methods discussed in previous sections, the photos in these later runs were more accurately timed than those of the earlier series through the use of an automatic camera-starting arrangement in all experiments. This insured that the Batchelor decay corrections⁽³⁾ were determined from the same base time relative to the grid passage for all trajectories.

The superior quality of the photographs taken in the more recent runs made it possible to select only complete trajectories for analysis; it was never necessary (as it had been before) to choose one whose beginning or end was obscured. Therefore, there were no missing particle-velocity data; the input data matrix fed to the computer was complete. This maximized the number of velocity products in each correlation estimate.

The horizontal and vertical Lagrangian autocorrelation curves obtained are shown as solid lines in Fig. 48; the points are the block-averaged correlation estimates. Included for comparison are the dashed lines representing the autocorrelation functions obtained in the preliminary trials (913-929 X and 913-929 Z). Both because of the improved conditions of the experiment and because the individual autocorrelation estimates obtained before block-averaging showed significantly less scatter in the newer runs, the solid curve is considered the more reliable. In its computation, mean flows consisting of individual drift components determined from each trajectory plus a constant 6.7-sec period oscillatory

surge of ± 1.0 mm per sec amplitude were subtracted from the horizontal turbulent velocity data; average buoyant vertical velocity components measured from each trajectory were subtracted from the vertical turbulent velocity data.

Fig. 48 Lagrangian autocorrelation curves obtained in turbulent trajectory experiments in neutrally stratified fluid; solid lines are the more recent findings (Runs 1638-85X-1,3,6 and 1638-85Z-1,5), while the dashed lines represent the results of preliminary trials (913-29X-5 and 913-29Z-2). All experiments made with $\frac{3}{8}$ -inch bar, 2-inch mesh grid driven at 25 cm per sec; average vertical temperature gradient within one hundredth of a degree C per cm of isothermal.

The two horizontal autocorrelation curves are not very different from one another in the first half except for a more rapid fall from unity in the recent results. On the other hand, the first portion of the newer vertical autocorrelation curve differs markedly from the preliminary results. This is probably due to the reduction of the effects of the apparent vertical mean flow component achieved both by an improvement in tracer density control during the experiments and by the application of individual trajectory buoyancy compensations during the analysis.

The principal differences in the two runs seem to show up in the latter portions of both the horizontal and the vertical autocorrelation curves; the "time to zero" appears to have been delayed. For a number of reasons, the difference may be less significant than it appears:

- a) the number of velocity products involved in each correlation estimate decreases toward the latter part of the curve;
- b) correlation estimates for larger lag intervals, ξ , are based upon combinations of one velocity from the beginning of a run and another from the end, and therefore these estimates are more sensitive to errors introduced by inaccuracies in the Batchelor decay corrections;
- c) the newer autocorrelation curve computation included velocity measurements taken one second closer to the time of the grid passage than was the case in the computation of the preliminary curve; and this may have contributed to a shift in the correlation estimates at high lag times in the more recent results. This last possibility was checked by running an autocorrelation computation with the current data after the first velocity measurement has been deleted. Only insignificant changes in the results were observed.

Conclusion

The experimental program will now continue by exploring a number of stable-fluid cases, the improved temperature gradient measuring system and the heat exchanger on the tank bottom making it possible to hold a particular degree of stable stratification for the duration of a complete experimental series. In this way it is hoped that significant trends in the Lagrangian autocorrelation function with changing fluid stability will be found.

A note of appreciation is due for the recent contributions of a number of individuals to this program. H. Cremer of the Electronic Division constructed the electronic flash equipment, adding a number of improvements to the original designs supplied by Professor Fultz's group at the University of Chicago. The optical temperature gradient measuring method was an outgrowth of several conversations with Dr. H. A. Schultz of the Radiological Physics Division, while R. F. Selman constructed the necessary equipment. The computer program has continued to improve under the ministrations of Mrs. C. C. Chamot of the Applied Mathematics Division, the recent achievement of compatibility with a foreign input code making the use of the services of the Dayton Data Processing group feasible.

With regard to the experiment itself, the author would like to express his thanks to Professor D. Fultz, University of Chicago, for his valued suggestions and encouragement.

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ERRORS IN WIND MEASUREMENTS ASSOCIATED WITH TOWER-MOUNTED ANEMOMETERS

Harry Moses and H. G. Daubek

I. Introduction

Measurements by anemometers mounted on towers are subject to errors arising from the obstruction to wind flow by the tower. Although this fact is commonly recognized, only rarely do meteorological installations have quantitative information on the magnitude of this effect. In addition, the literature on the subject is very sparse.

This paper describes an investigation of the wind shadow effect made by comparing the data from two Aerovane anemometers, one mounted on a boom extending from a tower of lattice construction and the other mounted on top of a utility pole. Both anemometers were located 18.75 feet above ground at the meteorological field site of the Argonne National Laboratory.

II. Description of Meteorology Site and Experimental Equipment

The meteorological field site is located 746 feet above sea level in an open meadow. A prominent feature on the site is the Meteorology Tower which is a standard fire lookout tower used to support meteorological equipment. The base of the tower cabin is 120 feet above ground and a steel mast extends upward through its roof to 150 feet.⁽¹⁾

The nearest obstruction to the Meteorology Tower is the Meteorology Building located approximately 300 feet due west of it (Fig. 49). This building is 65 x 20 x 13 ft and is oriented with its largest dimension in the east-west direction. Other structures in the area are the storage shed which is 20 x 11 x 9½ ft, and the pump shed which is 6 x 6 x 8 ft. The terrain is clear of trees 1400 feet to the north, 1500 feet to the east, 1600 feet to the south and 1200 feet to the west of the tower.

Figure 50 shows the anemometer which was mounted on a utility pole 8.4 inches in diameter with a 2-inch pipe attached to the pole support. The hub of the anemometer rotor is 23.5 inches above the pole; thus there is no wind shadow effect on this instrument due to its mounting structure.

The Meteorology Tower (Fig. 50) has a cross section of 25 feet square at the bottom, but tapers to 7 feet square at the base of the cabin. At 18.75 feet the cross section is 22 feet square. When the wind is from the north, the distance between the Aerovane rotor hub and the tower leg is at a minimum distance of 9 ft 1 in. Thus the tower subtends an angle of 68° at the anemometer.

Fig. 49 Schematic layout of buildings in relation to anemometer pole and Meteorology Tower.

To determine the degree of obstruction by the north-south oriented tower, measurements were made of the tower components such as the beams, catwalks, stairs, railings, conduits and conduit boxes. These were projected on a vertical plane, P_N , passing through the vertical axis of the tower and equidistant to its north and south faces. Measurements were made for sections of the tower bounded by the horizontal planes marked A and A' and also B and B' in Fig. 50. The vertical separation between planes A and A' is 13 ft 7 in., and between B and B', 26 ft 10 in. From this figure it may be observed that the anemometer is approximately halfway between A and A' and also B and B'.

The tower components projected on plane P_N occupy an area A_N . Let us designate as A_t the total area on plane P_N bounded by planes A and A' and located within the tower. The ratio of obstructed to unobstructed area, $A_N:A_t$ is a measure of the interference to wind flow. This ratio is 0.21 both for the section of the tower bounded by planes A and A' and for the larger section bounded by planes B and B'.

When the projection plane is rotated 90° so that it is equidistant from the east and west faces of the tower, the obstruction ratio between planes A and A' is 0.22 and between planes B and B', 0.20. The obstruction ratio varies somewhat with the separation of the horizontal planes such as A and A', i.e. with the size of the tower section considered and with the orientation of the projection plane.

The Aerovane operates with a propeller-type rotor attached to the shaft of a dc generator whose voltage output is proportional to the magnitude of the wind speed. Tests made on the equipment before, during, and after the period of study showed that the manufacturer's calibration curves

Fig. 50. View of Meteorology Tower facing southeast and reference anemometer pole

remained valid. A comparison test, made by operating the two anemometers side by side for a period of several hours, indicates that for speeds up to 13 miles per hour their readings checked to within one mile per hour. Higher speeds were not recorded during the comparison test period.

The orientation test of the Aerovanes, routinely made with a transit during major servicing, indicated that within the period of this study the tower anemometer was within 1° of the true direction and the pole anemometer was within 4°.

III. Analysis of Data

This study is based on observations beginning at 1000 CST, July 27, 1954, and ending at 2400 CST, February 28, 1955, covering a period of 5,199 hours. Wind-direction data from the tower were missing for twenty hours - from 1600 CST, October 10, 1954, through 1100 CST, October 11, 1954, and in addition, there were 135 hourly observations during which the wind direction was considered calm since the direction trace appeared as a straight line and the wind speed zero. An additional forty-four hourly observations in which the direction was extremely variable and essentially indeterminate were omitted. The study is, therefore, based on 5,000 hourly observations.

Wind direction and speed were recorded on Esterline-Angus recorders and read to the nearest mile per hour and to the nearest ten degrees.

Ten-minute averages of wind speed and direction beginning at five minutes before each hour were chosen for the comparison. These data were entered manually on data-evaluation forms and key punched on IBM cards for use with an IBM 650 computer.

In preparing the data for analysis, for each 10-degree wind direction, the wind speeds were divided into five-mile-per-hour groupings: 0-4, 5-9, 10-14 and greater than or equal to 15 miles per hour. For each 10-degree direction and wind speed group the following ratio, R, was determined.

$$R = \frac{\text{Sum of wind speeds on tower}}{\text{Sum of wind speeds on pole}} .$$

These ratios, plotted on polar coordinate paper, are shown in Fig. 51.

Fig. 51. Ratio of tower anemometer speeds to pole anemometer speeds as a function of wind direction measured on tower (relative size of Meteorology Tower and boom indicated in northeast sector)

IV. Discussion of Results

An inspection of Fig. 51 shows that there is a substantial reduction in the measured wind speed when the wind blows through the tower before reaching the anemometer. On a percentage basis, the reduction is greatest for light winds. For example, for a wind blowing from 20° , the ratio, R , is 0.55 for tower wind speeds of 0-4 miles per hour, 0.67 for speeds of 5-9 miles per hour and 0.74 for speeds of 10-14 miles per hour. The 20° direction is the direction for which the speed reduction was largest. It is of interest to note that as the wind blows along the sides of the tower before reaching the anemometer there is an increase in the measured wind speed of the tower anemometer as compared with that of the pole anemometer. This may be seen from the R values for the wind direction of 320° . For 0-4 miles per hour the ratio is 1.31; for 5-9 miles per hour, 1.17; for 10-14 miles per hour, 1.13; and for 15 miles per hour or greater, 1.15. A similar speeding up of the wind may be seen for the 120° wind direction. This velocity field about the tower, characterized by a retardation of the flow through the tower and an acceleration along the sides, is not surprising in view of the existing theory and accounts of experiments of turbulent flow about arrays of solid obstacles.(2)

Another maximum occurs in the diagrams for winds blowing from 230° . This may be due to the shadow effect of the Meteorology Laboratory upon the pole anemometer. Even though this building is about 160 feet from this anemometer, it appears to influence the air flow sufficiently to cause a reduction in the measured wind speed.

To study the effect of the tower on the wind direction, the average direction on the pole was determined for each 10° direction on the tower. The differences between the average tower and pole directions were examined and found to agree to within 11° . The tower appeared to influence the air to flow along its sides. Air flow from the northwest sector, where the maximum differences were measured, veered about the tower so that direction readings on the tower exceeded those on the pole. Except for winds from the northwest, differences in direction between the tower and pole were within about 5° .

V Conclusion

This study indicates that serious errors occurred in measurements made with the tower-mounted Aerovane, due to a tower wind shadow effect. The air flow on the lee side of the tower may be reduced to nearly one-half its true value for light winds and nearly 25 percent for speeds of 10-14 miles per hour. An increase in measured wind speed exceeding 30 percent occurred when the wind blowing toward the anemometer made an angle of 20-40 degrees with respect to the sides of the tower adjacent

to the anemometer, i.e., when the light winds were from the northwest or southeast sectors. The shadow effect is greatest percentage wise for the lower wind speeds. In Fig. 5ld the high values are not significant because of the paucity of observations in the ≥ 15 mile-per-hour speed group.

The effect on direction appeared to be relatively smaller with the greatest mean deviation of 11° occurring with winds making an angle of 70° with respect to the west side of the tower, i.e., the wind was west northwest.

These results show that measurements made with tower-mounted anemometers are subject to serious errors. When towers are used as platforms, the magnitude of the errors arising from the tower wind shadow effect should be ascertained for precise work.

Thanks are due to Professor Horace R. Byers, Chairman, Department of Meteorology, University of Chicago, for his counsel during the course of this study. The aid provided by Mr. Merlin H. Dipert of the Division of Biology and Medicine in programming the study for the IBM 650 computer is gratefully acknowledged. Messers. Frank C. Kulhanek and Gunther A. Zerbe helped materially with the reduction of the data and numerous measurements in the field.

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ATMOSPHERIC BOUNDARY LAYER SIMULATION TECHNIQUES FOR SMOKE DIFFUSION EXPERIMENTS*

Robert F. Weiss**

I. Introduction

The effect of convective turbulence in the atmosphere on the diffusion of stack gases has been recently studied in wind tunnel experiments⁽¹⁾ for the purpose of determining the relationships between scale and field diffusion data. In the development of the necessary scaling criteria, techniques, etc., it became apparent that accurate simulation of the atmospheric boundary layer was required. Most characteristic of this layer are its thickness, velocity profile, turbulence-intensity distribution, scale of turbulence, and spectrum of turbulent fluctuations. Ideally, these parameters should be controllable in a wind tunnel test, i.e., they could be designed, created and stabilized. The possibility of accomplishing this has now been investigated, this report discussing one approach to the problem and presenting some preliminary experimental results.

II. Notes on Experimental Technique

The first objective was the creation of a relatively thick boundary layer at the most upstream station of the wind-tunnel test section. Previous tests under the Argonne National Laboratory contract had indicated that even with three-quarters of the test-section floor roughened (artificially) the resulting boundary layer was insufficiently thick to immerse the smoke plume from the model Argonne Meteorology Experimental Stack. Of major importance also, was the preservation of a predesigned velocity and turbulence profile in a nearly constant-thickness layer over the length of the test section. Three difficulties in wind-tunnel simulation of the atmosphere result from size consideration: (1) boundary layer formation over a short flat plate differs from that over the surface of the earth in that a nearly constant thickness is not ordinarily approached (as in the atmosphere), (2) the boundary layer thickness is artificially limited by the tunnel ceiling, and (3) the size of the eddies formed (also, as a result, the scale and spectrum of turbulence) is restricted by tunnel dimensions, and not generally in accordance with scaling criteria.

*Work performed under contract between Argonne National Laboratory and New York University under the direction of Professor Gordon H. Strom, Director of Wind Tunnels, New York University, and supervised by Harry Moses, Radiological Physics Division, Argonne National Laboratory.

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Starting, then, with the pertinent meteorological information to be simulated (velocity and turbulence profiles, scale of turbulence), a straightforward mechanism was designed for constructing a boundary layer of desired characteristics. Using a series of channels, vertically stacked according to a geometric progression (Figs. 52 and 53) at the upstream end of the test section, a boundary layer was to be roughly pieced together

such that in its downstream travel it would become continuous, (with "smooth" variations of parameters across the layer). The design of the channels together with the dimensions and spacing of the roughness elements (see Table 24) was based on both theory and experiment.(2)

Fig. 52. Configuration of channels for velocity-profile control.

It was assumed that the velocity profile follows the exponential law

$$\frac{u_1}{u_2} = \left(\frac{Z_1}{Z_2} \right)^{\frac{n}{2-n}}$$

where u is wind speed, Z is height, and n a nondimensional turbulence

parameter. The velocity profile was "finely adjusted" by adding or removing roughness elements. However, a more efficient arrangement should be devised to raise or lower the $4' \times 7' \times \frac{1}{8}"$ aluminum channel plates (screw

Table 24

Channel design parameters

Channel height, inches	Height of roughness elements, inches	Spacing of roughness elements, inches
1	0.1	0.1 (sandpaper)
2	0.5	1.0
4	0.75	5.0
8	1.5	8.0
16	-	-

Fig. 53. Photograph of channels for velocity profile control

(Photograph courtesy of New York University)

jacks, possibly), and thereby change the velocity profile independently of the turbulence-intensity distribution. In general, it was necessary to remove a significant number of roughness pieces to increase the mean velocity in a channel. Also, it was found that the curvature of the velocity profile was more difficult to control in the region near the floor of the test section than in midstream or above. Temperature gradients in these preliminary tests were kept fairly close to $+\frac{1}{2}^{\circ}\text{F}$ per meter, i.e., the top of the tunnel was warmer.

The velocity-measuring instrument used was a manually-operated, constant-voltage, hot-wire bridge anemometer, capable of indicating rapidly fluctuating velocities on both coarse and sensitive scales. Readings of root-mean-square velocities, correlation coefficients and spectral distributions of fluctuating components will be possible with further improvement of the equipment. Control of the mean frequency of fluctuation and scale of turbulence might then be achieved.

Semiquantitative measurements of turbulence intensity were obtained from the following relation

$$I = \frac{\sqrt{(u')^2}}{u_{\infty}} = \frac{u'_{\text{rms}}}{u_{\infty}} \approx \frac{u_{\text{max}} - \bar{u}}{\sqrt{2} u_{\infty}}$$

where I is turbulence intensity, u_{∞} is the mean free stream velocity, and u' is the deviation from the mean velocity $\bar{u}$. The value of $u_{\text{max}} - \bar{u}$ can be determined from the hot wire anemometer fluctuations.

III. The Experimental Results

Measurement of the velocity profile at several stations downstream of the profile-producing apparatus was carried out with free-stream velocities of 2.5 and 1.5 feet per second. These data are presented in Fig. 54

Fig. 54. Velocity profiles at distance (X) downwind from stack

and Fig. 55, which show the creation and propagation of a smooth-profiled, "constant-thickness" (20 inches) boundary layer. A test run made with $\frac{1}{2}$ -inch floor roughness downstream of the apparatus revealed only a slight smoothing of the lower part of the layer. A logarithmic plot of the 2.5-foot-per-second data is given in Fig. 56, the straight line obtained indicating that the velocity profile was exponential. Turbulence-intensity distributions were also well-behaved at both velocities and are plotted for 2.5 feet per second in Fig. 57.

The effect of increasing the thickness of the boundary layer upon smoke-plume behavior was examined by comparing smoke plumes of the current series with those of previous experiments. With this in mind, one of the situations (Run No. 5a) previously used in the ANL tests⁽¹⁾ was reproduced: A free-stream velocity of 2 feet per second was set in the test section, and the velocity and turbulence-intensity profiles obtained are sampled in Fig. 58 at a position immediately upstream of the smokestack. Note that the 20-inch

Fig. 56. Velocity profile at 3 feet downwind from stack.

Fig. 55. Velocity profiles at distance (X) downwind from stack.

thickness of the boundary layer, as compared to a 10-inch thickness obtained without the profile apparatus, Fig. 59, placed the smoke plume in a relatively lower position in the layer. Being in a region of greater shearing stress and turbulence, it was not surprising to see greater diffusion of smoke and a wider plume formation downstream of the stack. This behavior is illustrated in multiple-exposure photographs (Figs. 60 and 61) which show plume formation with and without boundary-layer simulation. Furthermore, it is interesting to note that this tendency towards a wider plume brings more closely together the scaled and field results.

At 100 feet downstream of the plume's virtual origin, for example, the half-width becomes 3.7 meters with, as compared to 2.8 meters without simulation. The relative widening of the plume is even

Fig. 57. Turbulence-intensity profiles at distance (X) downwind from stack (mean free stream velocity, 2.5 feet per second)

Fig. 58

Velocity and turbulence-intensity profiles with profile-control apparatus (18 feet downwind from stack, mean free stream velocity 2 feet per second)

Fig. 59. Velocity and turbulence-intensity profiles without profile-control apparatus (immediately downwind from stack, mean free stream velocity 2 feet per second)

Fig. 60. Plume formation with boundary layer simulation
(Photograph courtesy of New York University)

Fig. 61. Plume formation without boundary layer simulation
(Photograph courtesy of New York University)

greater at distances farther downstream. Beyond the initial expansion region of the smoke jet, the aerodynamic characteristics of the boundary layer become more significant and those of the jet less important, resulting in greater differences in plume width with distance (at 100 feet the percentage difference is 30%, while at 200 feet it becomes 60%). Future tests will present sufficient data for comparison with field results. The most desirable relative position of the plume in a boundary layer must be determined from scaling criteria, allowing then the tailoring of the layer to this specification. It should be noted that the wavy outline of the plume in Fig. 60 is due to uneven background shading, rather than to meteorological phenomena.

IV. Conclusions

It has been shown, in this study, that:

- (1) It is possible to design and control velocity and turbulence profiles, under fairly low windspeeds, at the upstream end of a wind-tunnel test section.
- (2) The profiles constructed are relatively stable with regard to shape, thickness and turbulence intensity in downstream travel of more than 20 feet.
- (3) Smoke-plume formation is affected by the boundary-layer structure, and there is reason to believe that close correlation between scaled and full-scale plume behavior is quite possible with the use of boundary-layer simulation techniques.

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