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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

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Human Studies Project

Summary

PART I. RADIOACTIVE WASTE DISPOSAL

1. Liquid Injection into Deep Permeable Formations

Movement of radionuclides through porous media is due to movement of the transporting water, but is also affected by the ion exchange properties of the medium. Calculations were made for the movement of the solution and radiostrontium through a slab of Berea sandstone and were confirmed by experimental data.

The behavior of chloride as a water tracer is complicated by its repulsion by negatively charged soil colloids. The relative exclusion of chloride is especially complicated when clay minerals are present and restricted in their degree of lattice expansion. However, these effects would be decreased as the concentration of electrolyte is increased and would probably be rather insignificant in "clean" sandstones having low specific surface and high porosity, especially if most of the surfaces were sufficiently separated to permit full development of an electric double layer.

Anion exclusion gives a good measure of lattice expansion and collapse of hydrobiotite systems, thereby providing a rather accurate method for measuring the distribution of surface charge density of these interstratified materials.

2. Disposal by Hydraulic Fracturing

Four water injections were made in the injection well at 986 and 966 ft. These tests confirmed other evidence that a fluid loss additive is not a necessary component of the mix to be used in the waste injections. Also, since in all cases the fracturing pressure was well above the overburden pressure, these tests furnished presumptive evidence that horizontal fractures were formed.

On the last water injection, the sand-erosion technique for slotting failed to cut the casing after 20 min at a jet pressure of 2600 psi. To determine the reason for this failure, tests were run by the Halliburton Company in their test well. During the tests, it was observed that considerable oscillation of the jetting tool was cutting a much wider slot than previous experience had suggested. This was probably responsible for failure of the slotting operation at Oak Ridge, since more time would be required to erode a wider slot and no allowance had been made for such a factor. In any case, these tests showed that the $5\frac{1}{2}$-in. casing of the injection well could be cut despite the oscillation of the jet if a jet pressure of 4000 psi and a cutting time of 30 min were used.

An estimate of the probable operating cost of the shale fracturing plant shows that the cost of the mix and the well life are the significant variables. The well life can be significantly extended by making the batch size large. For this reason it seems probable that the direction of future work will be toward larger batch sizes than are being injected at the present time.

A hazards analysis of the shale fracturing experiment has been completed. It was concluded that the waste solution to be used in the first series of injections would be so low in specific activity that no significant hazard would result from its use. The other principal hazard involved in the shale fracturing experiment – that of working with high-pressure equipment – has been minimized where possible by installing this equipment in cells.

To verify that the Densometer system could be used to control the solids-to-solution ratio at very low proportions of cement, a mix test was run by the Halliburton Company. The Densometer controlled the density of the slurry to within 0.1 lb/gal of the desired density, which is entirely acceptable. It was found, however, that the

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density indicated by the Densometer was uniformly 0.8 lb/gal lower than the true density. This error in calibration can be compensated for by a change in the Densometer setting.

Following construction of the surface plant, a series of five injections was made to determine the performance of the plant and the underground behavior of waste slurries of different compositions.

In the first injection (February 13), 37,000 gal of synthetic concentrated waste solution was mixed with 23,400 lb of attapulgite drilling clay and injected at a depth of 945 ft. This injection was made to check the operation of the surface plant and to evaluate a nonsetting mix.

In the second injection (February 21), 27,300 gal of synthetic concentrated waste solution with 30 curies of ^{198}Au tracer was mixed with about 170,000 lb of a cement-base mix and injected at a depth of 924 ft. This injection was made to evaluate a mix with a low cement concentration and to determine whether the activity in the grout sheet could be detected at the observation well.

In the third injection (April 3), 33,500 gal of a mixture of synthetic concentrated waste solution and actual intermediate-level waste was mixed with about 518,000 lb of a cement-base mix and injected at a depth of 912 ft. This injection was made to demonstrate that concentrated radioactive waste solutions similar to those to be produced by the ORNL waste evaporator can be fixed permanently underground.

In the fourth injection (April 17), 36,000 gal of intermediate-level waste solution was mixed with about 381,000 lb of a cement-base mix and injected at a depth of 900 ft. This injection was made to evaluate a mix for dilute waste solutions.

In the fifth injection (May 28), 148,000 gal of a mixture of synthetic waste solution and actual intermediate-level waste was mixed with about 1,040,000 lb of a cement-base mix and injected at a depth of 890 ft. This injection was made to test the surface plant during an extended (11-hr) disposal operation and to determine the underground behavior of large injections.

Minor difficulties were encountered with various components of the surface plant during the injections. These difficulties were corrected as they appeared. In general, the operation of the surface plant has been smooth and satisfactory; the disposal operation has become almost routine as experience has been accumulated.

Evaluation of the mixes used in the five injections and determination of the underground behavior of the grout sheets cannot be accomplished until cores of the formation have been recovered. Coring is expected to begin in July.

3. Disposal in Natural Salt Formations

Renovation of the topside facilities at the Carey Salt mine, Lyons, Kansas, is essentially complete. A new 7-ton capacity headframe has been erected; the existing hoist has been reworked, all electrical circuits and switchgear have been replaced, and the hoist house has been renovated. Drilling has started on the 19.1-in.-ID waste-charging shaft.

Mining equipment for the underground work has been purchased and installed in the mine. Cleanup of the main access tunnels in the mine is in progress. Layout of the experimental area has been completed, and mining should start early in July 1964.

Concurrent with the mine cleanup, plastic flow gages are being installed along the access tunnels and near the experimental area. The information from these gages will be used to determine the effect of increased temperature on stability of the mine.

Design of equipment for handling and storing the radioactive fuel assemblies is essentially complete. Fabrication of all handling and storage equipment is in progress, with the exception of the fuel element canisters. A draft of the hazards report on the shipment of fuel assemblies between Idaho and Lyons, Kansas, has been completed.

Laboratory tests of 1000-hr duration on pillar models made from Lyons salt have been run by the U.S. Bureau of Mines. Tests were run at pillar stresses ranging from 4000 to 10,000 psi. From the results of these tests, an empirical equation was derived which correlates well with measured vertical closure rates in the Lyons and Hutchinson mines in openings up to 70 years old.

The isolated 2 × 8 × 10 ft heated model room (wall temperature approximately 140°C), after more than a year of heating, has a closure rate ten times higher than it had at ambient temperature.

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4. Clinch River Study

Analyses of stable chemical constituents in discharge-weighted water samples from the Clinch and Tennessee Rivers show that variations in constituent concentrations are not great; bicarbonate is the predominant anion and calcium the predominant cation. Seasonal variations in concentration of several of the constituents were detected. Calculation of correlation coefficients for constituent pairs indicated that turbidity, apparent color, centrifuged color, suspended solids, iron, and manganese concentrations varied in a similar manner.

Measurement of gross gamma radioactivity in 146 cores of Clinch River bottom sediment has shown that radioactive sediment up to 8.7 ft thick is present in the river. Variations in radioactivity with depth were observed in several of the cores. The general pattern of variation is strikingly similar to the pattern of annual ^{137}Cs releases at White Oak Dam, and is assumed to be the result of continuous sedimentation of cesium-bearing clay particles suspended in the water.

Regulation of flow in the Clinch River due to power generation at Melton Hill Dam will produce greater daily changes in flow conditions than did similar operations at Norris Dam. Tracer studies were conducted to study the effect of releases from Melton Hill Reservoir on dispersion of radioactive releases from White Oak Dam. Pulses of tracers released twice daily from the creek, simulating summer conditions, were observed to occur at about 10 AM and noon each weekday at the ORGDP water treatment plant intake. During the normal weekend shutdown of operations at Melton Hill Dam, the mass of tracer released from the creek built up in the river. Power releases on Monday moved this maximum concentration of tracer downstream. The minimum dilution for the momentary peak on Monday at ORGDP water intake was computed to be 50. This minimum dilution is only about ten times less than the median dilution observed prior to construction of Melton Hill Dam.

Change in flow conditions due to Melton Hill Dam has necessitated a change to automatic proportional sampling of the river water.

Analyses of fish taken from the Clinch River show that about 2.5% of the maximum permissible intake (MPI) would be received from eating 37 lb of fish per year. Radiation levels near filter beds in water plants upstream and downstream from White Oak Creek are at background.

Bottom sediments from the Clinch River were selectively leached. Salts between a pH of 6 to 8 removed less than 10% of the ^{60}Co, ^{106}Ru, and ^{137}Cs, though up to 80% of the ^{90}Sr. At lower pH's up to 80% of the ^{90}Sr and ^{60}Co were leached. At higher pH's up to 50% of the ^{106}Ru and up to 20% of the ^{60}Co are released.

5. Movement of Radionuclides in Terrestrial Environment

Core samples taken in and around waste pits 2 and 3 show that most of the ^{137}Cs and ^{90}Sr is concentrated in sludge and precipitates in the pits and on the first few inches of shale comprising the side walls and bottoms of the pits. By the use of tritiated water as a tracer, it has been observed that the normal rate of ground-water movement in the waste pit area is about 0.5 ft/day.

Most of the 700 curies of ^{137}Cs in the bed of White Oak Lake occupies highly selective exchange sites on the illitic clay fraction of the lacustrine sediment and can be desorbed only by disruption of the clay lattice structure.

Analyses of selected samples obtained from the upper 0.2 ft of White Oak Creek bottom sediment indicate that less than 5×10^5 $\mu\mu c$ of ^{137}Cs and substantially smaller quantities of ^{60}Co and ^{106}Ru are stored in this zone. The inference is drawn that the residence time for the less-than-63-μ sediment in this zone is relatively short and that the material is scoured during higher-than-normal stages and transported into White Oak Lake.

As an extension of information gained from laboratory studies on radionuclide sorption, field studies on deliberately contaminated plots, using ^{137}Cs, have been started in order to correlate areal and vertical redistribution of the nuclide with natural factors, including biologic, meteorologic, and pedologic factors.

6. Mineral Exchange Studies

In sorbing trace concentrations of strontium, ideal ion exchange behavior, based on mass action

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expressions, was exhibited by several of the clay minerals under special conditions. Hydrous sesquioxides sorbed strontium preferentially, and the strontium sorption curve did not yield the predicted slope for ideal behavior. Several soil materials were tested; the local Conasauga shale, whose sorption indicated strong influence of sesquioxides at alkaline pH, was shown to behave according to the mass action law when the free iron oxide was removed.

Ethylenediaminetetracetate (EDTA) forms a more stable complex with calcium than with strontium. In a waste solution containing calcium as a major ionic component, the major breakthrough of strontium through an ion exchange column can be delayed by complexing part of the calcium with EDTA. However, the delayed breakthrough of strontium is gained at the expense of increased "leakage" of strontium through the ion exchange column.

7. Engineering, Economics, and Safety Evaluations

Costs of permanent storage of calcined radioactive wastes in concrete vaults were five to seven times greater than those for storage in salt, and costs for storage in rooms mined out of granite formations were twice those for storage in salt mines. This economic advantage, plus greater safety, makes salt the preferred choice.

Study of the comparative costs for disposal in concrete vaults, hard-rock mines, and salt mines was based on the costs for disposal in salt developed in a previous study. In the preliminary study, costs for excavation of salt were assumed to be $2/ton. During the year, work has continued on a final report of costs for disposal in salt. Excavation costs have been calculated as a function of the amount of salt mined yearly, and $2/ton was found to be a reasonable figure for a relatively large operation. Costs per ton for small operations would be higher.

Estimates were made of the movement of ^{90}Sr, ^{106}Ru, and ^{137}Cs in Conasauga shale following a hypothetical release of reactor fuel-reprocessing waste from a tank. Computations were based on hydrologic and exchange characteristics of the local formation.

In Conasauga shale, ^{90}Sr would apparently move quite rapidly in an acid waste system, even if the acid would be neutralized by calcite ($CaCO_3$) in the formation. Movement of ^{90}Sr in an alkaline waste solution would be much slower due to the formation of insoluble compounds, in addition to the greater holdup by ion exchange. Ruthenium-106 would be expected to move rapidly in either acid or neutralized waste systems, but it presents little long-term hazard due to its relatively short half-life. The movement of ^{137}Cs would be expected to be slow because of its strong fixation by the ion exchange sites of the illitic fraction of the Conasauga shale.

8. Related Cooperative Projects

The U.S. Geological Survey has continued to obtain partial-record base-flow and crest-stage records for the area. In addition, a geologic map of White Oak Creek Basin, with an explanatory text, has been issued. A generalized soils map of the basin is being prepared.

The work of the multiagency steering committee for the Clinch River Study has continued.

Three alien guests have been on assignment during the past year.

Several members of the Section have continued to participate in the following activities: the ASA Committee N5, the AEC Advisory Committee on Deep-Well Disposal, the ASCE Committee on Sanitary Engineering Aspects of Nuclear Energy, and various ORNL special committees. Members have also contributed to the journal *Nuclear Safety* and have served with the Nuclear Safety Information Center.

In addition, courses and lectures have been given for ORSORT, Vanderbilt University, University of Tennessee, and the U.S. Public Health Service.

PART II. RADIATION ECOLOGY

9. Radioactive Waste Area and Radiation Effects Studies

Statistical analyses of sedges growing on White Oak Lake bed indicated that length of inflorescence was mostly correlated with the air dose rate of gamma radiation at 1 m above ground surface. Some of the natural vegetation around the unshielded Health Physics Research Reactor exhibited the first visible effects of radiation

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damage. After a period of winter dormancy, new growth produced in several species of forest trees contained many morphological abnormalities. The damaged vegetation occurred at distances of 10 to 20 m from the reactor, and total cumulated dose was approximately 600 to 140 rads (tissue dose in air) respectively. Radiation effects studies completed this year included fast-neutron exposures from the HPRR, and gamma-radiation exposures from both a ^{137}Cs field source and the White Oak Lake bed. Among the findings were that all acute fast-neutron doses ranging from 100 to 500 rads resulted in 100% mortality of pine seedlings although the time required to reach complete needle mortality varied widely within and between dose groups. Pine seedlings subjected to chronic gamma irradiation from White Oak Lake bed for 178 days exhibited 100% needle mortality, while controls averaged only about 10%. During the past fiscal year, a study was initiated to determine the seasonal changes in nuclear volumes of local tree species. Shoot apexes were measured monthly from 14 local species. An analysis of variance showed that most species increased significantly ($P < 0.01$) in size before shoot elongation. Initial spring activity of nucleic acids in shoot apexes is thus earlier in eastern Tennessee than would be anticipated from any external evidence. Continuing work on the use of metaphosphate glass dosimeters demonstrated that low-Z glass rods are subject to fading in sunlight. Predosed rods lost 37 to 46% of the dose during the first 6 hr of exposure to sunlight. Fading was also related to total predose since high predose rods had a significantly greater ($P < 0.01$) percentage loss than low predose rods.

Mud-dauber wasps that used radioactive mud from the liquid waste pits were found to be affected by the ionizing radiation. In the case of the species *Sceliphron cementarium*, the effect of the radiation is to reduce the successful emergence by a significant 40% ($P < 0.001$), and radioactive nest cells showed a higher mortality from parasites. The maximum integrated dose received by a developing wasp in this study was determined to be 10,630 rads. Continuing studies on radiocesium accumulation and elimination by various species of insects demonstrated the usefulness of these techniques for measuring food consumption and insect production under field conditions. Also, energy flow through the herbivorous insect population may now be estimated, by using food consumption rates for herbivores calculated with steady-state equilibria for radiocesium. Preliminary studies of radiation response of several insect species including crickets and grain beetles were reported.

Hematological studies of native mammals initiated in 1962 were completed this year. The objective of these studies was to determine the effects of endogenous and environmental factors on the blood of indigenous species. Over 10,000 hematological measurements were made for over 2000 mammals of 41 species belonging to 11 families representing 7 orders. These studies have shown that factors such as age, sex, certain diseases, certain parasites, injuries, confinement, and season govern hematopoietic changes. The largest and most consistent change in normal serum patterns is characteristically demonstrated by albumin. Significant alterations in this component have been observed as a seasonal variation and as a result of numerous stress factors. One seasonal variation has been a higher albumin value for the sera of animals sampled in the water than from the same animals sampled in other seasons. Laboratory-bred cotton rats had higher albumin percentages than field-trapped members of the same species. A coefficient of correlation of −0.360 between total white cell count and albumin-globulin ratio was calculated for the white-footed mouse ($0.05 > P > 0.02$). If subsequent calculations show this relationship to be a general one, it would imply that a low albumin in combination with a high total globulin value might well serve as readily as a high white blood cell count as an indication of general infection or trauma in an individual or a wild population.

10. Processes and Components of Terrestrial Ecosystems

An inventory of total ^{137}Cs distribution among the tree-soil compartments of the tagged tulip poplar ecosystem constructed from data collected during and at the end of the first growing season suggests that about 50% of the initial tag is found in the litter, soil, and smaller root components at the end of the first growing season. The maximum burden found in the canopy 4 to 6 weeks after inoculation was reduced by 60% in late September. This reduction is due to three processes: (1) export of cesium from foilage to woody tissue during

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mature and senescent stages of leaf development, which accounts for the greatest amount of reduction; (2) removal of radiocesium from foilage by rain and dew leaching prior to mid-September, which accounts for about 4% of the total reduction; and (3) reduction in concentration (μc per gram of dry weight) by the formation of new tissue. Accumulation of surface income of ^{137}Cs within the organic litter layer and surface 2 cm of soil was measured in sample units which prevented root entry. Results from these sample units indicate that half of the surface income was contained within the organic layer and more than 80% was contained within the organic layer plus the surface 2 cm of mineral soil. Dispersion of ^{137}Cs outside the tagged forest (20 × 25 m) area by falling leaves was measured at the end of the second growing season. Sampling measurements indicated that 2.67 mc (representing 5.34 μc per m^2 of study site) of ^{137}Cs was transported outside the study area by falling leaves. Based on this figure and on canopy concentrations in both years, it is estimated that 20% of the leaves from the tagged trees fell outside the study area. Of this 20% more than three-fourths of this activity is located within a 10-m zone around the perimeter of the study area. A new phase of work in the ^{137}Cs-tagged forest was initiated this year to broaden the understanding of the behavior of this radionuclide in the various life stages of tulip poplar trees. Seeds collected from tagged trees were germinated, and patterns of translocation were followed through the early growth phases. These studies seem to indicate that cesium follows similar pathways to sugars and other foods which may be stored in tissue such as cotyledons used to sustain the early growth of seedlings. Transfers of this radionuclide in young seedlings are also rapid, as has been demonstrated for mature trees of this species.

Microbial uptake of stable cesium and cobalt was proportional to their concentrations in the environment over a concentration range of 2 to 3 orders of magnitude. There was no marked isotopic selection for either of these elements. Uptake of ^{134}Cs was proportional to the concentration of ^{134}Cs in the substrate. When microbes were separated from a tagged substrate by water, agar, or semipermeable film, microbial uptake was proportional to the concentration of ^{134}Cs in these intermediates. Microbial concentration factors for different substrates varied with the availability of the tag. On fresh-leaf litter with readily available cesium, microbial concentration relative to the substrate was twice as high as on weathered, old litter with less readily available cesium.

11. Clinch River and Related Aquatic Studies

The series of analyses of stable strontium in white crappie (*Pomoxis annularis*) flesh and bone was completed, and it was possible to test the relation of the distribution of stable strontium and ^{90}Sr between the environment and the fish. The calcium and strontium concentrations in fish flesh and bone were relatively constant throughout the year with no seasonal variation and probably reflects the relative constancy of these elements in Clinch River water. Data on calcium and strontium concentrations in Clinch River water were published previously. There is a relatively good agreement between the specific activities in fish bone and water. These data suggest that for any constant release of ^{90}Sr to the river, it is possible to predict the consequent ^{90}Sr content of fish bone. The short biological half-life of strontium in fish flesh shows that measurements of ^{90}Sr in flesh will reflect the environmental concentrations at the time the fish were collected. The long biological half-life of strontium in fish bone suggests that ^{90}Sr concentrations in bone will reflect the average environmental concentrations during the lifetime of the fish. Studies on uptake and accumulation of strontium and calcium by algae and snails representing contrasting types of biogeochemical systems are reported. Uptake of calcium by the algae was found to be primarily a function of the media concentrations of calcium with a slight inverse influence by strontium in the media. A slight relative increase in calcium uptake at very low calcium concentrations indicates an increase in uptake efficiency at limiting levels of calcium and also suggests that calcium is required by the alga. Experiments also suggested that uptake is directly proportional to strontium in the media, with an enhancement in uptake at very low media concentrations of calcium, suggesting that strontium was used by the algae in place of calcium when calcium was in short supply. In snails the deposition of both calcium and strontium depends primarily on the respective concentrations of these elements in the immediate environment. The data also suggest

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that there is a simple substitution of strontium uptake for that of calcium by snail shell when environmental calcium is deficient. Strontium uptake increased with increasing environmental strontium concentrations and decreased with increasing environmental calcium concentrations. Increased environmental strontium depresses strontium uptake and increased environmental calcium enhances strontium uptake.

Previously, it was reported that there is an increase of newly occurring chromosomal aberrations in the natural population of *Chironomus tentans* from White Oak Creek. This population of Diptera, which is receiving a calculated dose of 230 rads/year of chronic environmental radiation, has been sampled annually since 1960. Two new inversions, each involving several bands, were observed once in the additional collection from the irradiated area. No new inversions were observed in the additional collections from the control populations. The finding of these two new inversions supports the previous conclusion that there is an increase of newly occurring aberrations in the White Oak Creek population. These newly occurring aberrations are eliminated by selection or genetic drift and are not maintained in the gene pool of the population. The increased mutation rate produced by the chronic irradiation in the White Oak Creek population has not reduced the amount of chromosomal polymorphism in this natural population.

12. Theoretical Systems

To provide a conceptual basis for understanding radiation effects and the fate of nuclides in ecosystems, analog computer solutions of competition equations, both with and without radiation stress, were generated. The equations investigated were the Lotka-Volterra system which has received major attention in classical population ecology. There are four possible cases of outcome between two competing Lotka-Volterra species: (1) s_1 or s_2, but not both, survives depending upon initial conditions; (2) s_1 survives to the exclusion of s_2 regardless of initial conditions; (3) s_2 only survives independent of initial conditions; and (4) both s_1 and s_2 coexist at equilibrium. For all four of these cases it was found that introducing radiation stress in the form of a rarefying term in the equations altered both transient and steady-state responses of the system. Where the final result was determined in the unstressed cases only by the degree of competition between the two species, under radiation stress their respective reproductive potentials became an important factor. In addition, it was found that effects of radiation tend to be minimized when interactions between the competing species are greater; the stronger the coupling between two competing systems, the less the effect of an outside perturbing influence.

Previous compartment models for transfer of substances in ecological systems are generalized in the form of a system of differential equations. The system is $\dot{\mathbf{V}} = \mathbf{VF}$, where $\dot{\mathbf{V}}$ is instantaneous change in the state vector $\mathbf{V}$, and $\mathbf{F}$ is a matrix of fractional transfer coefficients expressing the exchange of material between compartments. Estimations of $\mathbf{F}$ for a variety of real ecosystems are being made utilizing data from ecological and physiological sources to provide more realistic computer simulations.

PART III. RADIATION PHYSICS

13. Theoretical Radiation Physics

Distributions of dose and dose equivalent in anthropomorphic phantoms were computed for protons up to 400 Mev. A theory has been developed and a code written to give dose distributions in a proposed high-energy silicon dosimeter from isotropically incident protons of energies up to 400 Mev.

Dose distribution with linear energy transfer (LET) has been computed for a cylindrical anthropomorphic phantom irradiated with a beam of neutrons. A Monte Carlo code was used, and wide variations were found for the dose per unit LET at various points in the phantom for various neutron energies from thermal to 2.5 Mev.

A new fast-neutron spectrometer, the "Vectrometer," has been proposed. This device consists of two separated planar arrays of Geiger counters which measure the track lengths of recoil protons resulting from neutron irradiation of the hydrogen gas in the device. A simple expression yields the neutron energy from these track lengths. A prototype is under construction.

The simultaneous emission of bremsstrahlung and transition radiation from an electron-irradiated dielectric slab has been analyzed theoretically.

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It has been shown that proper measurements of these radiations may be analyzed to yield the mean square scattering angle of electrons in the material. The mean free path of hot electrons in a free-electron gas has been determined from many-body perturbation theory. Previous workers have neglected exchange scattering processes altogether. It is found that such processes may make an important difference in the calculated energy and angular distribution of the hot electrons after collision. The effect of surface losses upon the stopping power of charged particles has been analyzed using a semiclassical approach. The dispersion and damping of surface plasmons have been computed from a semiclassical theory. The results are presented as a function of the wave vector of the surface plasmon. In connection with work done by the Experimental Radiation Physics Group, computer codes have been written to evaluate the optical constants of materials from measured reflectivities and transmittivities of plane parallel foils made of these materials.

The analysis of the dispersion for a medium in which a linear proton beam interacts with a stationary ionized medium indicates that there is a multiple instability. Physical conditions under which a multiple instability may occur were studied, and various types of plasma beam systems were investigated with particular reference to the frequency, ranges, and rates of growth of the excited waves.

An analysis was made of a dispersion equation for a medium in which a helical electron beam interacts with a stationary plasma. The interaction was investigated with particular reference to superluminous, subluminous, and counterstreaming instabilities.

The interaction of an ion beam of small intensity moving with velocity $c\beta$ (where c is the velocity of light) through a stationary plasma was investigated under the assumption that the beam and the waves resulting from the interaction are aligned along the direction of an impressed static magnetic field and that the frequencies of the excited waves are sufficiently high so that the motion of the ions perturbed by these waves can be neglected.

A plasma-beam system is investigated in which an electron beam of small intensity moving with velocity βc interacts with a cold plasma immersed in a magnetic field. It is assumed that the beam and the excited wave resulting from the interaction are aligned along the direction of the magnetic field and that $(1 - \beta^2) \gg 1/\eta$, where η is the ion-to-electron mass ratio. The frequencies and growth rates of the excited waves are calculated.

14. Experimental Radiation Physics

The interaction of low-energy electrons with a number of benzene derivatives has been studied, using the swarm method. Control of the energy distribution in the electron swarm has been achieved, which allows discrimination between the different electron capture processes. The detailed study of the benzene derivatives aids in the understanding of the interaction of free electrons with more complicated organic molecules of great biological interest.

Improvements in the electron time-of-flight technique for electron diffusion measurements using a Geiger counter detector have been carried out by utilizing an empirical instrumental response characteristic to unscramble the drift time distributions observed in water vapor and ethylene. A new apparatus for drift measurements has been constructed and has undergone preliminary testing. This apparauts avoids problems associated with Geiger counter detectors by using an electron multiplier and differential pumping. A new method has been worked out and equipment has been constructed for determining oscillator strengths for electronic transitions in the vacuum ultraviolet region. This method uses pressure pulses of millisecond duration in the absorption cell and the pulsed light from a spark discharge source in order to avoid uncertainties in pressure and optical absorption associated with differential pumping. First application of the new method will be to determine oscillator strengths in argon.

An experiment is being developed which will determine the lifetime of temporary negative ions produced by unimolecular electron capture. The technique is designed to measure lifetimes from 10^{-9} to 10^{-6} sec and will be applied to the study of electron capture in water and other polar molecules. Monochromatic light from electron plasmon decay has been observed for the first time from aluminum foils a few hundred angstroms in thickness bombarded by 80-kev electrons. The sharp peak at 815 A determines the plasma energy precisely at 15.3 ev. The optical constants of evaporated silver films were determined between 2500 and 3700 A, the region of the Wood's

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transparency. Optical transmission curves derived from these constants showed a minimum whose depth was proportional to the projection of the E vector of the incident light on the foil normal, thus indicating a resonant coupling of the radiation with electronic motion across the foil. No optical constants in the vacuum ultraviolet spectral region are available for indium, so these were measured also for use in evaluation of plasmon decay experiments.

The Keplertron was used to study Auger electron production in infinite isotropic radioactive copper media. The continuous slowing-down model neglects Auger production, which therefore must be studied experimentally. Further calibration data for the instrument were taken, and an electron slowing-down spectrum in gold was measured for the first time.

15. Physics of Tissue Damage

The optical reflectivity of hot-pressed pyrolytic graphite was measured from 1100 to 3000 A, where no data were available. The optical constants showed that an electron resonance occurs at 6.9 ev, as predicted from energy loss experiments and as explained by Ichikawa as due to plasma excitation.

A new technique was developed for measuring charged-particle interactions in layers a few hundred atoms thick. These layers are vacuum evaporated on a thin silicon monoxide insulator and thus need not be self-supporting. Range measurements on protons from 2 to 10 kev were made with this method.

Further work on the time-of-flight electron beam monochromator resulted in a gun which produced a 2-nsec burst of electrons, an improved transistor amplifier, positive proof that the electron drift time is characteristic of its energy, and some preliminary observations of nitrogen vibrational excitation by 2-v electrons.

Vacuum diffusion pump oil was irradiated in the LITR and then used in the pump. A trace amount of the radioactive oil escaped into the vacuum system, where it deposited on the walls to a maximum depth of 6 A (less than a monomolecular layer). This technique should be useful in vacuum ultraviolet spectroscopy, where minute layers of oil change the optical properties of gratings, thin films, etc.

The so-called "gas effect" noted previously in thermoluminescence experiments was found to be a temperature effect with the gas playing the role of a heat transfer agent. Thus the doubt cast on previously obtained data is removed. Sample holders were fabricated from boron carbide since it is the only material we have found which is tissue equivalent, nonfluorescing, nonthermoluminescing, electrically and thermally conducting, structurally strong over a wide range of temperature, and capable of holding a high vacuum.

16. Professional Health Physics Training

A first-year graduate course, Applied Radiation Physics, was given at Vanderbilt University and the University of Tennessee for a total enrollment of 30 students. A course in Health Physics of 30 contact hr was given at ORSORT. Citizens of Pakistan, Austria, India, and Israel received training and experience with various sections of the Health Physics Division. Lectures were given at ORINS by a Health Physics Division staff member assigned to ORINS temporarily. Lectures and tours of the Laboratory were provided for personnel of the U.S. Public Health Service, the University of North Carolina, and the ORNL Orientation Program. Twenty holders of AEC Health Physics Fellowships completed a year of graduate training at Vanderbilt University and the University of Tennessee. Several fellowship holders elected to do their thesis work for the master's degree in the Health Physics Division at ORNL. A 1-year advanced graduate level course in radiation physics has been planned to be given at the University of Tennessee during the 1964–1965 academic year.

PART IV. RADIATION DOSIMETRY

17. Dosimetry Applications

The emphasis of the radiation dosimetry studies has been on the accumulation of human-exposure data, especially the dosimetry for the survivors of the nuclear bombings of Hiroshima and Nagasaki. Refinements were made in the "free-field" data and dose-distribution functions developed for typical Japanese houses. The equations were

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developed by a "multiple regression and correlation" fit to the extensive data from Operation BREN. Computed values for the "shielding factor" at a point in a Japanese house, using these equations, were within 6% of the experimental values from weapons tests in 50% of the cases.

18. Dosimetry Research

Other areas of investigation included small detector research, spectrometry, instrumentation, and dosimetry for radiobiological research. Fading characteristics of metaphosphate detectors were measured, advancements were made in several types of spectrometers, energy response functions were determined for dosimetry systems, and accurate dose-conversion factors were determined for PuBe and PoBe sources.

19. HPRR Operations and Special Projects

The Health Physics Research Reactor was used in a wide assortment of radiobiological, dosimetric, physical, and ecological studies with participating researchers from numerous laboratories and several countries. A feasibility study was completed for the construction of an intense 14-Mev neutron source for the BREN tower; construction began in June 1964.

PART V. INTERNAL DOSIMETRY

20. Internal Dose Estimation

The program of the Internal Dose Estimation Section continues in the study and improvement of methods for assessment of exposure due to radioactive materials that may enter the body. As in the past, data have been gathered and interpreted to serve as a basis for estimation of dose from a source within the body. Although animal data are not disregarded, the principal emphasis is on direct data on man.

The work reported this year includes a study of the retention of HTO by an employee exposed in the course of his work. The retention is closely approximated by the sum of two exponentials, one showing an elimination half-time of about 9 days and the other about 37 days. Although multiexponential retention functions have been observed in animals, this is the first direct observation of such a retention in man that has been reported; however, indirect evidence has been reported. This study involved members of the Health Physics Technology Section and Applied Health Physics Section, as well as members of the Internal Dose Estimation Section.

21. Stable Element Metabolism

Much useful data have been accumulated from the studies of trace elements in human tissues, diets, and excreta, although the newer studies involving diets and excreta are only partially complete at this time. Among other applications of these data, a distribution of stable strontium in the body has been studied and is found to differ rather significantly from the distribution of ^{226}Ra under chronic conditions of exposure. It is found that approximately 99% of the stable strontium is in the skeleton, whereas the skeleton burden of ^{226}Ra due to the natural background has been estimated to be only about 80% of the body burden.

22. A Two-Compartment Model with Random Variable Flows

Since estimates of dose depend upon a metabolic model, the Internal Dose Estimation Section must construct models for the metabolism of many substances and understand various factors that influence the retention of radionuclides in the body, particularly with a view to extrapolation from animals to man. Two such exploratory studies are reported here. A system of compartments has been constructed which are connected during periods of time distributed randomly, thus simulating bone sites that may be remodeling during various periods of time. Only the simplest of such systems, a two-compartment model, has been studied, and it is found that certain distributions of the time periods during which exchanges between compartments are permitted produce a power-function type of retention.

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23. Studies on the Internal Exposure/External Exposure Equivalence Relation

A second study is reported which suggests a possible method of relating effects due to an internal source with effects due to external radiation. In effect, this amounts to a method for interspecies extrapolation at low levels of exposure.

24. Comments on Intake Guides for Various Isotopes and Isotopic Mixtures of Uranium

In the application of maximum permissible concentration (MPC) values, or intake guides, for uranium as provided by NCRP and ICRP, questions arise frequently concerning a "curie" of natural uranium, which has a special definition in these recommendations. Other questions relate to the fact that limiting values for some of the isotopes are based on considerations of chemical toxicity, while for other isotopes it is the radiological hazard that is limiting; thus uncertainties in the computation of values for mixtures of the isotopes are encountered. In this study simple formulas, additional MPC values, and explanations are given as an aid in resolving these and other similar problems.

25. On the Estimation of a Systemic Body Burden of Plutonium

Two studies are reported which test, to a very rough degree, the validity of a computer program reported previously which purports to estimate the intake and body burden of ^{239}Pu on the basis of urinary excretion. The one study uses data on an employee who had a long record of exposure to ^{239}Pu. From the urinary excretion data, an intake pattern and retention are estimated and compared with autopsy estimates of the tissue burdens. The study suggests that the metabolic model is not inconsistent with the data, but certain parameters of the model may be rather different for this individual and this material from the values given in ICRP Publication 2. The second study uses data on dogs given Pu-nitrate by intravenous injection and by inhalation. The program seems to predict a qualitatively plausible pattern of intake to blood, and the estimates of lung deposition obtained by the code are within a factor of 2 of the estimates by the experimenters.

PART VI. HEALTH PHYSICS TECHNOLOGY

26. Aerosol Physics

Surface contamination measurement techniques, described in previous annual reports, have been used in a study of particle resuspension in a room. A tracer dust is dispersed and allowed to settle in an area simulating a small work room of average cleanliness. Contamination samples are collected by a person wearing coveralls and a respirator. The number of tracer particles found on the respirator filter is related to the number that would have been inhaled without a respirator. Three tests using 3.1-μ-diam ZnS (fluorescence) and one test using 2-μ CuO (neutron activation analysis) indicate that a person working in the room is likely to inhale 1 particle/hr for every 2900 (CuO) to 5600 (ZnS) particles per square meter of surface.

Agglomerates of submicroscopic thorium dioxide particles, produced by exploding thorium metal wires, were allowed to settle on a stainless steel surface. The settled aggregates were approximately 1 to 10 μ in diameter. Laboratory-scale studies show that these agglomerates can be redispersed by air jets, by mechanical abrasion, and by adhesion to contacting surfaces; however, the redispersion efficiencies are lower than those observed for 1.5- and 5-μ-diam solid ThO_2 particles.

The question of what happens physically just prior to, during, and after a particle is reentrained in an airstream is being studied with the use of a small wind tunnel. Particles adhering to test surfaces are exposed to a known flow structure of air in the wind tunnel. The dynamics of the particles moving on the surface are observed by microphotography using a pulsed light beam. Borosilicate glass beads (28.9 μ CMD; σ = 13%) on a glass surface display a variety of effects in an airstream of 5300 cm/sec average velocity (Re = 24,400). About half of the beads did not move during 2.5 sec of exposure, approximately one-third of the beads left the surface without having rolled, and the remainder moved along the surface in straight lines parallel to the direction of flow. Some of the beads that moved along the surface stopped, and some left the surface. There is an indication that the mechanisms involved in moving particles along the surface contributes a

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time-dependent factor to the redispersion of particles in an airstream.

Basic interactions between particles and surfaces, which determine the scale of forces required for resuspension, are being studied by applying known forces to remove particles from various surfaces. Borosilicate glass beads (28.9 μ, CMD), dispersed by settling on quartz surfaces, are subjected to known removal forces by vibrating the test surface. Attempts to remove the glass beads by applying 2.8×10^{-3} dyne of force normal to the surface were unsuccessful. This level of force was repeated for nonradioactive beads on friction-charged and on uncharged quartz. In a third experiment, glass beads were activated by thermal neutrons (~0.4 curie/g) and were dispersed on an uncharged quartz plate. After aging for 2.5 days, the test plate and radioactive particles were found to have a net positive charge of 7×10^{-10} coulomb/cm^2. Despite the presence or absence of electrostatic charge, there were no glass beads removed by the application of forces up to 90 times gravity.

Tests have continued in exploration of the capabilities of the exploding-wire aerosol generator to produce and disperse a wide variety of submicron aerosols. Well-dispersed gold aerosols with reproducible mass density can be produced by the electrical explosion of gold wires. The aerosol yield is observed to depend on the wire length, wire diameter, and voltage used for explosion. In other tests, gold and platinum wires were twisted together and exploded simultaneously by the electrical discharge. Particulate matter collected from the aerosol produced by such explosions consists of alloys of Au-Pt, giving some indication that metallic vapors were produced which mixed homogeneously.

27. Applied Internal Dosimetry

Emphasis of the ORNL whole-body counting program is on counting probable and potential exposure cases and on obtaining baseline counts on unexposed employees. During the past year, 1370 persons were counted for a total of 1568 human counts. Approximately 74% of the counts showed an essentially normal human spectrum. Several employees were counted for indications of ^{90}Sr-^{90}Y intake following three separate incidents of suspected exposure. One employee was estimated to have approximately 90% of the maximum permissible lung burden. Initial estimates in the other two incidents amounted to only 25 and 30% of the permissible total body burden; in both cases the estimates represent transient GI tract deposits which were eliminated rapidly. The computer program for processing whole-body count data has been made more flexible and can analyze data from about 75% of the routine counts. Computer plotting of in vivo spectra has been discontinued as not economical. Several different source-phantom configurations for calibrating the whole-body counter are being studied.

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Part V. Internal Dosimetry

W. S. Snyder

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20. Internal Dose Estimation

URINARY EXCRETION OF TRITIUM FOLLOWING EXPOSURE OF MAN TO HTO – A TWO-EXPONENTIAL MODEL

W. S. Snyder B. R. Fish
S. R. Bernard Mary R. Ford J. R. Muir

The metabolic course of HTO in the body may be considered to be relatively well understood. Numerous animal experiments supplemented by experiments on humans suggest that HTO introduced into the body becomes a part of the general pool of body fluids, and is eliminated from the body via the urine, perspiration, breath, and other avenues of elimination of water from the body. The experimental data prior to 1957 are summarized in the comprehensive review of the problem by Pinson and Langham.[1] In addition, there is a wealth of experience gained in working with tritium which has revealed other factors which influence the metabolism of HTO, in particular, age and seasonal variations.

Nevertheless, there are some data to suggest that the metabolic model which assumes HTO to enter and be nonpreferentially eliminated from the general pool of body water is not entirely accurate. Thompson[2] exposed mice and rats to HTO and observed that retention in a variety of tissues could be represented by a sum of two exponentials which were distinct from the single exponential representing the retention in body water. In addition to experimental work on mice which gave a multiexponential retention, Pinson and Langham report an experiment in which one human subject was exposed to tritium over a period of 8 months, and dried biopsy specimens of skin and fat taken 51 days after the end of the exposure showed a higher concentration of tritium than did urine at the time of the biopsy. Thus, there is the implication that in man, as in experimental animals, some fraction of the HTO entering the body is not eliminated at the rate characteristic of the elimination of water from the body, and that this fraction may be more closely incorporated in certain tissues of the body. Subsequent to the inhalation of HTO by a human subject, we have been able to measure the concentration of tritium in various body fluids and, in particular, have found that concentration in urine does follow a pattern described as a sum of two exponentials during 255 days following an intake.

The exposure occurred when an individual, working with a source of HTO in a hood, inhaled some of the material. The subject is a senior scientist, a male of age 41 years, weight 66.5 kg, height 161 cm, and apparently enjoying good health. He was aware of the general nature of the problem posed by HTO in the body, and we are indebted to him for his cooperation which made this extended study possible. After the first samples were evaluated, it was agreed that he would not attempt any drastic increase in his normal fluid intake, although he was encouraged to drink liberally.

During the first 11 days, samples of urine, blood serum, and sputum were obtained and analyzed for tritium. No significant difference in the levels found in these various body fluids was noted, and this is in line with measurements reported by Pinson and Langham.[1] Also, the data appeared to follow a single exponential as expected but indicated a half-time for elimination of something under

[1] E. A. Pinson and W. H. Langham, *J. Appl. Physiol.* **10**, 108–26 (1957).

[2] R. C. Thompson, *J. Biol. Chem.* **197**, 81–87 (1952); *op. cit.* **200**, 731–43 (1953).

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9 days. The data for the first 13 days are shown in Fig. 20.1. An early evaluation of the dose to be expected was made and indicated no apparent need for measures to hasten elimination. After discussion with the subject, it was agreed that he would not attempt to deliberately alter his normal habits for the purpose of hastening the elimination of the tritium.

At the time of writing, urine specimens have been collected periodically and analyzed during 255 days postexposure and have been analyzed by liquid scintillation counting. All counting times were for 30 min. All the urine data are shown in Fig. 20.2, and all the postexposure data are given in Table 20.1. During these 255 days, the subject has not been working directly with tritium. However, prior to this incident, he had shown urine concentrations in excess of the later values reported here. For example, the last sample taken before the incident was on October 25, just 3 days before the incident, and a concentration of 6.1×10^4 dis min^{-1} ml^{-1} was measured. It is possible that this earlier exposure may have some influence on the data.

From Fig. 20.2 it appears that two exponential terms are required to fit the urinary concentrations observed in this case. By the courtesy of Chester Richmond of LASL, we have obtained a least-squares fit of the data to a sum of two exponentials using the LASL code described in ref. 3 which does not require a logarithmic transformation of the concentrations. This fitting procedure yielded a formula for concentration in urine of

$$R(t) = 5.3 \times 10^6\, e^{-\alpha t} + 2.3 \times 10^4\, e^{-\beta t} ,$$

with

$$\alpha = \frac{0.693}{8.7} \quad \text{and} \quad \beta = \frac{0.693}{34} .$$

Cases of internal exposure at ORNL which warrant consideration of dose and intake on the basis of the characteristics of the individual are evaluated by an internal dose evaluation committee. This subject's total body water was estimated on the following bases:

1. On weight basis: for 66.5 kg of body weight the human contains 0.44 to 0.70 liter of water

[3]C. R. Richmond *et al.*, *Health Phys.* 10(1), 3–13 (1964).

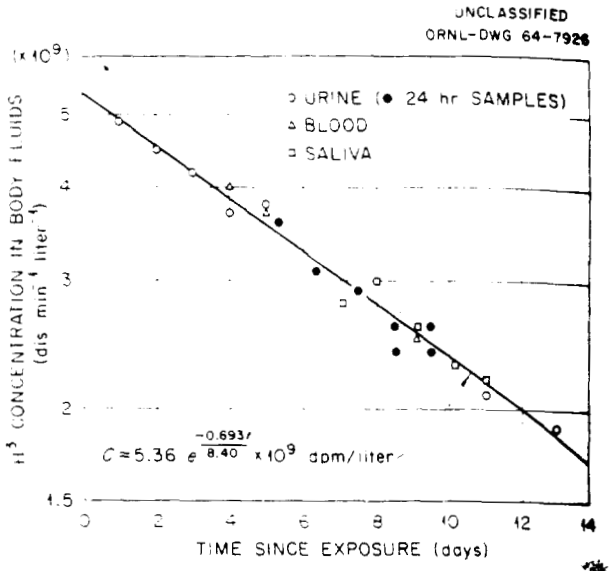

Fig. 20.1. Tritium Concentration in Body Fluids

per kilogram (the mean, 0.52 × 66.5 = 34.6 ≈ 35 liters).[4]

2. On age basis: humans 41 years of age con from 0.44 to 0.65 liter of water per kilogram mean, 0.525 × 66.5 = 34.9 ≈ 35 liters).[4]
3. On the basis of weight to height ratio: wei height = 0.41 kg/cm, the male human weight is 18 to 37% fat (mean 28%). L body weight (LBW) = (1 − 0.28) × 66.5 = kg.[5] The same reference suggests: total water = 0.72 LBW = 34.5 ≈ 35 liters.
4. On the basis of total potassium conte subject was counted in the ORNL Whole Counting Facility on November 8, 1963, the following results: age – 41; weight – kg; height – 161 cm; estimate 121 g of pot sium based upon ^{40}K measurement.

   Total potassium content = 0.28% of LBW; L 121 g/0.0028 = 43.2 kg.[6] This refere indicates that 72% of the lean weight and of the fat weight is made up of water;

[4]*Biological Handbook – Blood and Other Body Flu* Federation of American Societies for Experimental ogy, Washington, D.C., 1961.

[5]G. B. Forbes, J. Gallup, and J. B. Hursh, *Sc* 133, 101–2 (Jan. 13, 1961).

[6]E. C. Anderson, *Brit. J. Radiol. Suppl.* 7, 27 (19

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total body water = 0.72 × 43.2 + 0.2 × 23.3 = 35.76 ≈ 36 liters.

The Committee decided to use 36 liters as the amount of body water. Assuming a concentration of 5.3×10^6 dis min^{-1} ml^{-1} initially and that all the tritium is in the body water at early times post-exposure, this yields $5.3 \times 10^6 \times 36 \times 10^3 = 1.9 \times 10^{11}$ dis/min, or 86 mc, as the initial intake. The total dose to body water is then

$$D = \frac{51 \times t_{\frac{1}{2}} \varepsilon q(0)}{m \times 5.32 \times 10^6} \int_0^\infty dt\, R(t) = 15.5 \text{ rems},$$

where $t_{\frac{1}{2}} = 1$, $m = 3.6 \times 10^4$ g, $\varepsilon = 0.01$ Mev, $q(0) = 86 \times 10^3$ μc, and

$$\int_0^\infty R(t)\, dt = \frac{5.32 \times 10^6 \times 8.7}{0.693} + \frac{2.3 \times 10^4 \times 34}{0.693}$$

$$= 6.68 \times 10^7 + 1.13 \times 10^6 = 6.79 \times 10^7 .$$

It is apparent that the second exponential term contributes only about 1.7% to the total dose. The dose in various body tissues may be somewhat less

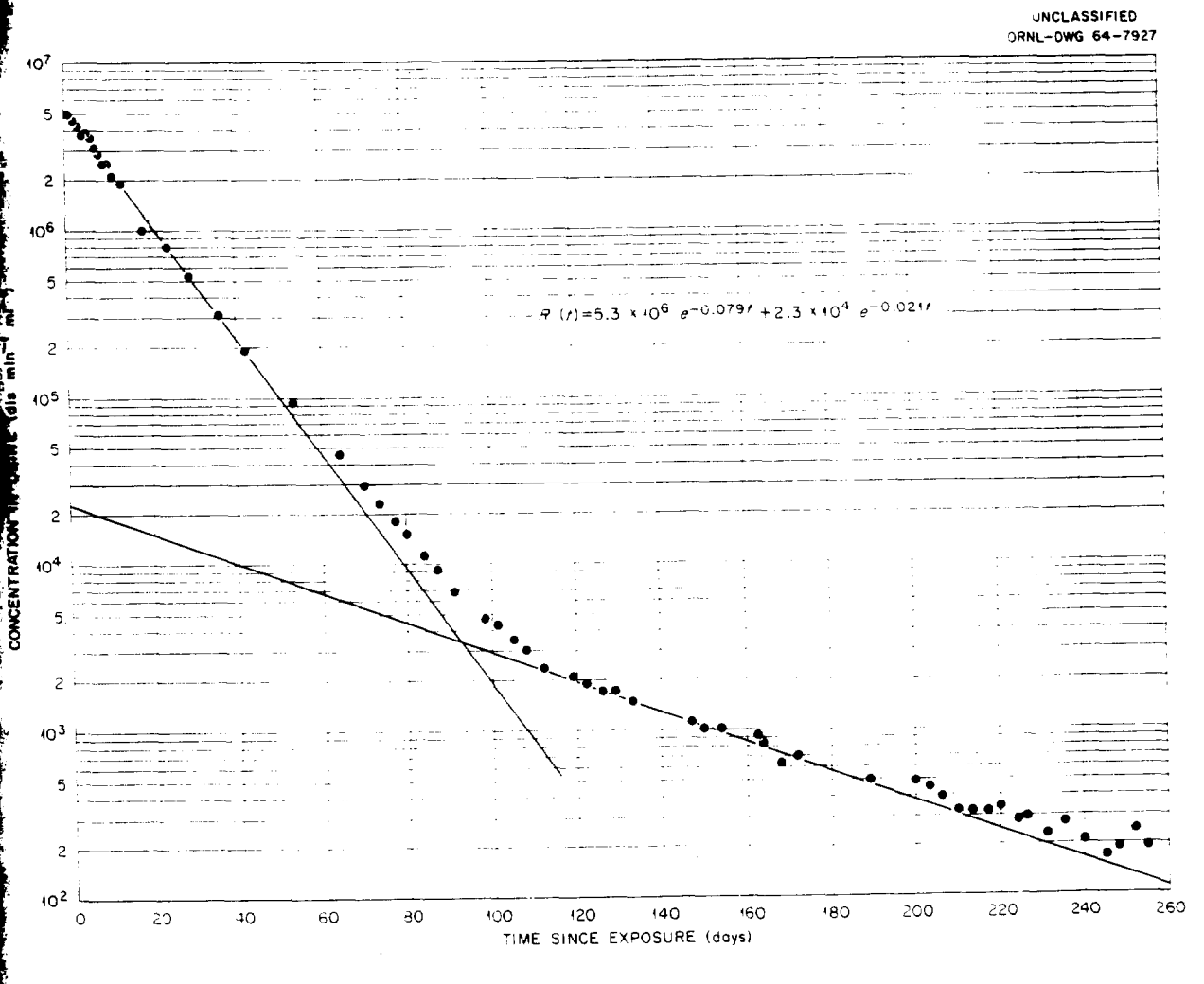

Fig. 20.2. Tritium Concentration in Urine.

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since no allowance has been made for the presence of organic material. However, there are some tissues in which water constitutes a preponderant part of the mass, and so the estimate is substantially, although not precisely, correct for such tissues. On the other hand, there remains the probability that some tissue may accumulate a higher concentration than body water, particularly at times much later than the exposure. However, the contribution of the second exponential term to the total dose is very small, and unless some tissue concentrates the material to a rather high degree, it seems unlikely that the dose would be significantly in excess of the dose to body water.

Table 20.1. Concentration of Tritium in Body Fluids

Time After Exposure (days)	Concentration (dis min^{-1} ml^{-1})[a]		
	Urine	Blood Serum	Sputum
1	$4.9 \times 10^6 \pm 5.1 \times 10^3$		
2	$4.5 \times 10^6 \pm 5.1 \times 10^3$		
3	$4.2 \times 10^6 \pm 3.5 \times 10^3$		
4	$3.7 \times 10^6 \pm 4.6 \times 10^3$	$4.0 \times 10^6 \pm 3.4 \times 10^3$	$3.3 \times 10^6 \pm 4.4 \times 10^3$
5	$3.8 \times 10^6 \pm 3.4 \times 10^3$	$3.7 \times 10^6 \pm 3.3 \times 10^3$	
6[b]	$3.6 \times 10^6 \pm 3.3 \times 10^3$		
7[b]	$3.1 \times 10^6 \pm 3.0 \times 10^3$		$2.8 \times 10^6 \pm 2.9 \times 10^3$
8[b]	$2.9 \times 10^6 \pm 2.9 \times 10^3$		$3.0 \times 10^6 \pm 3.1 \times 10^3$
9[b]	$2.6 \times 10^6 \pm 2.8 \times 10^3$	$2.5 \times 10^6 \pm 1.5 \times 10^3$	$2.6 \times 10^6 \pm 8.8 \times 10^2$
9	$2.4 \times 10^6 \pm 2.7 \times 10^3$		
10[b]	$2.6 \times 10^6 \pm 2.8 \times 10^3$		$2.3 \times 10^6 \pm 3.8 \times 10^3$
10	$2.4 \times 10^6 \pm 3.9 \times 10^3$		
11	$2.1 \times 10^6 \pm 3.6 \times 10^3$		$2.2 \times 10^6 \pm 3.9 \times 10^3$
13	$1.9 \times 10^6 \pm 3.4 \times 10^3$		
18	$1.0 \times 10^6 \pm 2.0 \times 10^3$		
24	$7.9 \times 10^5 \pm 1.8 \times 10^3$		
29	$5.4 \times 10^5 \pm 1.0 \times 10^3$		
36	$3.2 \times 10^5 \pm 7.8 \times 10^2$		
42	$1.9 \times 10^5 \pm 6.2 \times 10^2$		
53	$9.4 \times 10^4 \pm 3.8 \times 10^2$		
64	$4.6 \times 10^4 \pm 2.8 \times 10^2$		
70	$3.0 \times 10^4 \pm 2.2 \times 10^2$		
73	$2.3 \times 10^4 \pm 2.0 \times 10^2$		
77	$1.8 \times 10^4 \pm 1.8 \times 10^2$		
80	$1.5 \times 10^4 \pm 1.7 \times 10^2$		
84	$1.1 \times 10^4 \pm 1.5 \times 10^2$		
87	$9.2 \times 10^3 \pm 1.7 \times 10^2$		
91	$6.7 \times 10^3 \pm 1.3 \times 10^2$		
98	$4.6 \times 10^3 \pm 1.3 \times 10^2$		
101	$4.2 \times 10^3 \pm 92$		
105	$3.5 \times 10^3 \pm 86$		
108	$3.0 \times 10^3 \pm 82$		$3.0 \times 10^3 \pm 47$
112	$2.3 \times 10^3 \pm 71$		
115	$2.2 \times 10^3 \pm 75$		
119	$2.1 \times 10^3 \pm 73$		
122	$1.9 \times 10^3 \pm 74$		
126	$1.7 \times 10^3 \pm 59$		

[a]Me
[b]24

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Table 20.1 (continued)

Time After Exposure (days)	Concentration (dis min^{-1} ml^{-1})[a]		
	Urine	Blood Serum	Sputum
129	$1.7 \times 10^3 \pm 58$		
133	$1.5 \times 10^3 \pm 56$		
147	$1.1 \times 10^3 \pm 47$		
150	$1.0 \times 10^3 \pm 46$		
154	$1.0 \times 10^3 \pm 49$		
163	$9.2 \times 10^2 \pm 30$		
164	$8.3 \times 10^2 \pm 29$		
168	$6.3 \times 10^2 \pm 28$		
172	$6.9 \times 10^2 \pm 28$		
189	$4.9 \times 10^2 \pm 24$		
200	$4.8 \times 10^2 \pm 26$		
203	$4.4 \times 10^2 \pm 25$		
206	$3.8 \times 10^2 \pm 24$		
210	$3.2 \times 10^2 \pm 23$		
213	$3.2 \times 10^2 \pm 23$		
217	$3.2 \times 10^2 \pm 24$		
220	$3.4 \times 10^2 \pm 25$		
224	$2.8 \times 10^2 \pm 23$		
226	$2.9 \times 10^2 \pm 25$		
231	$2.3 \times 10^2 \pm 23$		
235	$2.7 \times 10^2 \pm 22$		
240	$2.1 \times 10^2 \pm 17$		
245	$1.7 \times 10^2 \pm 17$		
248	$1.9 \times 10^2 \pm 16$		
252	$2.4 \times 10^2 \pm 16$		
255	$1.9 \times 10^2 \pm 17$		

[a]Mean disintegration rate ± one standard deviation.
[b]24-hr samples; all others are spot samples.

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21. Stable Element Metabolism

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Mary Jane Cook
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Because a radionuclide follows the metabolic pathway of a stable element, one may predict the distribution pattern of a radionuclide under chronic conditions from the analysis of grossly normal human tissues for stable elements. Allowing for radioactive decay, the concentration of stable elements indicates the tissues where the corresponding radionuclide would deposit under chronic exposure and thus indicates the dose that tissues might receive under chronic exposure. Due to the heterogeneity of the human population, a sufficiently large sample is needed in order to permit an estimate of the variability of dose that might be received, and to correlate this variability with age, sex, occupation, geographical location, etc. Such dose estimates are essential because efficient use of atomic energy requires accurate estimates of maximum permissible concentrations (MPC) of radionuclides in air, in water, and in food, not only for atomic energy workers but also for the general population which includes babies, children, teen-agers, etc.

Under chronic conditions inorganic elements are taken into the body through food, water, and air, but little attention has been devoted to the amount of daily intake of the majority of trace elements. Also, very little is known about the quantities excreted in the urine or in the feces. In applied health physics programs, samples of urine of radiation workers are collected and analyzed routinely to determine the presence of various radionuclides to which the worker may have been exposed. The interpretation of such data requires a metabolic model for the handling of such substances by the body, and trace element analysis of tissues supplies evidence bearing directly on major sites for deposition of the material and indirectly on various parameters of the metabolic model. Such metabolic models are the basis for estimation of daily intakes that might be expected to produce a specified body burden of a radionuclide. Thus, studies of this kind are one of the major sources of data utilized for control of exposure to environmental contamination by radioactive material.

TISSUE ANALYSIS LABORATORY PROGRESS REPORT

Cyrus Feldman F. S. Jones

Spectrographic analysis of samples of hum tissue collected in earlier years was continued using the previously described procedures.[3] A proximately 430 soft tissues were analyzed 23 stable elements. These tissue samples predominantly the smaller organs such as thy gall bladder, and ovary, and small tissue sa from the larger organs such as kidney.

To test the possibility of analyzing tis for thorium, three tissues were analyzed by ra activation analysis. Equal portions of ash several kidney samples from Atlanta were and duplicate analyses were made. This procedure was followed for other kidney and lung samples. The analytical resul the mean values are presented in Table

For the past several months, Consumers U has been analyzing for ^{90}Sr, ^{89}Sr, Ca, and in teen-age diets collected from several citi the United States.[4] Because the elemental position of the diet is of importance in calcul MPC values and very little such data are avail samples of these diets were obtained from sumers Union in order to analyze them for

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[3]S. R. Koirtyohann and C. Feldman, "The graphic Determination of Trace Elements in Tissue," *Analytical Chemistry in Nuclear Technology, Fourth Conference, Gatlinburg, Te Oct. 12–14, 1960*, TID-7606, pp. 51–63.

[4]Irving Michelson, *Health Phys.* 9, 944–50 (19

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Table 21.1. Results of Radioactivation Analyses for Thorium in Human Tissues[a]

Samples	Micrograms of Th per gram of Ash		
	1st Analysis	2d Analysis	Mean
Kidney, Atlanta	0.10	0.096	
Kidney, San Francisco	0.06	0.07	0.073
Kidney, New York	0.06	0.05	
Bone, Atlanta	0.07	0.07	
Bone, San Francisco	0.05	0.06	0.067
Bone, New York	0.08	0.07	
Lung, Atlanta	0.12	0.12	
Lung, San Francisco	0.12		0.114
Lung, New York	0.10	0.11	

[a]Analyses performed by E. I. Wyatt.

ce elements. To date, 75 of these diet samples ve been analyzed. Some preliminary results are esented in Table 21.2. These data are being mpared with similar data on diets of adults as orted by other investigators and are being died in relation to questions of population posure to a variety of radionuclides.

In order to speed the analytical procedure and o eliminate the human errors of handling data, n automatic data-acquisition computer system as installed. The data output from the Quantom-er consists of recorder deflections which, when mpared with standards of known composition, are nverted and printed out in parts per million f ash. This was done previously by plotting urves on transparent overlays and interpreting the recorder chart deflections.

TRACE ELEMENTS IN HUMAN TISSUE: RIB AND VERTEBRA

Isabel H. Tipton Jane J. Shafer

Since the gross composition of bone is quite different from that of soft tissue, a different set of standards and a somewhat different method are required in the spectrographic analysis of bone. For this reason, in the program for spectrographic analysis of human tissue, all the samples of bone which were collected from individuals both from within and outside the United States were held for analysis as a group. This group of bone samples included rib, vertebra, sternum (only from Dallas), and unidentified bone. For this report, only the data for rib and vertebra are included. A summary of the total number of bone samples which were analyzed and the geographical distribution of these samples is given in Table 21.3.

The bone samples were analyzed in the spectrographic laboratory of the Analytical Chemistry Division of ORNL by methods which have been described.[5] Data from these analyses were processed at the University of Tennessee Computing Center using nonparametric methods previously described.[6] The Kruskal-Wallis test for two-way classification was used to test the hypothesis that two groups were from the same population.[7]

Descriptive statistics for rib and vertebra of adults from the United States are given in Tables 21.4 and 21.5 respectively. For the ash percent of wet, dry percent of wet, and ash percent of dry, the column "Fraction Observed" means the number of samples for which the parameter was calculated, divided by the total number of samples. For an

[5]W. S. Snyder *et al.*, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, p. 178.

[6]I. H. Tipton and J. J. Shafer, *Arch. Environ. Health* 8, 58 (1964).

[7]W. H. Kruskal and W. A. Wallis, *Am. Statist. Assn. J.* 47(260), 584 (1952).

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Table 21.2. Preliminary Results of the Spectrographic Analysis of Teen-Age Diet Samples

(The values are the means of the number of determinations in parentheses. The numbers in brackets indicate the lower limit of detection.)

City	% Dry	% Ash	Al (ppm)	Ba (ppm)	Cu (ppm)	Mn (ppm)	Pb (ppm)	Sn (ppm)	Sr (ppm)	Ti (ppm)	Zn (ppm)	C (%)	Ca (%)	Fe (%)	K (%)	Mg (%)	Na (%)	P (%)	S (%)	Si (%)
Chicago, Ill.	18.8 (3)	0.86 (3)	297 (3)	33 (3)	56 (3)	98 (3)		380 (3)	61 (3)	21 (2)	557 (3)	54.6 (3)	6.14 (3)	0.11 (3)	15.5 (3)	1.41 (3)	17.9 (3)	7.76 (3)	0.19 (3)	0.23 (3)
Duluth, Minn.	19.4 (3)	0.79 (3)	540 (4)	28 (4)	66 (4)	113 (4)	20 (3)	144 (4)	76 (4)		455 (4)	53.8 (3)	6.36 (3)	0.09 (3)	18.1 (3)	1.42 (3)	16.3 (3)	7.93 (3)	0.18 (3)	0.57 (3)
Knoxville, Tenn.	21.1 (4)	0.90 (4)	338 (4)	35 (4)	59 (4)	123 (4)		333 (4)	60 (4)	184 (2)	445 (2)	53.8 (4)	6.82 (4)	0.06 (4)	15.1 (4)	1.39 (4)	16.9 (4)	8.41 (4)	0.16 (4)	0.38 (4)
Little Rock, Ark.	17.0 (2)	0.75 (2)	665 (2)	45 (2)	46 (2)	145 (2)	[10] (1)	605 (2)	65 (2)	15 (1)	560 (4)	52.5 (2)	8.83 (2)	0.06 (2)	16.4 (2)	1.38 (2)	14.9 (2)	8.89 (2)	0.18 (2)	1.14 (2)
Los Angeles, Calif.	18.1 (3)	0.77 (3)	230 (3)	38 (3)	187 (3)	111 (3)	23 (3)	383 (3)	143 (3)	14 (2)	660 (3)	55.1 (2)	6.26 (2)	0.08 (2)	17.9 (2)	1.61 (2)	15.0 (2)	8.51 (2)	0.21 (2)	0.50 (2)
New Orleans, La.	15.8 (4)	0.71 (4)	470 (4)	54 (4)	109 (4)	143 (4)	[10] (1)	855 (4)	92 (4)	38 (3)	545 (4)	51.8 (4)	8.01 (4)	0.07 (4)	15.2 (4)	1.42 (4)	16.4 (4)	8.27 (4)	0.18 (4)	0.52 (4)
New York, N.Y.	18.4 (2)	0.86 (2)	500 (2)	29 (2)	85 (2)	125 (2)		370 (2)	76 (2)	114 (2)	415 (2)	54.1 (2)	6.47 (2)	0.06 (2)	15.4 (2)	1.36 (2)	17.5 (2)	7.64 (2)	0.19 (2)	0.73 (2)
Portland, Ore.	19.0 (4)	0.86 (4)	355 (4)	49 (4)	62 (4)	145 (4)		498 (4)	86 (4)	17 (3)	455 (4)	52.4 (4)	6.46 (4)	0.08 (4)	15.5 (4)	1.55 (4)	17.3 (4)	8.01 (4)	0.18 (4)	0.47 (4)
San Francisco, Calif.	20.8 (4)	0.88 (4)	228 (4)	53 (4)	70 (4)	123 (4)		403 (4)	95 (4)	161 (4)	965 (4)	53.8 (4)	6.92 (4)	0.08 (4)	15.8 (4)	1.51 (4)	17.0 (4)	8.13 (4)	0.18 (4)	0.64 (4)
Sioux Falls, Ida.	19.8 (4)	0.86 (4)	710 (4)	144 (4)	60 (4)	108 (4)	[10] (1)	463 (4)	92 (4)	98 (2)	443 (4)	53.8 (4)	6.83 (4)	0.08 (4)	14.9 (4)	1.45 (4)	17.9 (4)	7.87 (4)	0.18 (4)	0.2 (4)

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ement, the column "Fraction Observed" means e number of samples in which the element oc- urred in a concentration above the lower limit f detection (and was thus actually observed), ivided by the total number of samples. For those lements which were not observed in concentrations bove the lower limit of detection in every sample, t was impossible to calculate a mean value for oncentration, since it could only be assumed that the concentration of such an element lay somewhere between zero and the lower limit of detection. Therefore, the two limiting values of the mean were calculated for these elements, one assigning to those samples in which the element was not observed a value for the concentration equal to zero, and the other assigning a value equal to the lower limit of detection. These limiting values for the mean are given in the last

Table 21.3. Number of Samples of Rib and Vertebra

All Adult Subjects ≥ 20 Years	Total Number of Subjects	Rib	Vertebra
United States	150	91	53
Colorado, Denver		9	8
Illinois, Chicago		1	
Florida, Miami		19	1
Maryland, Baltimore		23	22
New York, New York			
Texas, Dallas			
Virginia, Richmond		22	20
Washington, Seattle		17	2
African Continent	54	1	
Near East	37	19	
India, Bombay		8	
India, Delhi		7	
India, Lucknow		4	
India, Vellore			
Lebanon, Beirut			
Far East	70	52	10
Formosa, Taipei		4	
Hong Kong		10	
Japan, Chiba		6	9
Japan, Kyoto		5	
Japan, Tokyo		8	
Philippine Islands, Manila		9	1
Thailand, Bangkok		10	
Switzerland, Bern	9	2	2
Alaska, Anchorage	2		
Hawaii, Honolulu	12	9	4
Males 20–59 Years			
United States	88	56	
African Continent	1	1	
Near East	24	14	
Far East	39	27	
Females 20–59 Years			
United States	30	16	

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Table 21.3. (continued)

Children 0—< 20 Years			
United States	23	11	7
Colorado, Denver		2	4
Illinois, Chicago			
Florida, Miami		3	
Maryland, Baltimore		2	1
New York, New York			
Texas, Dallas			
Virginia, Richmond		2	2
Washington, Seattle		2	
African Continent	5		
Near East	9	4	
India, Bombay		1	
India, Delhi		2	
India, Lucknow		1	
India, Vellore			
Lebanon, Beirut			
Far East	3	3	
Formosa, Taipei			
Hong Kong			
Japan, Chiba			
Japan, Kyoto			
Japan, Tokyo		2	
Philippine Islands, Manila		1	
Thailand, Bangkok			
Switzerland, Bern			
Alaska, Anchorage			
Hawaii, Honolulu			

column of the table as "Limits of Sample Mean." In some instances, when the element was observed in all but a few samples, (e.g., aluminum in vertebra), the difference between these two values for the mean lies in the second or third digit. The precision of the analytical method at this level is in general only good to one digit, and thus only one digit is reported; however, in such cases the two estimates of the mean do not differ significantly. The lower limit of concentration of those elements which were not observed in every sample is given as less than the lower limit of detection (< limit of detection). When an element was not observed in at least half the samples, the median value for concentration of that element is given in the tables as less than the lower li of detection. Cadmium and molybdenum w observed in only one sample of rib, and th elements and gold were not observed in an the vertebrae.

On the basis of the Kruskal-Wallis test, median values for the concentration in as iron, sodium, and potassium in vertebra significantly ($p \leq 0.01$) higher than the me values for the concentration in ash of the elements in rib. On the other hand, the me value for the concentration of calcium in as significantly higher in rib than in vertebra 21.1). The median value for concentratio strontium was higher ($p = 0.02$) and of lea

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Ash %
Dry %
Ash %

Calciu
Iron
Magne
Potass
Sodium

Lead
Strontiu
Zinc
Alumin
Cadmiu
Chromi
Cobalt
Copper
Gold
Mangan
Molybde
Nickel
Tin

[a]See tex

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er ($p = 0.05$) in rib than in vertebra. The ash cent of wet weight also was significantly higher rib than in vertebra (Fig. 21.1). This reflects difference in texture of the two types of wet e, rib being compact and vertebra porous with interstices filled with blood and "soft-type" sue which ashes with a low residue and which also high in iron, sodium, and potassium.

Since the concentration of certain elements in and vertebra showed a correlation with age, d since the concentrations of several of these ements in rib were also significantly different adult male and female subjects (Fig. 21.2), ly those values for males between the ages 20 and 59 were used in the two-sample tests r geographic differences. In general, only rib was collected from the foreign group, and so geographical comparisons have been made only in rib. (There were not enough samples of bone from African and Swiss subjects to consider them as subgroups in performing these tests.)

Geographic differences in the concentration in ash of calcium, strontium, and lead are shown in Fig. 21.3. The poor nutritional status of the Near Eastern subgroup is reflected in the significantly lower median value for the concentration of calcium in rib. The median value for strontium in rib in the Far Eastern subgroup was significantly higher than for either of the other two groups. Lead was in significantly higher concentration in the subgroup from the United States than in either of the other subgroups. In the Far Eastern subgroup this

Table 21.4. Concentrations of Elements in Bone – Rib for Adult Subjects from the United States

	Fraction Observed[a]	Total Range		Median	95% Confidence Interval for Median		Mean	Std Dev	Limits of Sample Mean[a]	
Ash % of Wet	91/91	7.6	50.	24.	22.	25.	24.	8.5		
Dry % of Wet	52/91	48.	79.	65.	62.	67.	63.	7.9		
Ash % of Dry	52/91	16.	56.	37.	34.	39.	36.	8,6		
				Grams per 100 Grams of Tissue Ash						
Calcium	91/91	29.	39.	36.	36.	36.	36.	3.8		
Iron	91/91	0.01	0.16	0.04	0.03	0.05	0.04	0.03		
Magnesium	91/91	0.25	0.59	0.46	0.45	0.48	0.46	0.10		
Potassium	91/91	0.29	2.6	0.62	0.56	0.72	0.70	0.29		
Sodium	91/91	0.9	4.4	1.3	1.3	1.5	1.5	0.57		
				Micrograms per Gram of Tissue Ash						
Lead	91/91	10.	140.	43.	41.	52.	47.	21.		
Strontium	91/91	38.	360.	110.	110.	120.	120.	49.		
Zinc	91/91	100.	450.	200.	190.	210.	210.	46.		
Aluminum	85/91	<5.	110.	10.	5.	10.			12.	13.
Cadmium	1/91	<5.	5.	<5.	<5.	<5.			0.	5.
Chromium	3/91	<2.	20.	<2.	<2.	<2.			0.	2.
Cobalt	4/91	<2.	21.	<2.	<2.	<2.			0.	2.
Copper	82/91	<2.	35.	3.	2.	3.			3.	4.
Gold	0/91	<2.	<2.	<2.	<2.	<2.			0.	2.
Manganese	87/91	<2.	10.	2.	2.	2.			2.	2.
Molybdenum	1/91	<2.	2.	<2.	<2.	<2.			0.	2.
Nickel	21/91	<2.	69.	<2.	<2.	<2.			2.	4.
Tin	7/91	<5.	20.	<5.	<5.	<5.			0.	5.

[a]See text for explanation.

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Table 21.5. Concentrations of Elements in Bone – Vertebra for Adult Subjects from the United States

	Fraction Observed[a]	Total Range		Median	95% Confidence Interval for Median		Mean	Std Dev	Limits of Sample Mean[a]	
Ash % of Wet	53/53	7.5	27.	15.	14.	16.	15.	3.4		
Dry % of Wet	34/53	40.	62.	51.	49.	53.	51.	6.4		
Ash % of Dry	34/53	15.	41.	29.	26.	31.	29.	5.7		
Grams per 100 Grams of Tissue Ash										
Calcium	53/53	32.	37.	35.	35.	35.	35.	2.9		
Iron	53/53	0.02	0.40	0.09	0.07	0.11	0.10	0.07		
Magnesium	53/53	0.30	0.62	0.51	0.49	0.54	0.50	0.07		
Potassium	53/53	0.50	2.7	0.99	0.91	1.1	1.0	0.29		
Sodium	53/53	1.2	3.9	2.0	1.7	2.3	2.0	0.44		
Micrograms per Gram of Tissue Ash										
Lead	53/53	5.	130.	51.	45.	56.	54.	21.		
Strontium	53/53	58.	200.	100.	90.	110.	110.	34.		
Zinc	53/53	94.	340.	220.	210.	250.	220.	45.		
Aluminum	47/53	<5.	78.	5.	5.	10.			9.	9.
Cadmium	0/53	<5.	<5.	<5.	<5.	<5.			0.	5.
Chromium	1/53	<2.	5.	<2.	<2.	<2.			0.	2.
Cobalt	1/53	<2.	15.	<2.	<2.	<2.			0.	2.
Copper	52/53	<2.	10.	5.	2.	5.			4.	4.
Gold	0/52	<2.	<2.	<2.	<2.	<2.			0.	2.
Manganese	51/53	<2.	10.	2.	2.	3.			2.	2.
Molybdenum	0/53	<2.	<2.	<2.	<2.	<2.			0.	2.
Nickel	11/53	<2.	43.	<2.	<2.	<2.			2.	3.
Tin	7/53	<5.	20.	<5.	<5.	<5.			1.	5.

[a]See text for explanation.

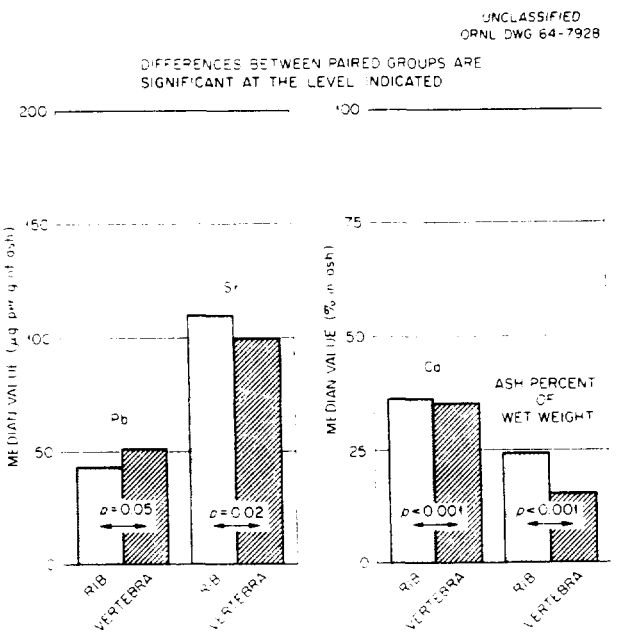

Fig. 21.1. Differences Between Rib and Vertebra, Adults ≧ 20 yr.

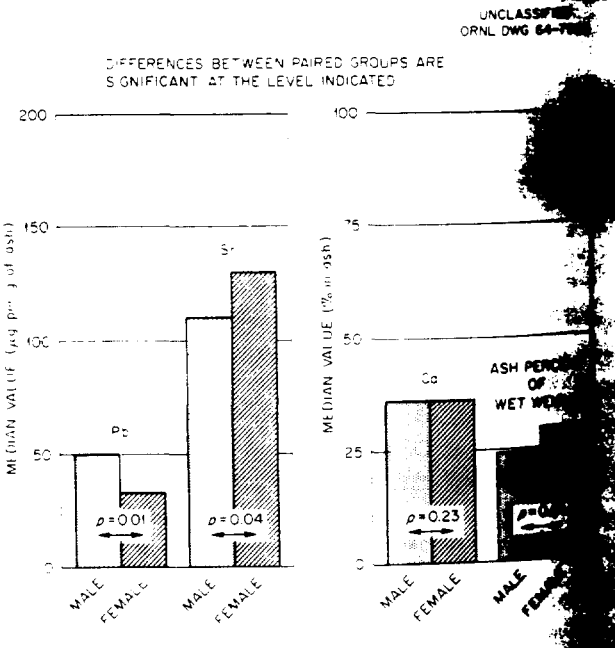

Fig. 21.2. Rib – Sex Differences, U.S. Adults 59 yr.

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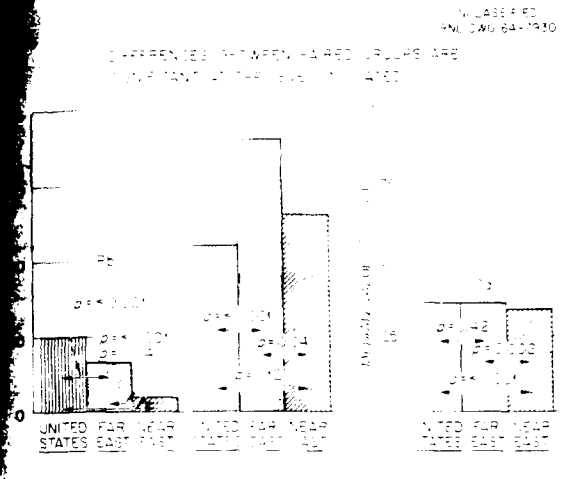

Fig. 21.3. Rib – Geographic Differences, Males 20–yr.

lement was significantly higher than in the Near astern. This may reflect the differences in posure to automobile exhaust fumes of these ree subgroups, since at the time of collection these samples, lead was a common additive in ost gasolines.

TRACE ELEMENTS IN DIETS AND EXCRETA – PRELIMINARY STUDY

Isabel H. Tipton Peggy L. Stewart

To obtain some idea of the average amounts and day-to-day variation of trace elements in ordinary diets, two subjects for a month saved duplicate samples of all food ingested and all excreta. The subjects were husband and wife, ages 35 and 34, respectively, who for several years have maintained weights of ~165 and 136 lb respectively.

All fluids except water, tea, and coffee were included in the daily samples of food. A record was kept of the daily intake of water, tea, and coffee, and samples of each also were supplied for analysis.

Complete 24-hr samples of urine were collected, and analyses were made on these daily samples. Feces were composited for a 3-day period since the chances of no fecal output during a 24-hr period are high.

The average daily intake of food and output of urine and feces are shown in Table 21.6. The total intake was arrived at by adding the weight of food to the weight of drink (assuming a specific gravity of 1.0). The amount of water in the food samples was determined by distillation with toluene. Thus, it was possible to calculate the total intake of water.

The average daily intake and output of trace elements as shown in the table seem to indicate a positive balance for most elements. In some instances this is due to a limitation of the spectrographic method which was set up for the analysis of carefully controlled diets in a program where the range of variation for most elements was not very great. In the *ad libitum* diets the range was very great, and in many cases the measurements were made at and extrapolated beyond the upper limits of the method where exact values were difficult to obtain. This was especially true for the feces which had extremely high concentrations of trace elements. In urine the reverse was true – concentrations were so low that many values were uncertain. There is also the possibility that some of the excretion might be via perspiration, exhalation, etc.

For some elements (e.g., zinc, silver, and nickel) the concentration in food was too low to be detected by the spectrographic method employed in the study. More sensitive methods are being developed. The concentrations of these elements in feces (and of nickel in urine), however, were in good measurable range. For this preliminary report, the average daily intake of these elements has been assumed to be equal to the output.

The figures show the daily amounts of calcium (Fig. 21.4) and strontium (Fig. 21.5) ingested and excreted in the feces and urine by each of the subjects. Although the variation in intake is wide, the amounts vary around a mean which remains fairly constant with time.

The daily intake of stable strontium in these subjects was about twice that generally reported, but the mean intake of calcium was 1.1 g/day, which agrees with the generally accepted value of 1 g/day. Thus, the ratio of strontium to calcium in their diets was somewhat higher than the average values by other investigators.[8] As indicated, this report is preliminary, and the data and analysis are not complete at this time.

[8] W. S. Snyder, M. J. Cook, and M. R. Ford, *Health Phys.* 10, 171–82 (1964) (especially Table 2).

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Table 21.6. Balance Studies June 12–July 11, 1963 for Two Subjects, *ad libitum* Diets

	Female, Age 34, Weight ~136 lb			Male, Age 35, Weight ~165 lb		
	Intake	Output	Urine/Fecal Ratio	Intake	Output	Urine/Fecal Ratio
Total	2372 g/day[a]	1570 g/day[a]	26.0	2487 g/day[a]	1200 g/day[a]	15.0
Food (including milk, beer, etc.)	1278 g/day			1769 g/day		
Drink (water, tea, coffee)	1094 cc/day			718 cc/day		
Total water (food and drink)	1952 cc/day			1939 cc/day		
Dry solids	461 g/day			547 g/day		
Urine		1511 cc/day			1131 cc/day	
Feces		59 g/day			74 g/day	
Ca	1.1 g/day	0.78 g/day	0.32	1.7 g/day	1.4 g/day	0.39
Mg	0.23	0.23	0.55	0.40	0.13	0.11
Cu	2.9 mg/day	2.4 mg/day	0.31	2.2 mg/day	2.0 mg/day	0.11
Mn	6.4	4.8	0.0004	7.5	2.1	0.0005
Zn[b]	6.2	6.2	0.00	22.0	22.0	0.00
Ag[b]	0.082 mg/day	0.082 mg/day	0.03	0.11 mg/day	0.11 mg/day	0.05
Ba	1.4	1.2	0.06	1.5	0.62	0.11
Cr	0.033	0.006	0.50	0.007	0.005	3.0
Ni[b]	1.7	1.7	1.4	0.81	0.81	0.21
Pb	1.0	0.81	0.84	2.9	0.26	0.44
Sn	1.9	2.1	0.06	3.4	3.6	0.00
Sr	2.7	1.4	0.20	4.0	0.53	0.26
Ti	0.79	1.1	1.0	0.50	0.25	0.92

[a]Assuming the specific gravity of water, tea, coffee, and urine to be 1.

[b]Intake assumed equal to output.

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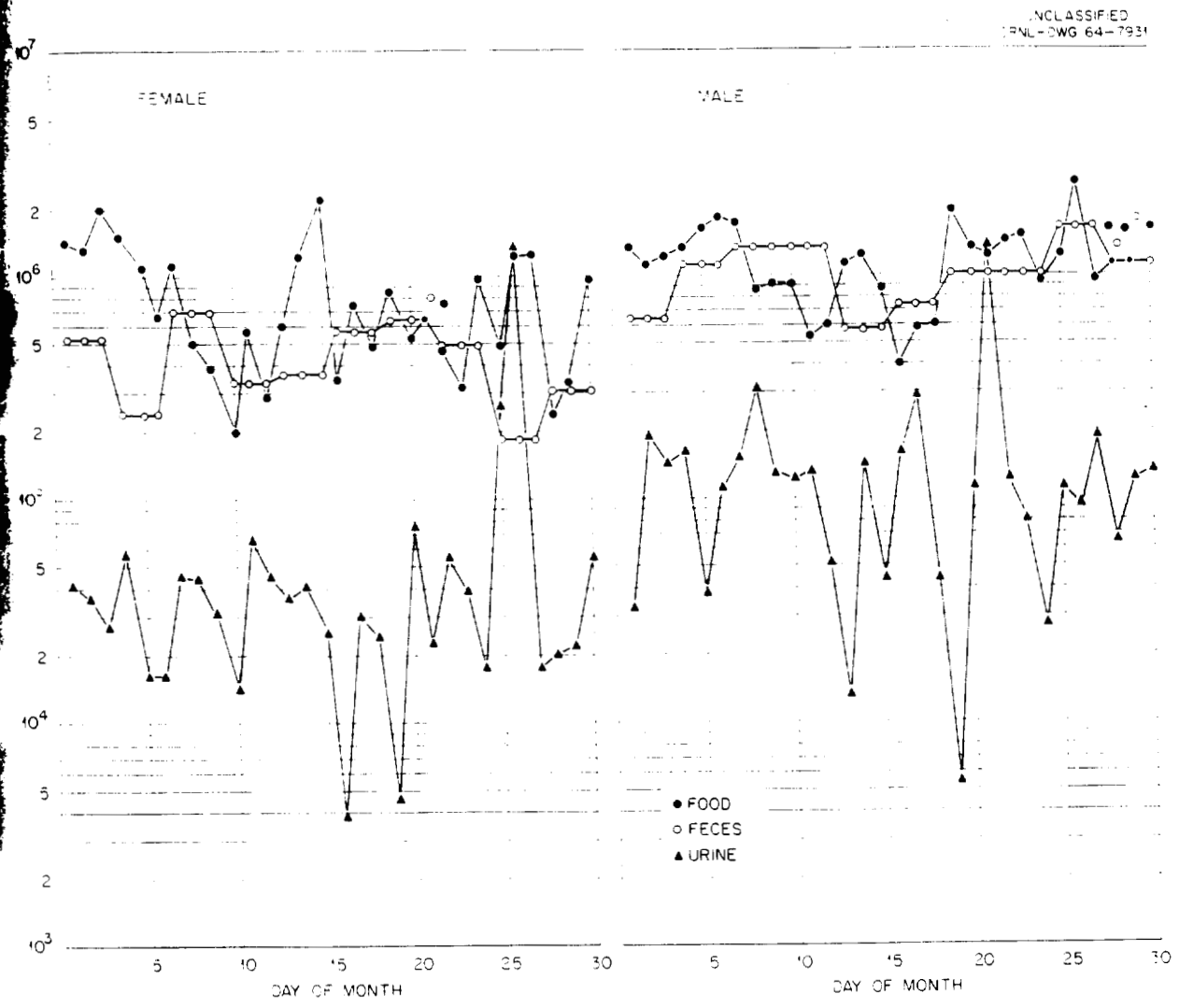

Fig. 21.4. Daily Intake and Excretion of Calcium.

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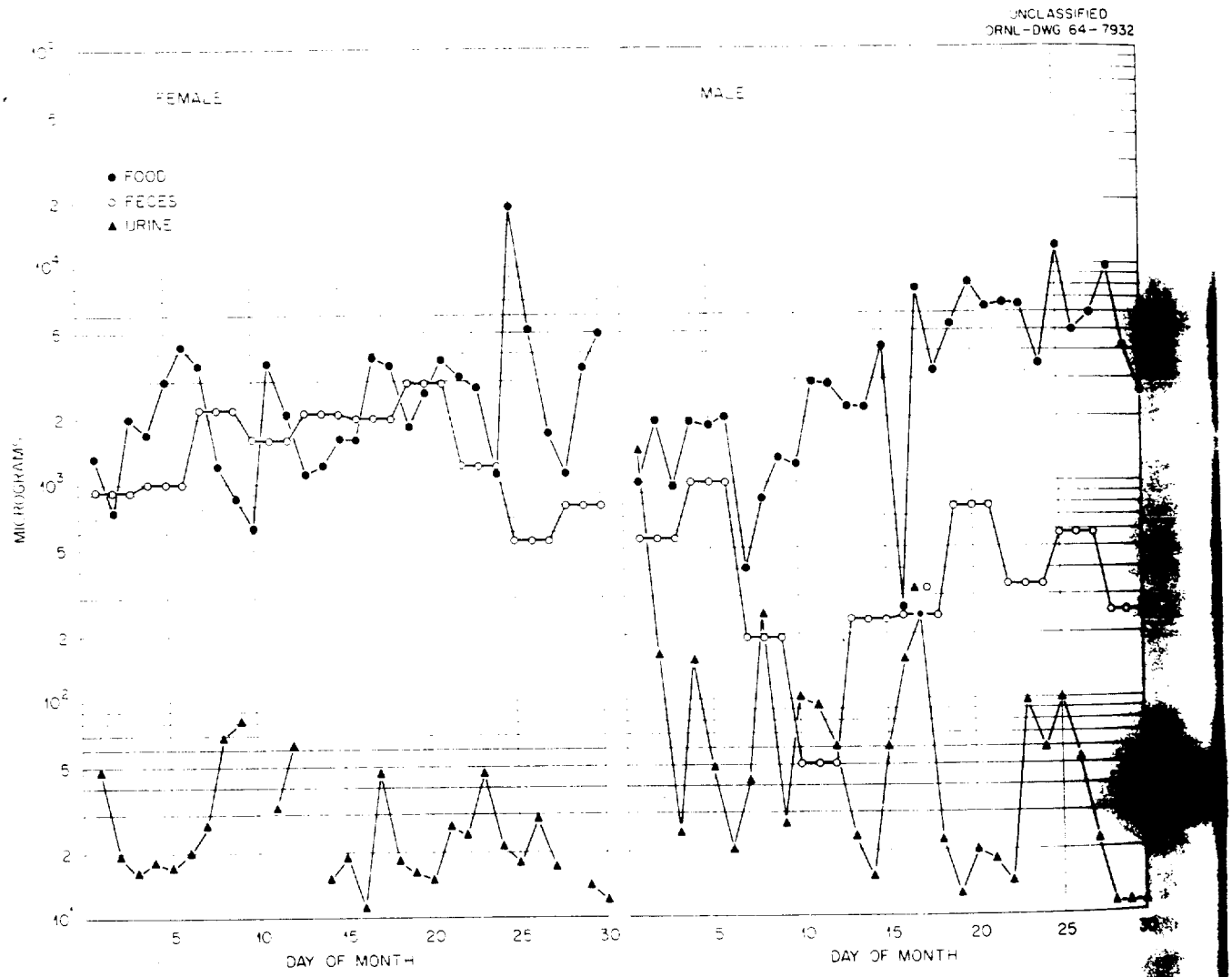

Fig. 21.5. Daily Intake and Excretion of Strontium.

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[9] J. B.
(1963).

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DISTRIBUTION OF STABLE STRONTIUM IN BONE AND SOFT TISSUES AND ITS APPLICATION TO METABOLISM OF STRONTIUM-90

W. S. Snyder M. J. Cook

Strontium has been shown to be a bone seeker both human and animal studies. Little attention been focused upon the distribution of strontium soft tissues; however, every tissue examined our program on the analysis of human tissues been found to contain a measurable amount stable strontium. For this study the age of the dividuals was restricted from 20 through 59 years, hich would include the majority of working people. ver 2200 soft tissue samples and 160 bone samples re examined spectroscopically, and the data analyzed here to estimate the distribution of ontium under near equilibrium conditions. It found that bone contains ~99% of the strontium resent in the body, thus differing significantly rom ^{226}Ra for which estimates of as low as 80% f the body burden have been given for bone.[9]

In Fig. 21.6 are presented the means of the oncentration for 27 different soft tissues and one, with the number of samples of each tissue nalyzed given in parentheses. The length of the vertical line is equal to two standard deviations on the vertical scale. If the vertical line representing the mean concentration of any tissue is turned 90° to either the right or left, plus or minus wo standard deviations may be ascertained. Based n the concentration of strontium, the tissues eem to fall into three classes: (1) bone, which is in a class to itself; (2) trachea, larynx, and aorta, which are higher in calcium than the majority of soft tissues; and (3) the remaining soft tissues.

At the meeting of the International Commission on Radiological Protection (ICRP), December 2–5, 1963, Committee 2 proposed to calculate maximum permissible concentration (MPC) values for nuclides that are incorporated into gonadal tissue on the basis of mean dose equivalent delivered to the whole gonadal tissue. On this basis of averaging over the entire organ, it is found that at the 95% confidence limit the concentration of stable strontium in the ovary differs from that in the testes. The mean concentration in the ovary is about $2\frac{1}{2}$ times that in the testes.

Since no whole bodies are available for analysis, the estimate of strontium for the whole body had to be based on samples available, and several compromises were necessary. First, many samples only contained a fraction of the entire organ or tissue being sampled, and the weight of the entire organ or tissue was not known. The amount of strontium in various organs was estimated by multiplying the weight of the organ as given for standard man[10] by the mean concentration observed in the wet tissues. Second, it was not feasible to separate bone and marrow in the course of analysis; therefore, the analytical values reported for bone also include strontium present in the marrow. Since no samples of marrow were available, we have assumed that the concentrations of strontium in red and yellow marrow are comparable to the concentrations of strontium in spleen and fat respectively. The calculated estimate of strontium content of marrow was subtracted from the analytical values reported for bone plus marrow. Finally, some tissues were not sampled, and assigned weights could not be found in literature for some other tissues. However, these tissues only account for about 1200 of the 70,000 g of standard man. The average concentration of the soft tissues which were sampled and analyzed but for which no body weights could be assigned was used as the mean concentration value for the remaining tissues for which no weights were found and for tissues not sampled. By this method the total body burden of stable strontium was estimated to be ~0.33 g.

In calculating MPC and body-burden values for radioisotopes, the fraction of the body burden in an organ is needed. This factor is designated by f_2 in the ICRP[10] and NCRP[11] internal dose reports. In Fig. 21.7 these values are presented. Of the stable strontium in the body, 98.5% is found in the bone. Although muscle has a very low concentration on a microgram per gram wet basis, it may be noted that due to its mass, which comprises

[9] J. B. Hursh and Arvin Lovaas, *Nature* **198**, 265–68 (1963).

[10] Recommendations of the International Commission on Radiological Protection, ICRP Publication 2, *Report of Committee II on Permissible Dose for Internal Radiation (1959)*, p. 151, Pergamon, London.

[11] "Maximum Permissible Body Burden and Maximum Permissible Concentrations of Radionuclides in Air and in Water for Occupational Exposure," *Natl. Bur. Std. (U.S.), Handbook 69*, GPO, Washington, 1959.

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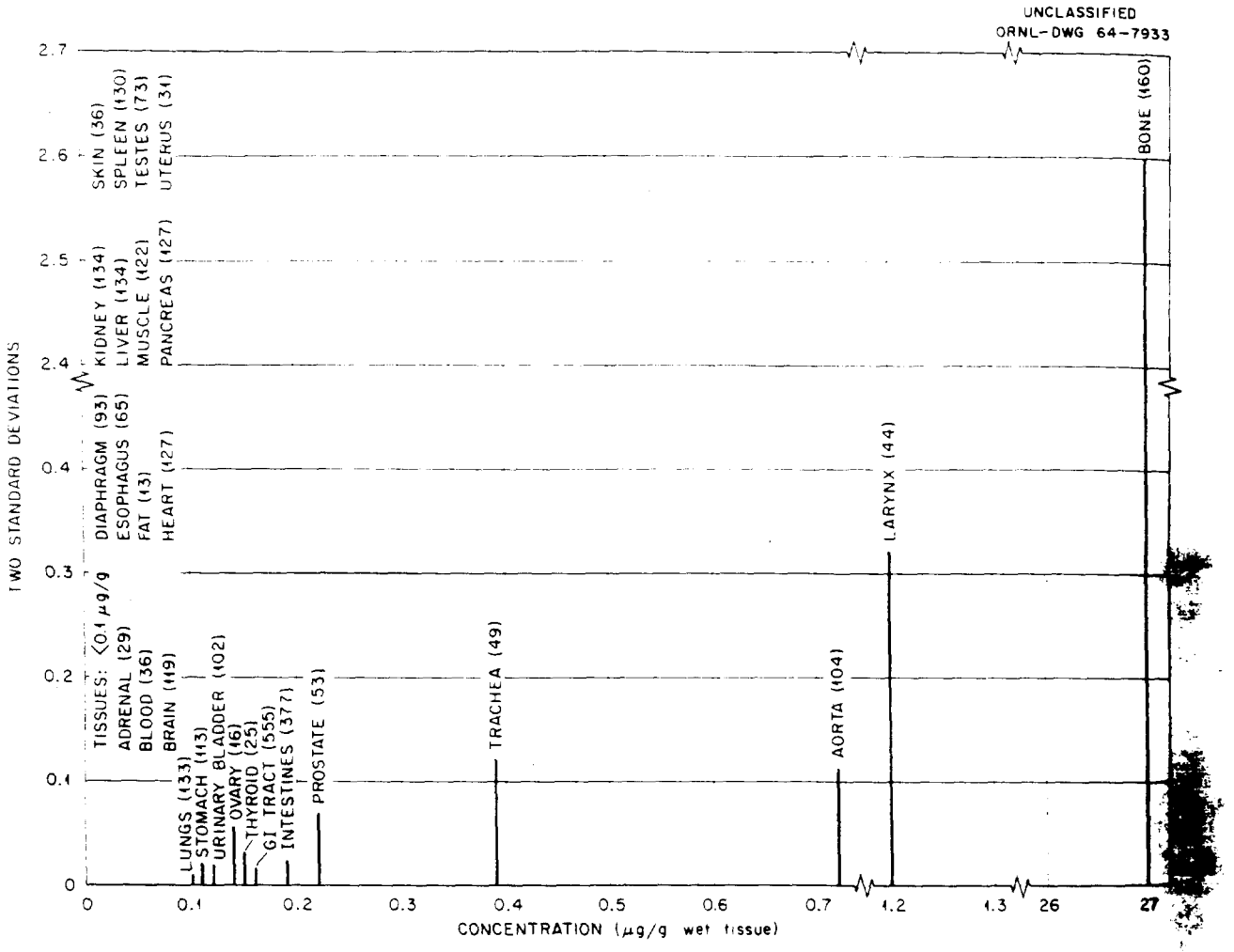

Fig. 21.6. Concentration of Stable Strontium in Wet Tissue.

almost half of the total weight of the body. muscle has the largest fraction of strontium of any of the soft tissues.

The dose which an organ receives from a radioactive element may be calculated from the concentration of this radionuclide in the tissues. A stable isotope of the same element should indicate, on the average, the pattern of distribution to be expected at near-equilibrium conditions for the radioactive element, provided suitable allowance is made for radioactive decay. Statistical methods given by Brunk[12] and by Dixon and Massey[13] have been described and applied to the data for some elements[14] and have now been applied to stable strontium in the bone. These data are presented in Fig. 21.8. The small, dotted, center line is the cumulative sample distribution of strontium in wet bone for people in the United States between ages of 20–59. The two solid lines determine a band such that at each concentration value one may assert with 95% confidence that the true distribution function for the population being sampled lies below (above) the upper (lower)

[12]H. D. Brunk, *An Introduction to Mathematical Statistics*, Ginn and Co., New York, 1960.

[13]W. J. Dixon and F. J. Massey, Jr., *Introduction to Statistical Analysis*, McGraw-Hill, New York, 1957.

[14]W. S. Snyder and M. J. Cook, *Health Phys.*, 417–23 (1963).

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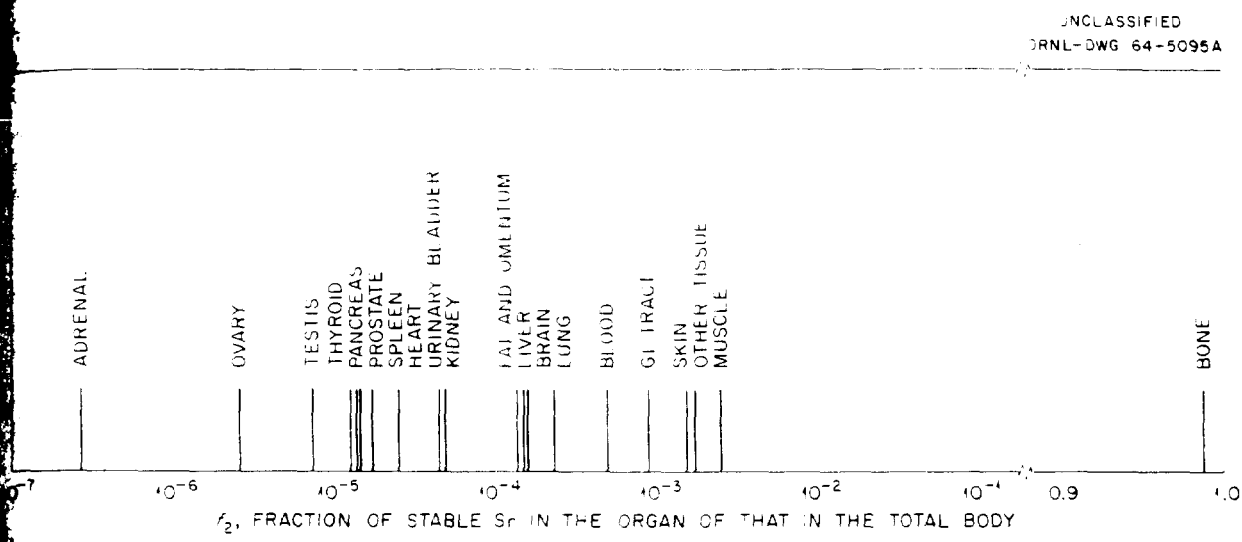

Fig. 21.7. f_2 – Fraction of Stable Strontium in the Organ of That in the Total Body.

boundary of the band. For example, at the sample mean, 27 μg of strontium per gram of wet bone, it can be asserted with 95% confidence that at least 32% of the individuals are above this value and 56% are below. For some estimates it is desirable to use the entire distribution function. This can be bracketed by use of a statistical method of Massey which defines a region bounded by the two outer curves in Fig. 21.8. It can be asserted with 95% confidence that the entire distribution curve of the population being sampled lies within this band.

Because the mean value exceeds the median value, the values are not normally distributed but are skewed to the right. Also, from this statistical analysis, 95% confidence intervals for different percentiles may be determined from the Brunk curves. As an example, the 10th percentile is between 10 and 13 μg/g and the 90th percentile is between 45 and 55 μg/g. Thus, with 95% confidence one may say that the 80% range is above 10 μg/g and, with the same confidence, that it does not extend beyond 55 μg/g.

Both strontium and calcium have been determined in each sample of tissue. However, many investigators have suggested and prefer studying strontium on the basis of the ratio of the concentrations of these elements. These ratios have been determined for 27 soft tissues and bone. The ratio of strontium to calcium in bone samples appears to be remarkably consistent, showing little dependence on age or bone type in the specimens analyzed for this study, and the study of these is continuing.

Although it is not possible to give an exact assessment of the distribution of dose in the body due to chronic exposure to ^{90}Sr, one can use these data to obtain rather good estimates of such exposure. For example, since man's exposure to ^{90}Sr in weapons fallout occurs largely via the diet, the ^{90}Sr would be expected to metabolize much as does the stable strontium of concern in this study. Because of radioactive decay, the levels of ^{90}Sr in various tissues that would occur after many years of exposure at a constant level might be expected to be somewhat less than the levels observed for comparable exposure to stable strontium. This reduction would be greatest in the tissue where the biological turnover is longest; thus the bone burden of ^{90}Sr would be relatively lower and soft tissues relatively higher than for stable strontium. However, the reduction in the bone burden cannot exceed 30% unless we postulate average turnover times in excess of 10 years. The decrease in the burden of the soft tissues would be expected to be much less.

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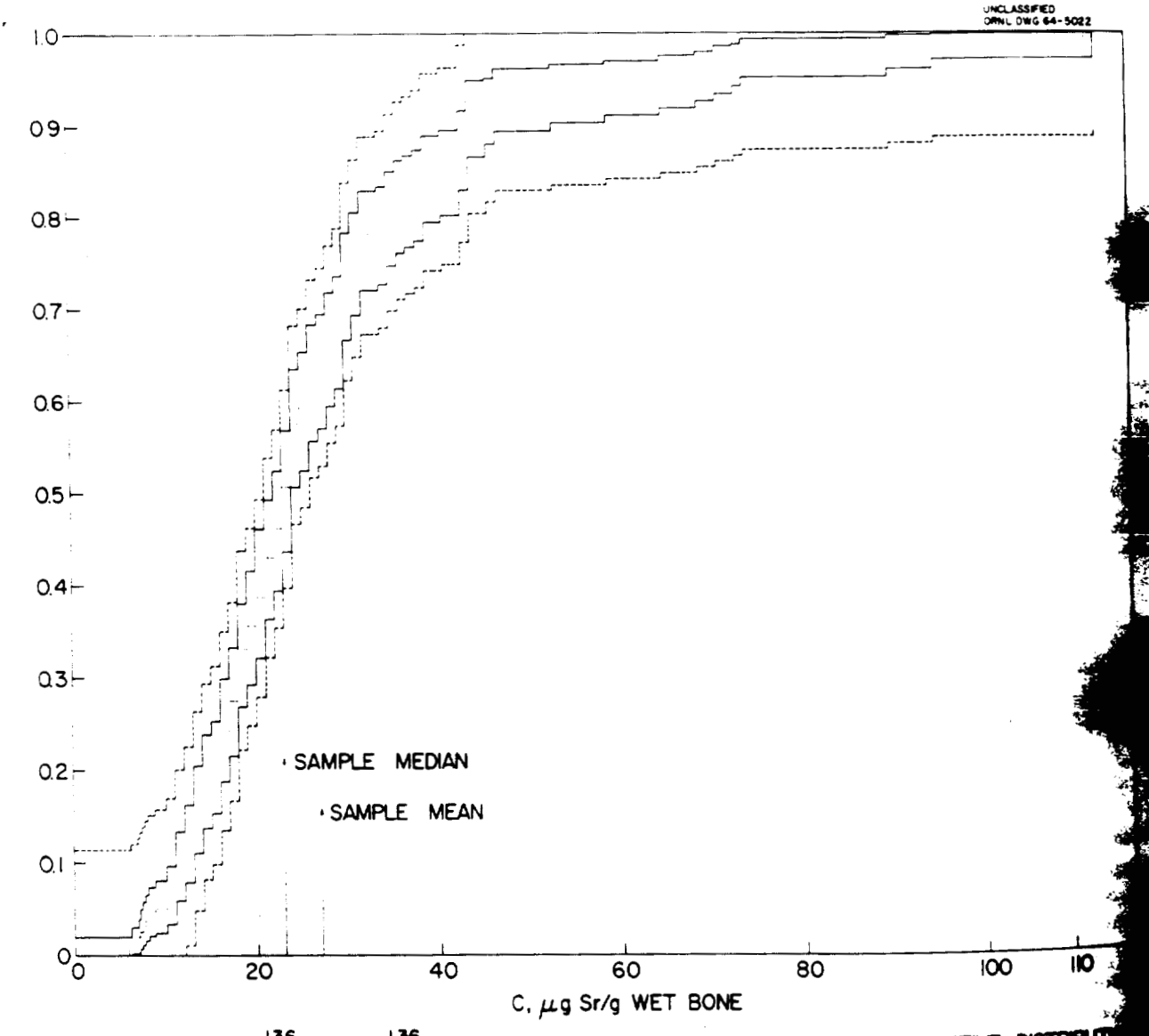
As one illustration of the usefulness of this more detailed estimate of distribution in the body, we note that the concentration of strontium in bone is about 190 times greater than the concentration in the ovary. Even if we allow for a reduction in bone of 30% due to radioactive decay, this would indicate that the ovary gets no more than $\frac{1}{130}$ of the bone dose. Thus, if the average dose to bone does not exceed 0.5 rem/year, the radiation protection guide recommended by the Federal Radiation Council, then the dose to the ovary should not exceed about 4 mrem/year and the dose to testes should be even less. This is much lower than estimates based on the values listed for the whole body which have sometimes been used, in the absence of anything better, as a basis for assessing genetic dose.



---- MASSEY $m + \frac{1.36}{\sqrt{160}}$ AND $- \frac{1.36}{\sqrt{160}}$ CONFIDENCE BAND FOR THE POPULATION CUMULATIVE DISTRIBUT

m SAMPLE CUMULATIVE SAMPLE SIZE, $n = 160 \frac{1}{160} = m, \frac{2}{160} = m, \frac{160}{160} = m$

— BRUNK 95% CONFIDENCE BOUNDS AT EACH CONCENTRATION (c)

Fig. 21.8. Ninety-five Percent Confidence Bands for the Distribution of Stable Strontium in Human Bones (A 20–59) ORNL Trace Element Data.

1147675

22. A Two-Compartment Model with Random Variable Flows

S. R. Bernard V. R. R. Uppuluri[1]

The purpose of this section is to consider the blem of nonuniform distribution of bone-seeking ionuclides, such as the alkaline-earth elements, d the effect of age on retention of strontium by gs via a mathematical study of a compartmental stem in which the connections between the comrtments are random variables. In most compartental studies it is generally assumed that the ntents of the compartments are uniformly disuted. This is not a realistic assumption for case of bone-seeking elements such as radium, ere it has been well demonstrated that hot spots activity occur as many as 20 or 30 years after take of ^{226}Ra by man.[2] Rowland states that e concentrations in the hot spots exist in regions f bone where new mineral was laid down at the me the radium was acquired and that in this neral the original concentration of Ra, expressed the ratio of Ra to Ca, was essentially the same the Ra-to-Ca ratio that existed in the blood asma at the time the new mineral was formed. here is also a second type of distribution which s much lower in concentration and rather uniform.[2] his is believed to be the result of an exchange rocess which continually transfers Ca and/or Ra oms from blood to bone and back again and which characterized by an unusually long time constant. Also, there is a growing trend to consider turnver rates in bone in terms of at least two "subcompartments," trabecular bone and compact bone. The turnover rates in these two types of bone are observed to be different – faster in trabecular bone and much slower in the compact bone. Lucas[3] has suggested that the biological half-life for Ra in trabecular bone is ~7 to 14 years and twice as long in compact bone. Also, the hot spots of Ra would disappear from the bone after perhaps 200 years since by then the whole bone would be remodeled and turned over. How may we more adequately represent these phenomena in terms of a compartmental approach?

One way to gain more insight into this problem is to consider the bone-blood system as a set of j randomly connected compartments; let the first compartment represent blood, in which the element is uniformly mixed, and the other $j - 1$ compartments represent bone. Take a subset of r compartments, $r < j - 1$, and call these trabecular bone. Let the other $j - r + 1$ compartments be cortical bone. Assume that the flows between the blood and bone are random, that is, of the $j-1$ compartments only l of them are connected to the blood in the interval 0 to t_1, while in the interval t_1 to t_2, l of them are also connected but not necessarily the same ones that were connected in the first interval of time, and so on for other intervals of time. Thus some of the $j - 1$ compartments trap the concentration that existed in the interval 0 to t_1. (This represents the laying down of new bone.) They may release it at a later time (this represents the remodeling of bone), or they may never reopen and connect with the bloodstream (this represents a hot spot), thereby maintaining the same concentration that existed at time t_1.

The above constitutes the general compartmental system we want to study. We need to determine the behavior of the system – what are the concentrations in a given compartment as a function of time – how long must one wait until a compartment releases the trapped material – how many randomly connected compartments are required to represent the available data on retention in bone? We find that these are difficult questions and that before we can get some answers to the general case we have to initiate our studies on a simpler system, namely, that of only two compartments with random flows. Here we gain some insight into the behavior of the general system and we find that the two-compartment system gives some information on the age effect on retention of ^{85}Sr by beagles. Thus we present the results of the study of this simpler model in this section.

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[1]Mathematics Division.

[2]R. E. Rowland, "Late Observations of the Distribution of Radium in the Human Skeleton," p. 339 in *A Symposium on Radioisotopes in the Biosphere*, ed. by R. S. Caldecott and L. A. Snyder, Univ. of Minnesota, Minneapolis, 1960.

[3]H. E. Lucas, personal communication, 1964.

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TWO-COMPARTMENT MODEL

Let us suppose that we have two compartments of the same volume, with a gate between them which can be opened or closed at will (Fig. 22.1). Initially, say we have unit concentration in compartment 1 and zero concentration in compartment 2. Also, suppose that there is provision for an inlet and outlet through compartment 1 which will enable the concentration (in 1 only if the gate is closed or in both if the gate is open) to reduce by a factor of $e^{-\lambda\Delta t}$ in a time interval Δt. (Here λ is a constant.)

Take a fair coin, that is, with $P(H) = \frac{1}{2} = P(T)$, and toss it once. If a head appears, open the gate and assume that instantly the material in both the compartments becomes homogeneous, that is, of concentration $\frac{1}{2}$ in each. Wait for a time interval Δt and suppose that during the Δt the inlet and outlet through compartment 1 are functioning. After time Δt, the concentration in 1 and 2 will become $\frac{1}{2}\, e^{-\lambda\,\Delta t}$ each.

If a tail appears, keep the gate between compartments 1 and 2 closed, and after time Δt (during which the inlet and outlet through compartment 1 are functioning) the concentration in 1 will become $e^{-\lambda\,\Delta t}$ and of course the concentration in compartment 2 is still zero.

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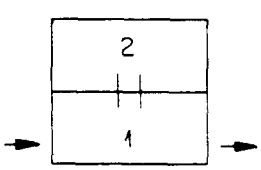

Fig. 22.1. A Two-Compartment Model.

The equations for this system are set up as follows:

Let $C_{0,1}$ denote the initial concentration in compartment 1 and $C_{0,2}$ denote the initial concentration in compartment 2, that is,

$$\begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}.$$

This corresponds to an experiment where initially the gate is always closed. More generally, let $C_{n,i}$ denote the concentration at time t_n, $n = 1, 2, 3, \ldots$, in compartment $i = 1, 2$.

Let X denote the binomial random variable, corresponding to the fair coin, that is,

$$X = \begin{cases} 1 \text{ if } H \text{ appears (with probability } \frac{1}{2}) \\ 0 \text{ if } T \text{ appears (with probability } \frac{1}{2}) \end{cases}.$$

Then, it is easily seen that

$$\begin{pmatrix} C_{1,1} \\ C_{1,2} \end{pmatrix} = \begin{pmatrix} \frac{1}{2}\, e^{-\lambda\,\Delta t}(C_{0,1} + C_{0,2}) & e^{-\lambda\,\Delta t}\, C_{0,1} \\ \frac{1}{2}\, e^{-\lambda\,\Delta t}(C_{0,1} + C_{0,2}) & C_{0,2} \end{pmatrix} \begin{pmatrix} X_1 \\ 1 - X_1 \end{pmatrix},$$

where X_1 denotes the binomial random variable at the first stage of the experiment, that is, more simply,

$$\begin{pmatrix} C_{1,1} \\ C_{1,2} \end{pmatrix} = \begin{pmatrix} \frac{1}{2}\, e^{-\lambda\,\Delta t} & e^{-\lambda\,\Delta t} \\ \frac{1}{2}\, e^{-\lambda\,\Delta t} & 0 \end{pmatrix} \begin{pmatrix} X_1 \\ 1 - X_1 \end{pmatrix},$$

which is a compact way of representing all the four possible outcomes, namely,

if heads, that is, $X_1 = 1$, $\quad C_{1,1} = \frac{1}{2}\, e^{-\lambda\,\Delta t} = C_{1,2}$;

if tails, that is, $X_1 = 0$, $\quad C_{1,1} = e^{-\lambda\,\Delta t}$; $C_{1,2} = 0$.

Next, we shall obtain a recursive relation between

$$\begin{pmatrix} C_{1,1} \\ C_{1,2} \end{pmatrix} \text{ and } \begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix},$$

connected through the random variable X_1.

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$$
\begin{pmatrix} C_{1,1} \\ C_{1,2} \end{pmatrix} = C_{0,1} \begin{pmatrix} \frac{1}{2} e^{-\lambda \Delta t} & e^{-\lambda \Delta t} \\ \frac{1}{2} e^{-\lambda \Delta t} & 0 \end{pmatrix} \begin{pmatrix} X_1 \\ 1 - X_1 \end{pmatrix}
$$

$$
+ C_{0,2} \begin{pmatrix} \frac{1}{2} e^{-\lambda \Delta t} & 0 \\ \frac{1}{2} e^{-\lambda \Delta t} & 1 \end{pmatrix} \begin{pmatrix} X_1 \\ 1 - X_1 \end{pmatrix}
$$

$$
= C_{0,1} \begin{pmatrix} e^{-\lambda \Delta t} - \frac{1}{2} e^{-\lambda \Delta t} X_1 \\ \frac{1}{2} e^{-\lambda \Delta t} X_1 \end{pmatrix}
$$

$$
+ C_{0,2} \begin{pmatrix} \frac{1}{2} e^{-\lambda \Delta t} X_1 \\ 1 - (1 - \frac{1}{2} e^{-\lambda \Delta t}) X_1 \end{pmatrix}
$$

$$
= \begin{pmatrix} e^{-\lambda \Delta t} - \frac{1}{2} e^{-\lambda \Delta t} X_1 & \frac{1}{2} e^{-\lambda \Delta t} X_1 \\ \frac{1}{2} e^{-\lambda \Delta t} X_1 & 1 - (1 - \frac{1}{2} e^{-\lambda \Delta t}) X_1 \end{pmatrix} \times \begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix}
$$

where we recall that

$$
\begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}.
$$

By following the same reasoning we obtain, if X_m denotes the binomial random variable at the mth stage,

$$
\begin{pmatrix} C_{m,1} \\ C_{m,2} \end{pmatrix} = \begin{pmatrix} e^{-\lambda \Delta t} - \frac{1}{2} e^{-\lambda \Delta t} X_m & \frac{1}{2} e^{-\lambda \Delta t} X_m \\ \frac{1}{2} e^{-\lambda \Delta t} X_m & 1 - (1 - \frac{1}{2} e^{-\lambda \Delta t}) X_m \end{pmatrix} \times \begin{pmatrix} C_{m-1,1} \\ C_{m-1,2} \end{pmatrix} m = 1, 2, \ldots .
$$

If we assume that $X_1, X_2, \ldots, X_n$ are independent, identically distributed, random variables and E denotes the expectation operator, then

$$
E \begin{pmatrix} C_{n,1} \\ C_{n,2} \end{pmatrix} = \begin{pmatrix} \frac{3}{4} e^{-\lambda \Delta t} & \frac{1}{4} e^{-\lambda \Delta t} \\ \frac{1}{4} e^{-\lambda \Delta t} & \frac{1}{2} - \frac{1}{4} e^{-\lambda \Delta t} \end{pmatrix}^n \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (1)
$$

because $E(X_m) = \frac{1}{2}$; $m = 1, 2, \ldots, n$; $\Delta t = t/n$.

Let $a_n = e^{-\lambda \Delta t}$ and μ_1, μ_2 denote the eigenvalues of the matrix

$$
S = \begin{pmatrix} \frac{3}{4} a_n & \frac{1}{4} a_n \\ \frac{1}{4} a_n & \frac{1}{2} + \frac{1}{4} a_n \end{pmatrix} = \begin{pmatrix} s_{11} & s_{12} \\ s_{12} & s_{22} \end{pmatrix}. \quad (2)
$$

It turns out that

$$
\mu_1 = \frac{1}{4} [2 a_n + 1 + \sqrt{2 a_n^2 - 2 a_n + 1}]
$$

and

$$
\mu_2 = \frac{1}{4} [2 a_n + 1 - \sqrt{2 a_n^2 - 2 a_n + 1}],
$$

and

$$
S = \frac{1}{(\mu_2 - \mu_1) s_{12}} \begin{pmatrix} s_{12} & s_{12} \\ \mu_1 - s_{11} & \mu_2 - s_{11} \end{pmatrix} \times \begin{pmatrix} \mu_1 & 0 \\ 0 & \mu_2 \end{pmatrix} \begin{pmatrix} \mu_2 - s_{11} & -s_{12} \\ -(\mu_1 - s_{11}) & s_{12} \end{pmatrix};
$$

therefore

$$
S^n = \frac{1}{(\mu_2 - \mu_1) s_{12}} \begin{pmatrix} s_{12} & s_{12} \\ \mu_1 - s_{11} & \mu_2 - s_{11} \end{pmatrix} \times \begin{pmatrix} \mu_1^n & 0 \\ 0 & \mu_2^n \end{pmatrix} \begin{pmatrix} \mu_2 - s_{11} & -s_{12} \\ -(\mu_1 - s_{11}) & s_{12} \end{pmatrix},
$$

$$
S^n \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \frac{1}{(\mu_2 - \mu_1) s_{12}} \times \begin{pmatrix} s_{12} [\mu_1^n (\mu_2 - s_{11}) - \mu_2^n (\mu_1 - s_{11})] \\ (\mu_1 - s_{11})(\mu_2 - s_{11})(\mu_1^n - \mu_2^n) \end{pmatrix}. \quad (3)
$$

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It is of interest to determine the solution for $n \to \infty$, $\Delta t \to 0$. Now, as $n \to \infty$, we have:

$$a_n \to 1,\ s_{11} \to \tfrac{3}{4},\ s_{12} \to \tfrac{1}{4},\ \mu_1 \to 1,\ \mu_2 \to \tfrac{1}{2},$$

$$(\mu_2 - \mu_1) \to -\tfrac{1}{2},\ \mu_1^n \to e^{-\frac{3}{4}x\lambda t},\ \text{and}\ \mu_2^n \to 0.$$

Therefore

$$\lim_{n \to \infty} S^n \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \tfrac{1}{2} e^{-\frac{3}{4}x\lambda t} \begin{pmatrix} 1 \\ 1 \end{pmatrix}.$$

In Eqs. (1), (2), and (3) we see that the expected concentrations are functions of two parameters, λ and Δt, and the initial conditions. We investigate the effect of Δt on the concentrations by graphical techniques. Figure 22.2 presents a plot of the sum of the concentrations in both compartments for $\Delta t = 0+$, 1, 2, 4, and $\lambda = 0.693$. As can be seen, when the interval width increases, the total concentrations decrease more rapidly, initially, but then it decreases more slowly along a second exponential component. Note also the intercept of this second exponential decreases with increasing Δt. It may be that the most appropriate value of Δt varies with the age of the animal, and thus the model might suggest an effect of age on the biological turnover time. It has been noted that the biological half-life for retention of Cs is larger for young than for old dogs.[4] Now, if a second exponential component would occur, does it have a longer half-life and a smaller intercept in older dogs? In the case of ^{85}Sr retention in dogs of different ages, as observed by Glad, Mays, and Fisher,[5] it appears that this model gives approximately the same quantitative picture as far as the intercept of the long-term component is concerned, but not the biological half-life, T_b. Figure 22.3 shows a plot of the data of Glad *et al.* Note the half-life of the second exponential component indicated in these figures is generally larger for young dogs that are injected than is the T_b for the second component fitting retention in old dogs. Note also that the intercept of that second component decreases with age. We find

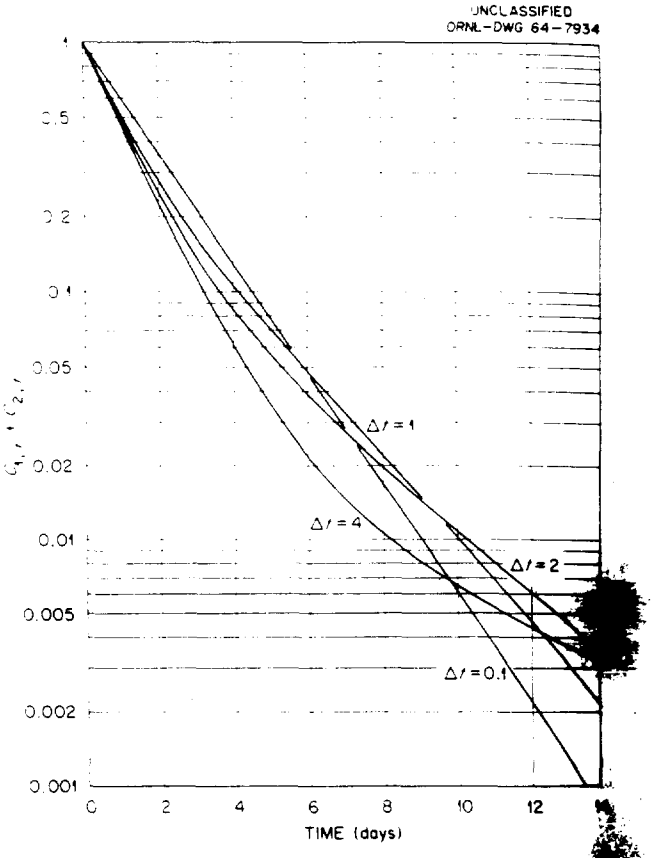

Fig. 22.2. Plot of the Sum of Concentrations In Compartments 1 and 2 vs Time for Case of Equally Spaced Subintervals of Time.

that we can get a better interpretation of data in terms of increasing subinterval wi

Also, Glad and Mays have shown that t data can also be represented with a power functio It is possible to obtain something akin to a powe function from this two-compartment model wit random flows simply by taking a partitioning o time intervals which increase monotonically. this case we have to have a more general notatio Let

$$\Delta t_i = t_i - t_{i-1}$$

which are not necessarily equal intervals. The instead of Eq. (1), we have

$$E\begin{pmatrix} C_{t_i,1} \\ C_{t_i,2} \end{pmatrix} = \left(\prod_{i=n}^{1} S(\Delta t_i) \right) \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

[4]W. P. Norris, *Late Effects of Radiation Exposure*, presented at Radiation Research Society meeting, Miami Beach, Fla., May 18–20, 1964.

[5]B. W. Glad, C. W. Mays, and W. Fisher, *Radiation Res.* 12, 672–81 (1960).

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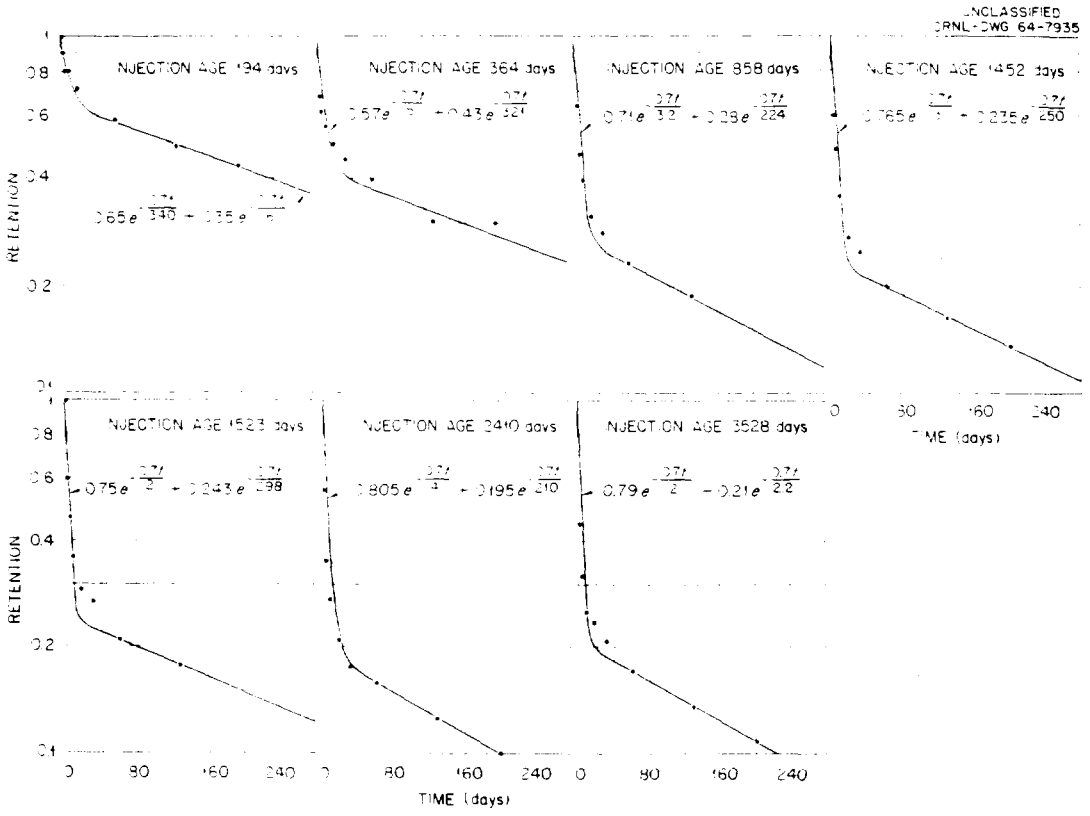

Fig. 22.3. Strontium-85 Retention in Dogs.

where S is the matrix which has already been defined in Eq. (2). When all Δt's are equal, we recover Eq. (3) again. Now, taking a specific case, that is, $\Delta t_i = i$ and $i = 1, 2, \ldots, 7$, and letting $\lambda = 0.693$, we insert into Eq. (4) and calculate the expected concentrations. Figure 22.4 presents a plot of the total concentrations, that is, $(C_{t_i 1} + C_{t_i 2})$, as a function of time. As can be seen, the slope of this function is monotonically decreasing. It can be seen from inspection of Eqs. (2) and (4) that as Δt_i gets larger

$$S(\Delta t_i) \to \begin{pmatrix} \sim 0 & \sim 0 \\ \sim 0 & \frac{1}{2} \end{pmatrix}$$

and hence the concentration in the second compartment decreases by $\frac{1}{2}$, but the time interval for this reduction to occur is larger the farther out we go. This behavior is similar to a power function.

To gain some further insight into the effect of age on retention from this model, consider the case where we let $\Delta t_i = i + T$, $\Delta t_0 = 0$, and $i = 0, 1, 2, 3, \ldots$. Here we let T correspond to the age of an animal at the time of injection. This corresponds to a simulation of aging. The gate opens and closes in a shorter time interval in a younger animal and in a longer time interval in an older animal. For example, in a newborn animal the gate first opens (or closes) after only one unit of time, while in a 3-year-old animal the gate first opens (or closes) after three units of time. We calculate the sum of the expected concentrations in compartments 1 and 2 for this case, and the results are presented graphically in Fig. 22.5. As can be seen, the curves are of the same qualitative shape as Glad and Mays observed in their experiments on ^{85}Sr retention in dogs of different ages.

It is to be pointed out here that the trend with age observed in Fig. 22.5 could be reversed by simply changing the initial conditions on the column vector. Recall that we dealt with the specific case where the gate between compartments 1 and 2 is initially closed. If we were to decide whether

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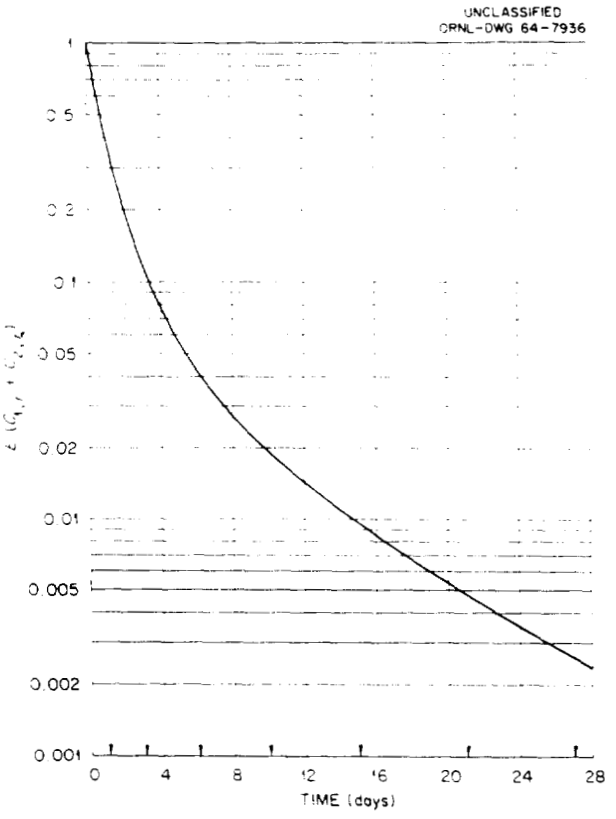

Fig. 22.4. Expected Sum of Concentrations in Compartments for Case of Increasing Subinterval Width.

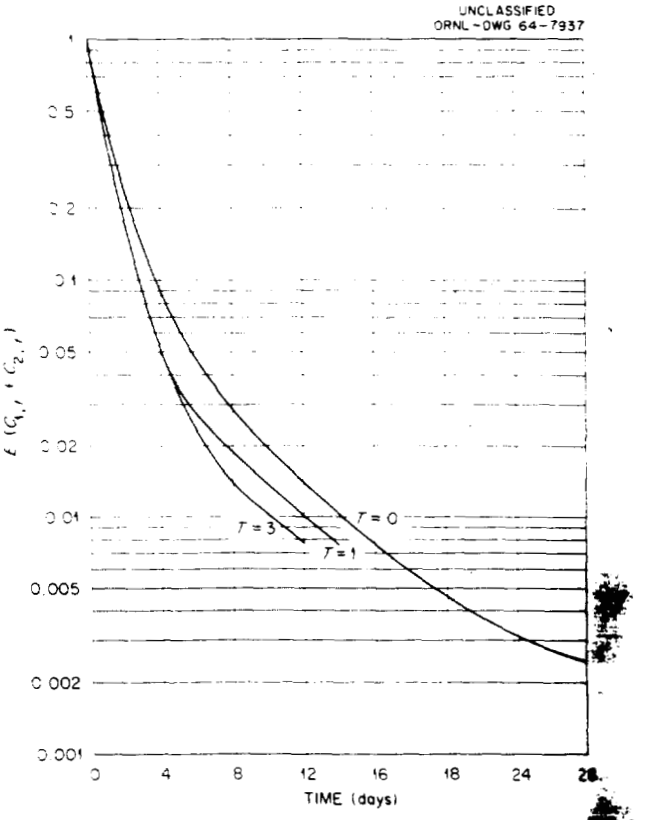

Fig. 22.5. Expected Sum of Concentrations for Co[illegible] of Increasing Subinterval Width and for Different Ages.

this gate was initially opened or closed by a random toss of the coin, then the expected value of the initial concentrations would be

$$E\begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix} = \begin{pmatrix} 3/4 \\ 1/4 \end{pmatrix}.$$

If we used this vector of initial concentrations, then the sum of the expected concentrations in

$$E\begin{pmatrix} C_{n,1} \\ C_{n,2} \end{pmatrix} = \begin{pmatrix} \frac{1}{8}(3\,e^{-\lambda_1 \Delta t} + e^{-\lambda_2 \Delta t}) & \frac{1}{8}(e^{-\lambda_1 \Delta t} + e^{-\lambda_2 \Delta t}) \\ \frac{1}{8}(e^{-\lambda_1 \Delta t} + e^{-\lambda_2 \Delta t}) & \frac{1}{2} + \frac{1}{8}(e^{-\lambda_1 \Delta t} + e^{-\lambda_2 \Delta t}) \end{pmatrix}^n \begin{pmatrix} C_{0,1} \\ C_{0,2} \end{pmatrix}.$$

compartments 1 and 2 would behave differently for different interval widths of time. One would expect that as the interval was increased, then, since compartment 2 initially starts out with material in it, it would keep it longer the longer the intervals. The chances for release would [illegible] greater the shorter the intervals.

Before closing, we list the equation for the [illegible] compartment model, that is, the case where [illegible] between compartments 1 and 2 are random [illegible] flow out of compartment 1 also is random. Let [illegible] denote one rate of flow and λ_2 the other rate [illegible] flow out of compartment 1. Assume these ha[illegible] equally probable occurrences. Then it can [illegible] shown that

This solution is only for the case where one [illegible] two fair coins at the same time, that is, it is [illegible] sumed that both gates are opened (or closed) [illegible] the equally spaced intervals of time accor[illegible] as the respective coins fall heads or tails.

Some [illegible] jected [illegible] have bee[illegible] between [illegible] or what [illegible] life shor[illegible] previous [illegible] equivale[illegible] would b[illegible] end poin[illegible] a metho[illegible] points as [illegible] First. [illegible] lined in [illegible] argument [illegible] that if s[illegible] where S_0 [illegible] irradiatec[illegible] jected (o[illegible] is propor[illegible] by D, tha[illegible]

For an [illegible] eliminate[illegible]

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where [illegible] bone has [illegible] ported d[illegible] $S_0 - S$, [illegible] that this [illegible] it was no[illegible] read

[1]W. S. S[illegible] Rept. June [illegible]

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23. Studies on the Internal Exposure/External Exposure Equivalence Relation

S. Bernard

Some data on the life-shortening effects of injected ^{239}Pu and ^{226}Ra in rats, rabbits, and dogs have been interpreted with the equivalence relation between internal exposure and external exposure, or what is really the same as the linear theory of life shortening by internal dose discussed in the previous progress report.[1] Also, by using this equivalence relation, an MPC is derived which would be related only to life shortening as an end point. This is offered as an illustration of a method which might be applied to other end points as well if appropriate data are available.

First, some familiarity with the approach outlined in the previous report is assumed. The argument in that report could be modified to show that if shortening of life is denoted by $S_0 - S$, where S_0 is the life span of uninjected (or nonirradiated) animals and S is the life span of injected (or radiated) animals, then life shortening is proportional to the total absorbed dose, denoted by D, that is,

$$S_0 - S = \alpha' D \,. \tag{1}$$

For an internal emitter, which is exponentially eliminated from the body,

$$D = \beta \int_0^S q_0 e^{-\lambda t}\, dt \,, \tag{2}$$

where β is the rem/unit time/μc injected. From Eqs. (1) and (2) we find

$$S_0 - S = \frac{\alpha q_0}{\lambda}\left(1 - e^{-\lambda S}\right) , \tag{3}$$

where $\alpha = \alpha'\beta$ with units of μc^{-1}. The dose to bone has been estimated on the basis of the reported data and the observed life shortening, $S_0 - S$, plotted against dose, and it was found that this linear theory was not obeyed. However, it was noted that when Eq. (3) was rearranged to read

$$q_0 = \frac{S_0 - S}{(\alpha/\lambda)(1 - e^{-\lambda S})} \tag{4}$$

and when an (S, q_0) plot of the data was made, then this function would fit the data fairly adequately if a suitable value of λ was used. We have estimated the parameters α and λ from the (S, q_0) data and have compared this λ with that estimated in metabolism studies. The two values of λ generally differed. Let us illustrate this point with the rat data of Moskalev.[2] Figure 23.1 shows a graph of his data – a plot of q_0/m, the microcuries of ^{239}Pu/kg injected into the rats vs S, the median life span in days. The equation and curve fitting the data are also shown. Note that the effective half-life is 36 days. Moskalev did not have any metabolic data from which to estimate a metabolic effective half-life (T). However, when we compared it with the T estimated from Fink's (S, q_0) data on rats,[3] we found $T = 72$ days, differing by a factor of only 2.

When we attempt to interpret the data of Bukhtoyarova[4] on rabbits or the Utah data on beagle dogs[5] with this model, we find they are not really adequate as yet, because the studies are not complete. However, we can make some tentative calculations with these data. Figure 23.2 shows the ^{239}Pu injection data on rabbits, and the Utah data on dogs injected with ^{239}Pu and ^{226}Ra are shown in Figs. 23.3 and 23.4 respectively. (In these latter two figures, the mean life spans are estimated for dogs dying from osteosarcoma only and not the mean life span for all causes of death.) Note, however, that a linear fit of the long time data has been estimated and plotted together with the data in Figs. 23.2 to 23.4. From the estimated intercept, $\lambda S_0/\alpha m$, we can see that dogs are more sensitive to ^{239}Pu than rabbits which, in turn, are more sensitive than rats, that

[1] W. S. Snyder *et al.*, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, pp. 163–65.

[2] Yu. I. Moskalev, Joint Publications Research Service translation 16,900, p. 12, 1962 (Russian translation).

[3] R. M. Fink, *Biological Studies with Polonium, Radium, and Plutonium*, McGraw-Hill, New York, 1950.

[4] Z. M. Bukhtoyarova, Joint Publications Research Service translation 16,900, p. 65, 1962 (Russian translation).

[5] T. F. Dougherty *et al.*, *Some Aspects of Internal Emitters*, Pergamon, New York, 1962.

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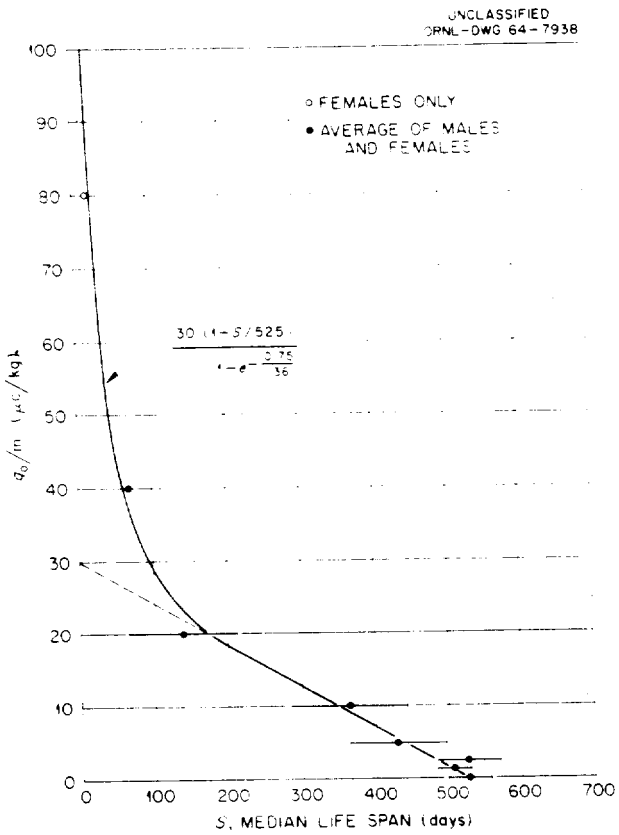

Fig. 23.1. Life Shortening by Injected ^{239}Pu – Rats.

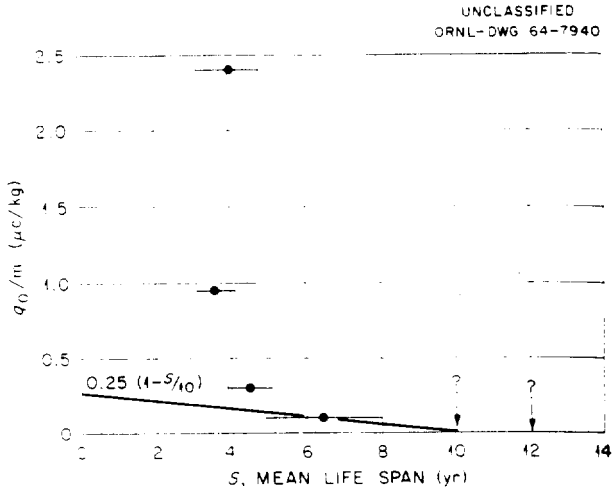

Fig. 23.3. Life Shortening by Injected Plutonium – Dogs.

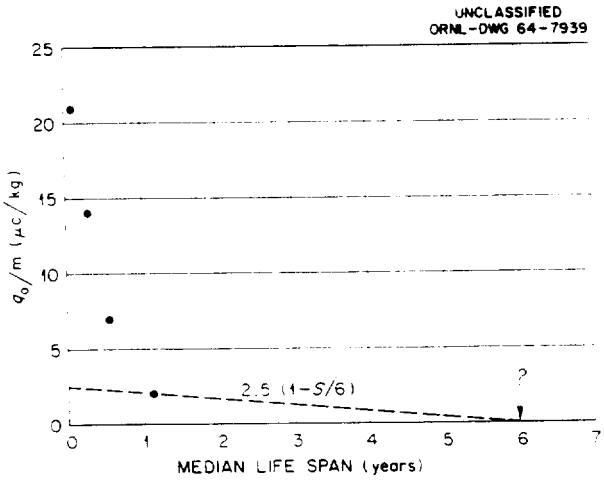

Fig. 23.2. Life Shortening by Injected Plutonium – Rabbits.

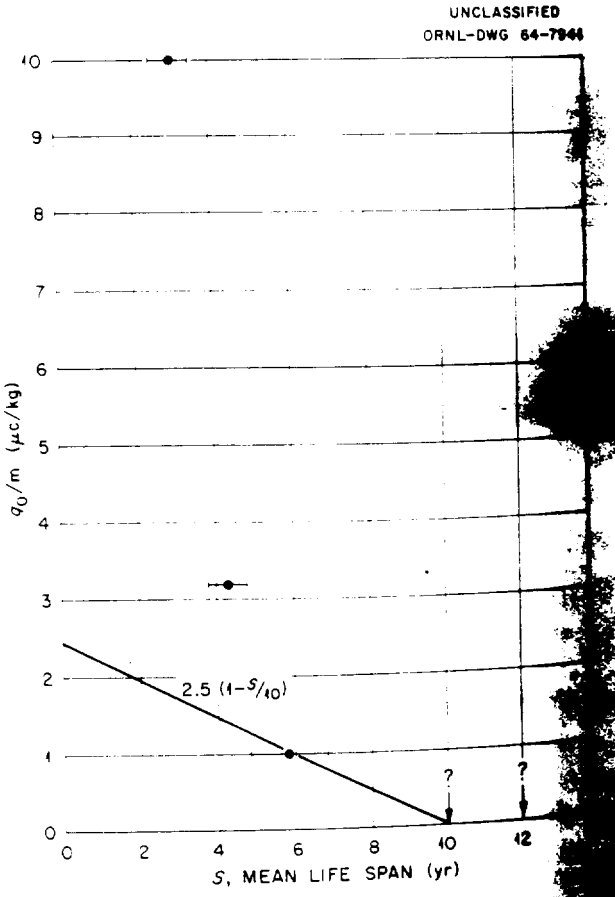

Fig. 23.4. Life Shortening by Injected Radium – D

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[6]Cited by

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the equations for the linear (or assumed-to-be-ear) regions are

$$
\begin{aligned}
/m &\longrightarrow 30\,(1 - S/525 \text{ days}) \text{ rats} \quad \text{(Fig. 23.1)}, \\
&\longrightarrow 2.5\,(1 - S/6 \text{ years}) \text{ rabbits} \quad \text{(Fig. 23.2)}, \\
&\longrightarrow 0.25\,(1 - S/10 \text{ years}) \text{ dogs} \quad \text{(Fig. 23.3)}.
\end{aligned}
\tag{5}
$$

hat does this mean in terms of MPC values for an? Let us review the method we proposed for estimating MPC values.

Recall that in the previous report we noted that a plot of R, the daily whole-body dose rate in rads/day, to mice in the (S,R) plane became a inear function too for values of $S/S_0 \leq 0.5$; it was

$$R \sim 40\,(1 - S/S_0) \tag{6}$$

in r/day (mice). Now, from Braestrup's data on American radiologists,[5] we estimate

$$R \sim 5\,(1 - S/S_0) \tag{7}$$

in r/day. Next we set up an equivalence between the whole-body external exposure data and the injection data by noting from Eq. (4) that

$$q_0 \longrightarrow \left(\frac{\lambda S_0}{\alpha}\right)(1 - S/S_0) \tag{8}$$

and from Eqs. (7) and (8)

$$q_0 = \left(\frac{\lambda S_0/\alpha}{5}\right) R\,. \tag{9}$$

Now, the question is which value in Eq. (5) should be employed for the intercept $(\lambda S_0/\alpha)$ in Eq. (9). We attempt to answer this question by plotting the $(\lambda S_0/\alpha)$ in Eq. (5) vs the mean life span of the species. Figure 23.5 shows a log × log graph of the intercept plotted vs the life span of the species. The power function 440 S^{-3} fits the data fairly well. Thus, if this function is used to extrapolate to the 70-year life span of man, we find

$$\frac{\lambda S_0}{\alpha m} = 0.00129\ \mu c/kg\,.$$

Inserting this into Eq. (9) and letting $R = 0.02$ rad/day, then

$$q_0/m = 0.56 \times 10^{-5}\ \mu c/kg$$

and

$$q_0 \sim 39 \times 10^{-5}\ \mu c\,.$$

Recall that the interpretation of this is that a single injection of 39×10^{-5} μc of ^{239}Pu into man would produce the same life shortening as the permissible whole-body external dose rate of 5 rads/year. What $(MPI)_w$ does this correspond to? It turns out to be ~ 0.07 μc/quarter. The ICRP report[7] recommends ~ 10 μc/quarter. This would imply that the ICRP report value is not a conservative value.

We interpret the ^{226}Ra rat data in the same manner and find from Fink's rat data $\lambda S_0/\alpha m = 350$ μc/kg, and from Fig. 23.4, on dogs, $\lambda S_0/\alpha m = 2.5$ μc/kg. Assuming the power function relation

[6] Cited by P. R. J. Burch, *Nature* **185**, 135–42 (1960).

[7] "Report of Committee II on Permissible Dose for Internal Radiation (1959)," *Health Phys.* 3 (1960).

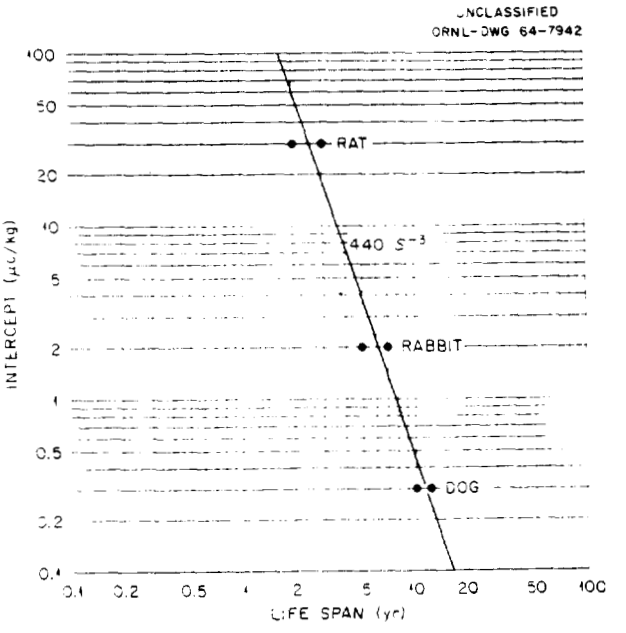

Fig. 23.5. Intercept of the Formula for Life Shortening $(\lambda S_0/\alpha)$ vs Life Span.

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between this intercept and mean life span of the species, we find $\Lambda S_0 \cdot m \sim 7210\ S^{-3.3}$. Extrapolating to man ($S = 70$ years), $(\Lambda S_0 \cdot m)_{man} = 0.006$ $\mu c \cdot kg$ for ^{226}Ra. Calculating Eq. (9) with $R = 0.02$ rad/day, we obtain $q_0 \cdot m \cong 3 \cdot 10^{-5}$ $\mu c \cdot kg$ and $q_0 \sim 210 \cdot 10^{-5}$ μc. This is lower than the current permissible ^{226}Ra burden of 0.1 μc by a factor of ~500, and it yields a correspondingly lower $(MPI)_w$ too. This implies that if the 0.1-μc value is a valid body burden, then this method of estimating MPC values is either too conservative, or invalid, or perhaps the dog is equally as sensitive to internal exposure from ^{239}Pu and ^{226}Ra as man is. If we adopt the latter assumption and recalculate the $(MPI)_w$, we find for ^{239}Pu, $(MPI)_w \sim 14$ μc/quarter (compared with ICRP value of 10 μc/quarter) and for ^{226}Ra, $(MPI)_w \cong 0.01$ μc/quarter (compared with ICRP value of 0.02 μc/quarter).

The data on dogs are not adequate as yet, and hence it would be premature to draw any firm conclusions from these calculations. Also, we mention here that this estimation method should not be used without checking with data on more species of animals which were administered lower injection levels or which experience continuous feedings and inhalation of the radionuclides. In addition to the need for data to permit interspecies extrapolation, other biological effects should be considered also. When such data become available, a better basis for estimating body burdens and MPC's may evolve.

24. Comments on Intake Guides for Various Isotopes and Isotopic Mixtures of Uranium

Mary R. Ford

In the application of maximum permissible concentration (MPC) values, or intake guides, for uranium as provided by NCRP[1] and ICRP,[2,3] questions arise frequently concerning a "curie" of natural uranium which has a special definition in these recommendations. Other questions relate to the fact that limiting values for some of the isotopes are based on considerations of chemical toxicity, while for other isotopes it is the radiological hazard that is limiting; thus, uncertainties in the computation of values for mixtures of the isotopes are encountered. In this study simple formulas, additional MPC values, and explanations are given as an aid in resolving these and other similar problems.

A CURIE OF NATURAL URANIUM

In the NCRP and ICRP Recommendations,[1,2,3] a curie of natural uranium is considered to correspond to 3.7×10^{10} dis/sec from ^{238}U, 3.7×10^{10} dis/sec from ^{234}U, and 1.7×10^{9} dis/sec from ^{235}U. Even before 1962, when the curie was redefined as a unit of activity rather than a quantity of a radionuclide,[4] this definition was confusing to many because a curie by the usual definition corresponded to only 3.7×10^{10} dis/sec. Evidence of this confusion is present in the Recommendations themselves, where the number of disintegrations per

[1] "Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure," *Natl. Bur. Std. (U.S.), Handbook 69*, GPO, Washington, 1959.

[2] *Report of Committee II on Permissible Dose for Internal Radiation*, ICRP Publication 2, Pergamon, London, 1959.

[3] *Recommendations of the International Commission on Radiological Protection (as amended 1959 and revised 1962)*, ICRP Publication 6, Pergamon, London, 1964.

[4] "Radiation Quantities and Units," ICRU report 10a, *Natl. Bur. Std. (U.S.), Handbook 84*, GPO, Washington, 1962.

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nd from ^{235}U was listed incorrectly, but dif-
tly, in both the NCRP and ICRP texts. This
been corrected by errata, and fortunately it is of
demic interest only, since ^{235}U is relatively un-
rtant in computations involving the natural
ture.

The first appearance of the special curie in
mal dose recommendations was in the ICRP
commendations of 1955.[5] In that report the
RP was influenced by the English, who for many
rs had used such a definition in industrial work.
ce natural uranium is more than 99 wt % ^{238}U
d since chemical toxicity is the principal con-
rn, it was natural to neglect the ^{234}U and ^{235}U,
hich contribute very little to the total mass.
hus, the convention developed of computing the
ass of ^{238}U that would correspond to 1 curie of
ivity of ^{238}U and calling this amount 1 curie
atural uranium. Since a counter, in measuring
disintegrations from such a curie of natural
anium, would register about twice the usual
umber of disintegrations, the "double curie"
ame into use.

CHEMICAL TOXICITY VS RADIATION DAMAGE AS A BASIS FOR ESTIMATING PERMISSIBLE INTAKE VALUES

It is quite generally known that natural uranium is very poisonous to the body chemically and that kidney is the organ most severely injured.[6,7] Therefore, it is not surprising to kind kidney as an organ of reference for each of the uranium isotopes. As stated in the Recommendations, however, chemical toxicity is the limiting criterion only for the longer-lived isotopes; thus some of the kidney values are based on chemical toxicity and others on radioactivity, and no designations appear with the values as to which criterion was used in the estimations. Thus, confusion arises when intake guides for mixtures of the isotopes are needed, since values on both bases are needed in the computations.

MPC values computed by both criteria are given in Table 24.1. The pertinent biological and physical parameters as well as the computational methods of the NCRP and ICRP Recommendations were used in arriving at the values. Underscores indicate that these values, rounded to one significant place, are the ones appearing in the reports. As seen from this table, radiation damage results before the permissible chemical dose is reached in

[5] *Recommendations of the International Commission on Radiological Protection, Brit. J. Radiol. Suppl.* 6, Pergamon, London, 1955.

[6] Carl Voegtlin and H. C. Hodge, *Pharmacology and Toxicology of Uranium Compounds*, McGraw-Hill, New York, 1949.

[7] A. J. Luessenhop *et al.*, *Am. J. Roentgenol.* 79(1), 83–100 (January 1958).

Table 24.1. Uranium MPC Values (in μc/cc) with Kidney as the Organ of Reference

(appropriate for an exposure of 168 hr/wk)

Isotope	Based on Radioactivity		Based on Chemical Toxicity	
	$(MPC)_w$	$(MPC)_a$	$(MPC)_w$	$(MPC)_a$
^{230}U	$\underline{2.4 \times 10^{-5}}$	$\underline{10^{-10}}$	8.1×10^{5}	3.5
^{232}U	$\underline{4.4 \times 10^{-5}}$	$\underline{1.9 \times 10^{-10}}$	3.6×10^{2}	1.5×10^{-3}
^{233}U	$\underline{9.6 \times 10^{-5}}$	$\underline{4.2 \times 10^{-10}}$	0.16	7.0×10^{-7}
^{234}U	$\underline{9.8 \times 10^{-5}}$	$\underline{4.2 \times 10^{-10}}$	0.11	4.5×10^{-7}
^{235}U	10^{-4}	4.5×10^{-10}	$\underline{3.7 \times 10^{-5}}$	$\underline{1.6 \times 10^{-10}}$
^{236}U	$\underline{10^{-4}}$	$\underline{4.4 \times 10^{-10}}$	1.1×10^{-3}	4.7×10^{-9}
^{238}U	1.1×10^{-4}	4.8×10^{-10}	$\underline{5.9 \times 10^{-6}}$	$\underline{2.5 \times 10^{-11}}$
^{240}U	$\underline{0.12}$	$\underline{5.2 \times 10^{-7}}$	4.2×10^{8}	2.4×10^{3}

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every case except for the very long-lived isotopes, ^{235}U and ^{238}U.

MAXIMUM PERMISSIBLE EXPOSURE VALUES FOR MIXTURES OF THE URANIUM ISOTOPES

In the isotopic separation plants, various enrichment processes result in the production and handling of many different mixtures of the uranium isotopes, both depleted and enriched, and reactors operating at a given enrichment of uranium result in a constant change in the isotopic mixture. Thus, a simple method of estimating maximum permissible exposure values for these various mixtures is needed. Since analyses of the isotopic composition of uranium resulting from these processes are routinely made and recorded as the fraction by weight (f) of each isotope in the mixture, it is convenient to begin the computation with these fractions. Considering that 1 g of a mixture contains f_i grams of isotope i, the microcuries of isotope i per gram of mixture is

$$\mu_i = \frac{f_i \times 6.02 \times 10^{23} \times \lambda_i}{A_i \times 3.7 \times 10^4 \times 3600 \times 24},$$

where

A = atomic weight,
λ = decay constant (days),
$3600 \times 24 \times 3.7 \times 10^4$ = dis day^{-1} μc^{-1},

and the relative activities of the mixture, ξ_i, = $\mu_i / \Sigma \mu_i$.

Since a concentration $(MPC)_i$ $\mu c/cc$ of isotope i produces the limiting concentration, C, in the critical organ, it follows that 1 $\mu c/cc$ of isotope i produces the concentration $C/(MPC)_i$ in the critical organ. Hence,

$$(MPC)_{mix} \frac{\mu c}{cc} = \sum_i (MPC)_{mix}\, \xi_i \frac{\mu c}{cc} \text{ produces}$$

$$\frac{\sum (MPC)_{mix}\, \xi_i C}{(MPC)_i} = C$$

and

$$(MPC)_{mix} = \frac{1}{\sum_i [\xi_i/(MPC)_i]}.$$

Figure 24.1 shows $(MPC)_a$ values computed by this equation for mixtures of ^{234}U, ^{235}U, and ^{238}U, where the weight percentages of ^{234}U and ^{235}U were enriched or depleted by the same factor, with ^{238}U constituting the remaining percent by weight (i.e. mixtures which are somewhat analogous to those resulting from the gaseous diffusion process). These values, which are applicable for the 40-hr work week, are plotted in units of dis min^{-1} m^{-3}, and the point shows the position of natural uranium in the scheme. The curves are

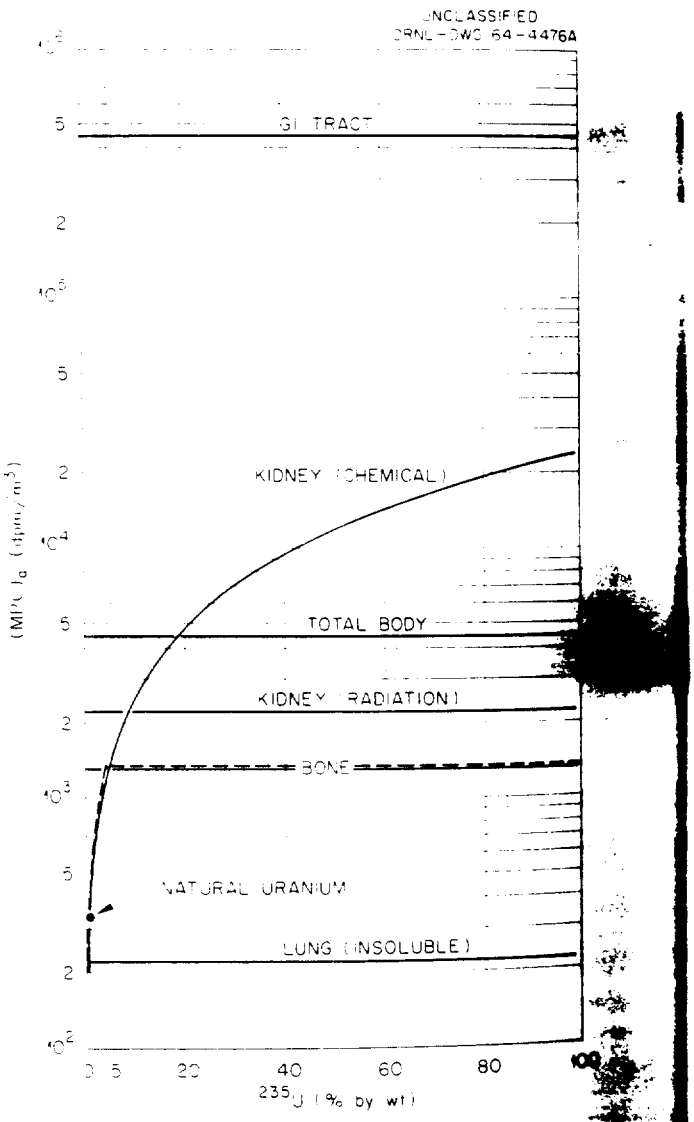

Fig. 24.1. MPC_a for Mixtures of ^{234}U, ^{235}U, and ^{238}U Appropriate for 40 hr/wk.

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beled according to the organ of reference used the computation. The curve labeled "lung" applies to insoluble uranium, and all the others apply to the soluble form. As indicated by portions of curves overlaid with a dashed line, chemical toxicity is limiting for soluble mixtures up to an enrichment of about 5% ^{235}U. For higher enrichments, radioactivity is limiting, with bone as the critical organ. It should be noted, too, that unlike enriched uranium, the $(MPC)_a$ for natural uranium is not acceptable for soluble depleted uranium.

This is more clearly discernible in Fig. 24.2, where the scale for mixtures up to 1% enrichment is enlarged. As shown in this figure, the limiting $(MPC)_a$ values for soluble material decrease with depletion to about one-half the natural uranium values. In Fig. 24.3 the same values are plotted in mass units ($\mu g/m^3$). In these units the MPC values based on radioactivity decrease appreciably with enrichment. For example, values for a 90% enrichment are less than those for natural uranium by about two orders of magnitude. Thus, it becomes apparent that controlling the dust

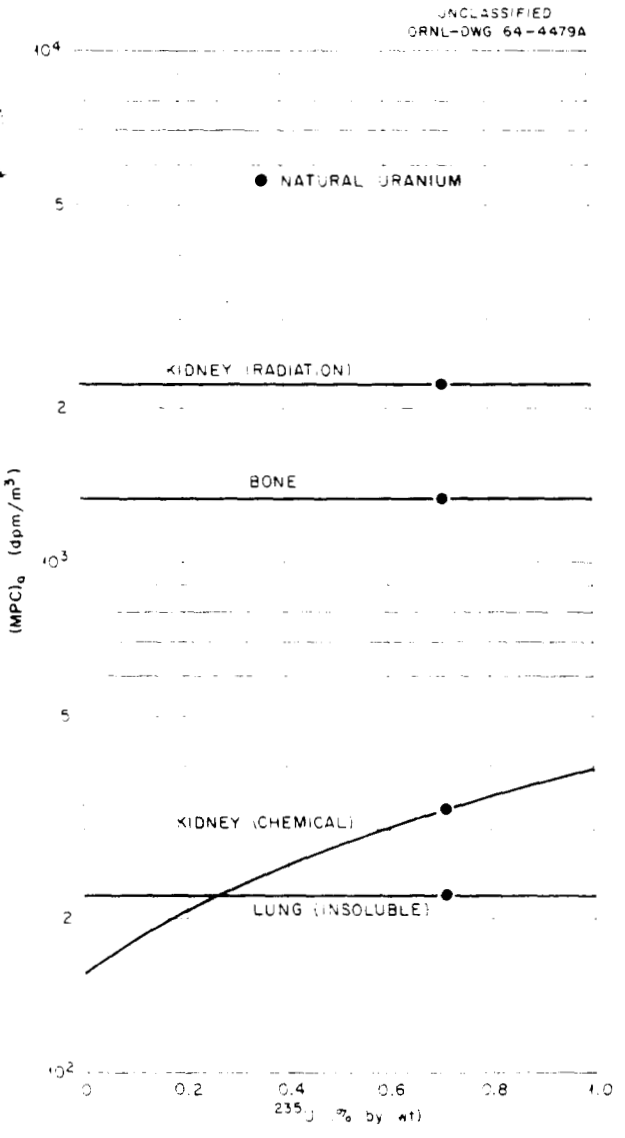

Fig. 24.2. MPC_a for Mixtures of ^{234}U, ^{235}U, and ^{238}U Appropriate for 40 hr/wk.

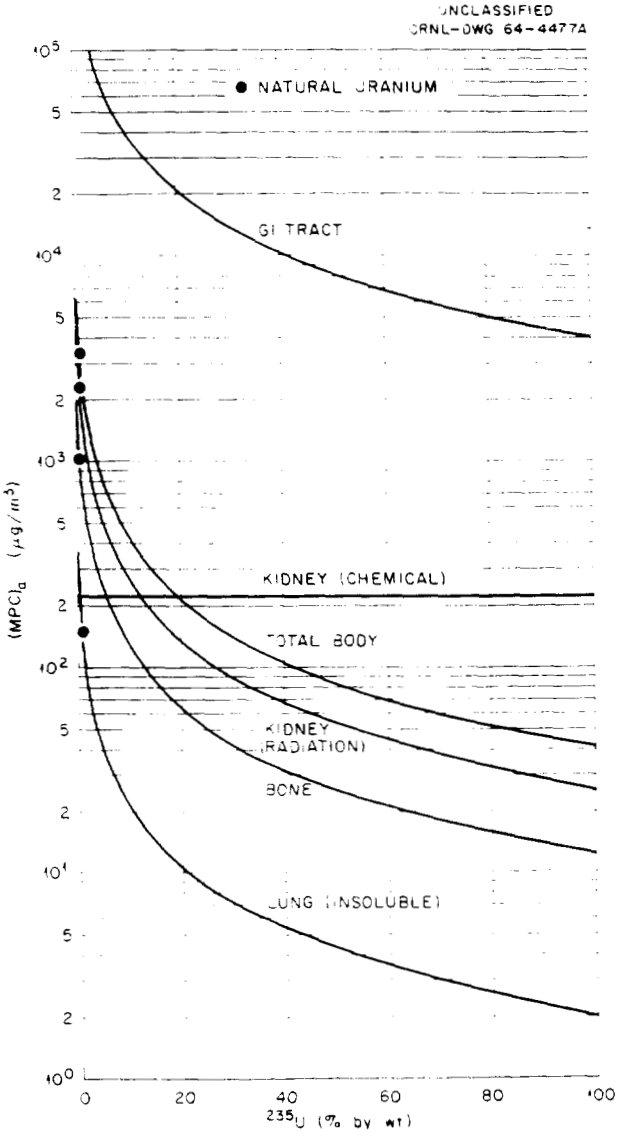

Fig. 24.3. MPC_a for Mixtures of ^{234}U, ^{235}U, and ^{238}U Appropriate for 40 hr/wk.

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hazard is a much greater problem for enriched uranium than for natural or depleted mixtures even though the limiting $(MPC)_a$ values for enrichments greater than about 5% are all equal in units of dis min^{-1} m^{-3}.

SINGLE INTAKE GUIDES FOR URANIUM

Thus far, these comments have dealt with uncertainties of long standing concerning uranium values. Reference is made now to the recommendation released a short time ago in ICRP Publication 6 (ref. 3) regarding single-intake values of uranium. In this recommendation it is stated that because of the chemical toxicity of natural uranium, ^{238}U, ^{236}U, or ^{235}U, the inhalation of soluble uranium of any isotopic composition should not exceed 2.5 mg in 1 day, or the ingestion averaged over 2 days should not exceed 150 mg. Since these single-intake values are stated in units of mg/day, the question will arise as to whether or not they may be used every day, that is, for continuous exposure, without exceeding the permissible values based on radioactivity. Figure 24.4 shows the limiting $(MPC)_a$ in mg/day for continuous exposure based on radioactivity for these isotopes and for the same mixtures used in the previous examples. The levels for the individual isotopes are shown along the right-hand ordinate, and levels for the mixtures are shown by the curve on which natural uranium appears. A comparison of these MPC values with 2.5 mg/day reveals that many of them lie considerably below this value. Thus, 2.5 mg/day would not be limiting for continuous exposure in every case. In like manner, it can be shown that one must be watchful that the radioactivity limit is not exceeded when applying the new single-intake value for ingestion.

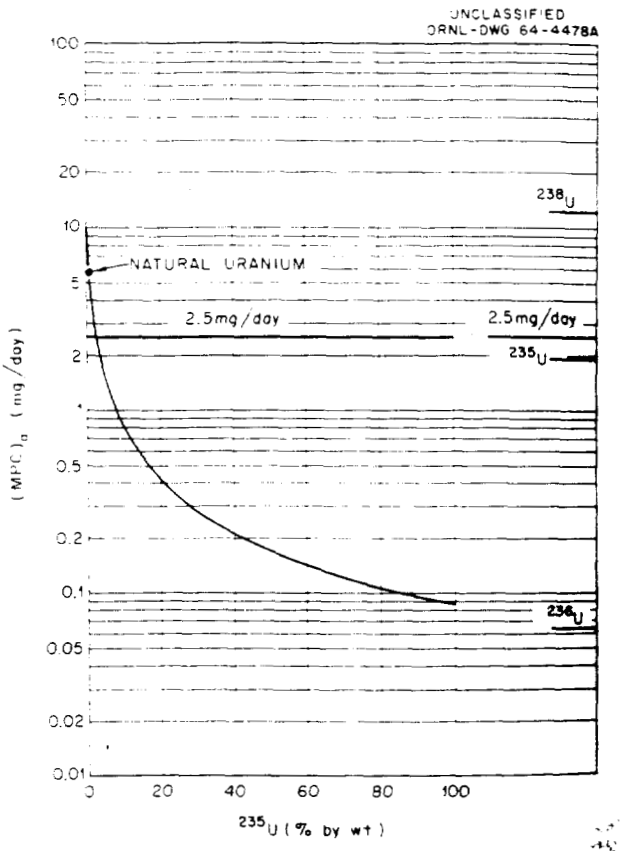

Fig. 24.4. MPC_a Based on Radioactivity Com[illegible] with 2.5 mg/day.

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25. On the Estimation of a Systemic Body Burden of Plutonium

W. S. Snyder

Employees exposed to plutonium generally excrete detectable amounts of the isotope in urine and feces. In fact, analysis of urine specimens is the more usual means of monitoring for possible exposure to ^{239}Pu. As the number of years of experience with processing of plutonium has increased, more and more cases have been found of employees who regularly show measurable amounts of ^{239}Pu in their urine. Often there is no known incident to which the exposure can be plausibly attributed, and the problem of estimating the employee's body burden is a very difficult one for the health physicist.

Computer codes have been designed[1,2] which provide an estimate of the intake and/or retention on the basis of the urinalysis data of the employee. These codes assume that the ^{239}Pu reaching blood is retained and excreted from the body according to a metabolic model devised by Langham[3] to fit the data on hospital patients given ^{239}Pu-citrate by intravenous injection. It is clear that all individuals do not follow the same metabolic model.[4] Only one human case has come to autopsy,[5] and, while the computer codes gave a rather accurate estimate in this case, the accuracy of these codes for estimation of body burden is not known in general.

During the past year the estimates of intake to blood as given by the code reported by Lawrence[1] have been used in an attempt to test the internal consistency of the model for metabolism of ^{239}Pu in the lung and also after reaching the blood. It is found that by postulating intakes via inhalation (which is considered to be the most likely mode of exposure in this case[5]), and assuming a constant half-time for elimination from the lung, the pattern of intake to blood indicated by Lawrence's code can be realized. Also, by selection of appropriate constants to represent the fraction of an intake to the blood that deposits in liver and in bone, the distribution found at autopsy can be obtained. It is noteworthy that this distribution can be obtained only by changing some of the currently accepted constants of the metabolic model as given by ICRP Publication 2 (ref. 6). In particular, a half-time in lung of about 30 days must be used, and approximately equal fractions of the intake to blood must deposit in liver and in bone to obtain agreement with the autopsy data. These values are rather at variance with those currently given in ICRP Publication 2, although it must be recognized that it has been known that the half-time in lung might vary considerably depending on the size of the aerosol as well as on its physical and chemical form. It is concluded that, in view of the many unknown factors involved in the exposure (particle size, chemical form, time of exposure, etc.), the observed distribution in the body is not known to be grossly at variance with the metabolic model used as the basis for estimation of MPC values by ICRP and NCRP. For the same reason, it cannot be concluded that the model is validated by these data. The study does suggest that certain parameters involved in the model probably should have somewhat different values, namely, that deposits in liver and in bone should be approximately equal and that in some cases the half-time for elimination from the lung may be substantially less than 1 year. The detailed analysis which is the basis for these suggestions is being published in *Health Physics* as part of the proceedings of the Symposium on Inhaled Radioactive Particles and Gases, held at Hanford Laboratories on May 4–6, 1964.

A second attempt to check on the accuracy of currently available computer codes for estimation

[1] J. N. P. Lawrence, *Health Phys.* 8(1), 61–66 (1962).

[2] W. S. Snyder, *Proceedings of 7th Annual Meeting on Bio-Assay and Analytical Chemistry, Held Oct. 12 and 13, 1961, at Argonne National Laboratory*, ANL-6637, p. 13.

[3] W. H. Langham *et al.*, *Distribution and Excretion of Plutonium Administered Intravenously to Man*, LA-1151 (Sept. 20, 1950).

[4] W. S. Snyder, *Health Phys.* 8(6), 767–72 (1962).

[5] H. Foreman, W. Moss, and W. Langham, *Health Phys.* 2(4), 326–33 (1960).

[6] *Report of Committee II on Permissible Dose for Internal Radiation (1959)*, ICRP Publication 2, Pergamon, London, 1959.

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of a systemic body burden of ^{239}Pu was undertaken, using data on dogs exposed intravenously and by inhalation to ^{239}Pu at Hanford Biology Laboratory. Bair *et al.*[7] injected three dogs with plutonium nitrate (0.14 *N* HNO_3), and the dogs were sacrificed ~30 days postinjection. The urinary excretion data are plotted in Fig. 25.1 together with a curve representing an average of the data. It is apparent that there are very wide fluctuations from day to day and that the dogs differ rather markedly so far as day-to-day excretion is concerned. Nevertheless, the code mentioned above was used to estimate the intake. In Fig. 25.2 the daily intakes as estimated by the code are shown in the form of a bar histogram. It is clear that, despite the erratic character of the excretion data, the code did correctly find that the intake

[7] W. J. Bair *et al.*, *Health Phys.* 8(6), 639–49 (1962).

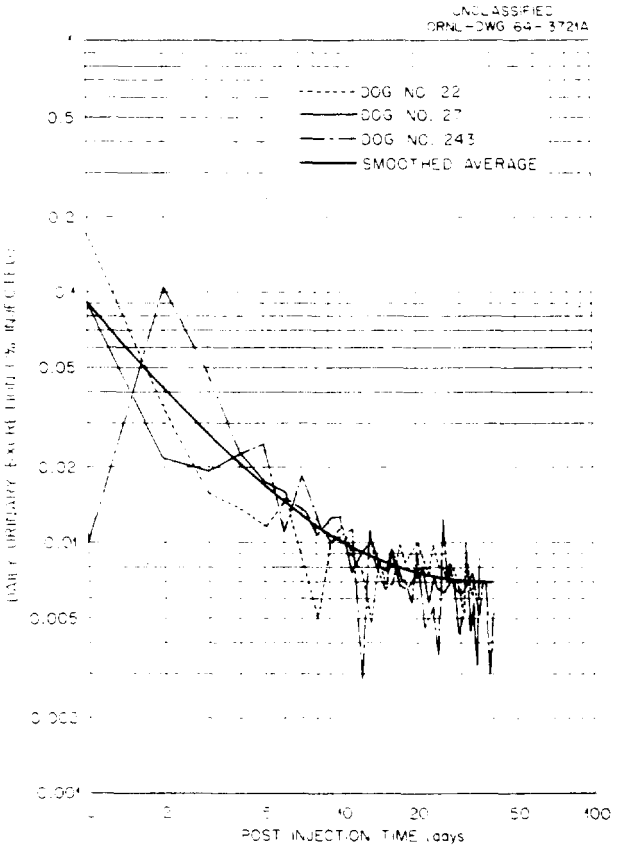

Fig. 25.1. Urinary Excretion Following Intravenous Injection of Plutonium Nitrate.

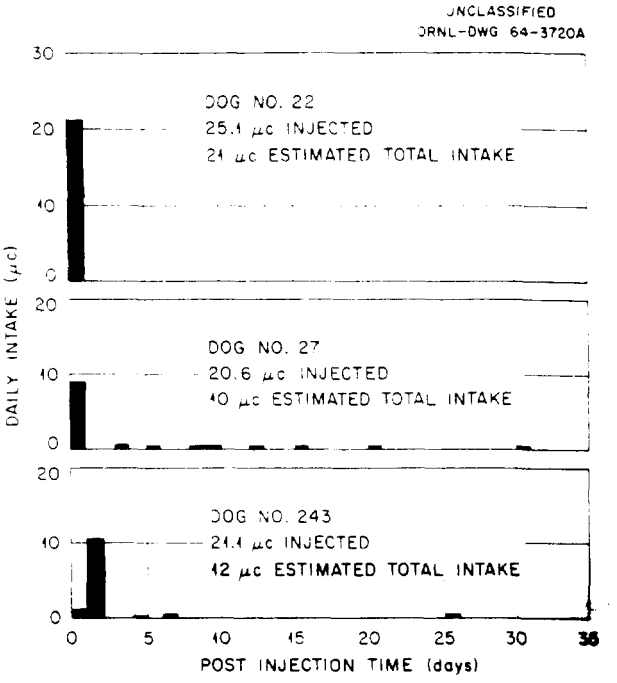

Fig. 25.2. Computer Estimates of Intake to Blood: Single Intravenous Injection Cases.

was confined largely to the first day or two. The small intakes to blood on succeeding days are the result of unusually high fluctuations of the excretion data. Thus, the code did give a qualitatively correct interpretation so far as time of intake is concerned. The estimated total intake is shown together with the injected amount, and it is evident that the estimates are within a factor of 2. In view of the erratic character of the data, this is considered to be encouraging.

Bair *et al.*[7] also exposed dogs to plutonium nitrate (0.14 *N* HNO_3) by inhalation. The urinary excretion data of those dogs exposed to the nitrate were analyzed by the code also, and the estimated intakes to blood are shown in Fig. 25.3. Although the time sequence is erratic, significant intakes to blood do seem to occur over a succession of days and tend to decrease with time. Thus, the qualitative aspects are as we might expect. However, the large number of days of no intake undoubtedly is an artifact produced by sporadic high excretions early in the period of observation. The estimated total intake to blood is shown also on Fig. 25.3, together with the difference of lung deposition and

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terminal lung burden as estimated by the experimenters. Again, the results generally agree within a factor of 2.

It seems clear that the present methods cannot be relied upon to give more than a very rough indication of the intake to blood. There are a number of possible improvements that suggest themselves. Some preliminary smoothing of data, perhaps by the computer, might reduce the error. However, it seems likely that marked improvements will come only with a better understanding of the metabolic model and of the probable source of the wide individual differences and day-to-day fluctuations of the excretion data. It would seem that carefully controlled experiments might help in understanding these difficulties and that, even if the experiments did not enable us to overcome these difficulties, we might in this way obtain a firmer estimate of the uncertainty of our method of estimation.

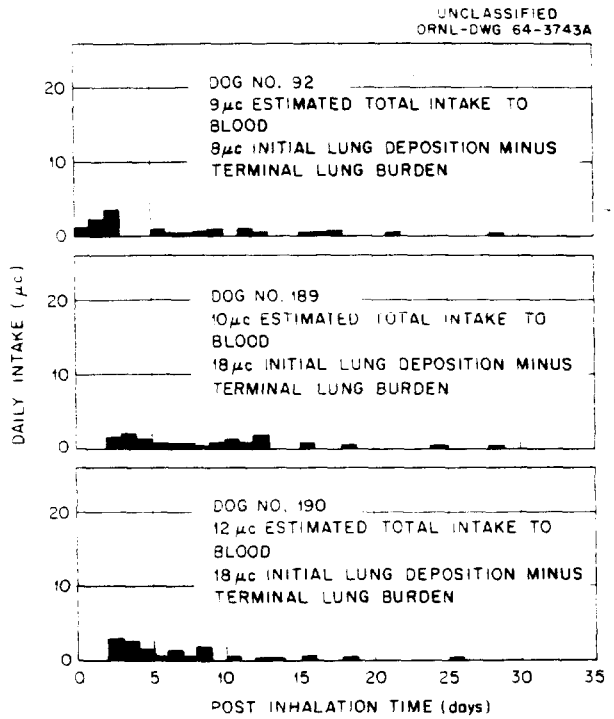

Fig. 25.3. Computer Estimates of Intake to Blood: Inhalation Cases.

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INTERNATIONAL SYMPOSIUM ON SURFACE CONTAMINATION

B. R. Fish (Chairman)
G. W. Royster, Jr. (Treasurer)
D. D. Cowen (Local Arrangements Chairman)
Blanche Reed (Ladies Activities)

The first International Symposium on Surface Contamination was held in Gatlinburg, Tennessee, June 8–12, 1964. The meeting was sponsored jointly by the Oak Ridge National Laboratory, the American Association for Contamination Control, and the Health Physics Society.

There were 54 papers presented in 12 sessions. Four sessions were devoted to fundamental research and development in the fields of aerosol physics, surfaces, adhesion-redispersion, and transport-deposition. Other administrative and technical problems were discussed in sessions on radioactive surface contamination control criteria, measurement techniques, environmental control of surface contamination, radioactive contamination control applications, dissemination of airborne microorganisms, biological and chemical surface contamination, insurance and economics, and decontamination.

The meeting was attended by 222 persons; 37 were from outside the United States. There were 14 attendees from France; 9 from the United Kingdom; 3 each from Canada, Germany, and Japan; 2 from Belgium; and 1 each from Australia, Austria, and Venezuela.

27. Applied Internal Dosimetry

B. R. Fish

ORNL IN VIVO GAMMA-RAY SPECTROMETRY FACILITY

P. E. Brown, L. B. Farabee
G. R. Patterson, Jr., Rose J. McBath
W. H. Wilkie, Jr.
L. S. Barden[1], D. L. Mason[3]
J. H. Dobkins[2], J. K. Eddy[4]

The ORNL in vivo Gamma-Ray Spectrometer (IVGS) Facility continues to be used primarily as a monitoring device to aid in the detection and measurement of internal exposures. Additional improvements in instrumentation, scheduling, and the operating program have made it possible to increase the rate of routine counting for monitoring purposes beyond the increase mentioned in the previous report.[5] Emphasis is placed on the routine counting of persons suspected of having experienced an exposure or of having a potential for exposure and on obtaining base-line counts for persons prior to their entering new work involving potential internal exposure. The further increase in rate of counting people has made it possible to expand the base-line counting and obtain counts on essentially all persons with even a remote chance for exposure. During the past year, 1370 persons were counted for a total of 1568 human counts; 1159 of the counts or approximately 74% showed a normal human spectrum. By way of comparison, the totals for the period July 31, 1961, to July 31, 1962, were 142 human

[1]ORINS trainee, Hendrix College, Conway, Ark.

[2]Temporary summer employee, North Carolina State College, Raleigh.

[3]Temporary summer employee, Austin Peay State College, Clarksville, Tenn.

[4]Temporary summer employee, West Virginia University, Morgantown.

[5]B. R. Fish *et al.*, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, p. 194.

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counts on 102 persons; the total for the period ending May 31, 1963, was 492 human counts. Table 27.1 summarizes the data for those cases in which measurable amounts of internal radioactive contamination were observed.

The current rate of counting is about 130 persons per month, excluding instrumentation downtime. Theoretically, one could count about 170 persons per month. This represents practically the maximum rate attainable with the present equipment operating 40 hr per week. We do not anticipate operating more than one shift per day, except when it is necessary to count persons regularly working only one of the night shifts. The increase of fiscal-year 1964 counting to more than three times the 1963 counts represents the realization of a number of improvements in equipment and operating program. Listed roughly in order of the favorable impact they have had, these improvements include installation of a second 512-channel analyzer, a paper-tape-to-typewriter data converter, the rolling scan bed, a new system for scheduling persons to be counted, and the computer program for data analysis. The rolling scan bed and the computer program were mentioned in some detail in the last report in this series;[5] the other improvements were implemented during the present period.

Table 27.1. Measurable Radioactivity Found in Routine Whole-Body Monitoring Program, June 1, 1963–June 30, 1964

Isotope	Individuals Counted	Maximum Amount (μc)	MPBB (μc)
^{24}Na	1	*a*	
^{51}Cr	10	0.042	800
^{58}Co	29	0.038	30
^{59}Fe	6	0.015	20
^{60}Co	34	0.220	10
^{64}Co	1	<0.005	10
^{65}Zn	32	0.016	60
^{75}Se	12	0.250	90
^{90}Sr-^{90}Y	17	4.500	2
^{95}Zr-^{95}Nb	67	0.047	20
^{106}Ru-^{106}Rh	47	0.047	3
^{125}Sb	43	0.018	40
^{131}I	29	0.073	0.7
^{137}Cs	160	0.310	30
^{144}Ce-^{144}Pr	17	≦0.007	5
^{155}Eu	3	Trace	70
^{198}Au	1	*a*	
^{226}Ra	3	<0.005	0.1
U (enriched)	1	Trace	0.06

[a]Skin contamination.

IN VIVO DETECTION AND MEASUREMENT OF ^{90}Sr-^{90}Y INTERNAL CONTAMINATION

During this report period, the ORNL IVGS Facility was used to estimate the ^{90}Sr-^{90}Y body burden of several ORNL employees who had been involved in airborne particulate contamination incidents. In two of the incidents the contamination was $^{90}SrTiO_3$; in the other incident the contaminant was $^{90}SrCO_3$. The practical limit of detection of ^{90}Sr for the ORNL in vivo counter is approximately one-tenth of the maximum permissible body burden (MPBB) or ~0.20 μc. In the observed cases the bremsstrahlung continuum was readily discernible at levels estimated to be around 5% of the MPBB. Of course, at these relatively low levels, estimation of ^{90}Sr internal contamination by in vivo counting is subject to large uncertainties, particularly when other contaminants may be present. In the first incident involving $^{90}SrTiO_3$, three persons were found to have obvious indications of bremsstrahlung in their spectra. Initial estimates of their whole-body strontium burden, based on the application of a strontium-in-skeleton calibration to whole-body scan counts, suggested 0.8, 0.9, and 6.2 μc for cases A, B, and C, respectively, as of three days postexposure. Figure 27.1 is a profile scan showing the relative counting rate as the body of employee C passed under the crystal. Note that even at this late date the largest fraction of the

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deposit is in the lower abdomen. In view of this fact, the estimates were revised by using a combined GI tract–skeleton calibration. Revised estimates based on this calibration were 0.46, 0.55, and 4.5 μc for the three men. Figure 27.2 shows the net spectrum from 0 to 2.04 Mev for the initial scan count of case C. After much of the abdominal deposit was eliminated, the chest counts taken with a lead-collimated 8 × 4 in. crystal were felt to provide a more accurate estimate of the strontium burden. Figure 27.3 shows the in vivo estimates of chest burden for case C up through the most recent count, 169 days post-exposure.

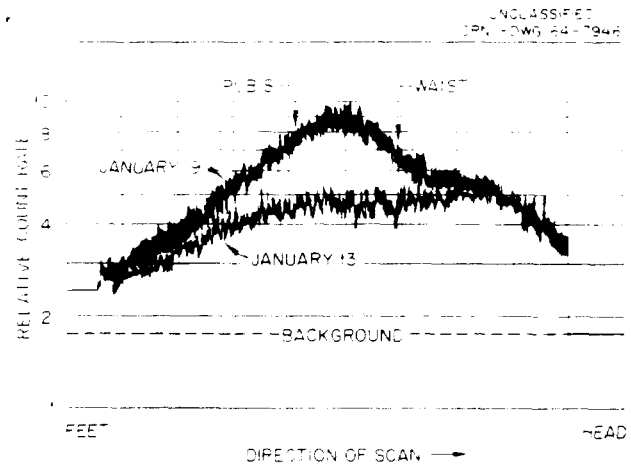

Fig. 27.1. Profile Scans of Two Whole Body Counts ($^{90}SrTiO_3$ exposure case C).

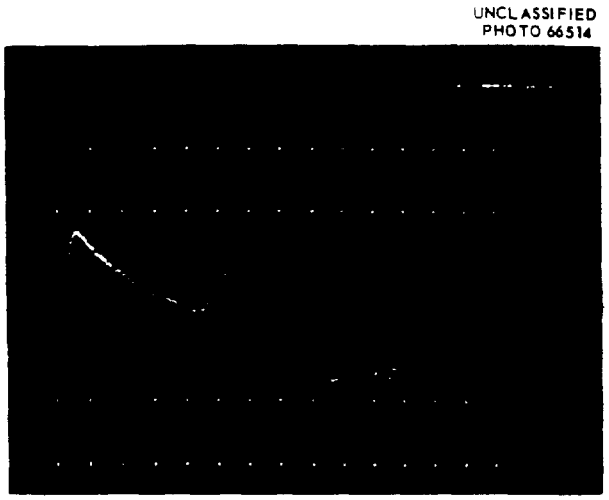

Fig. 27.2. Net Spectrum Whole Body Scan ^{90}Sr Inhalation Exposure.

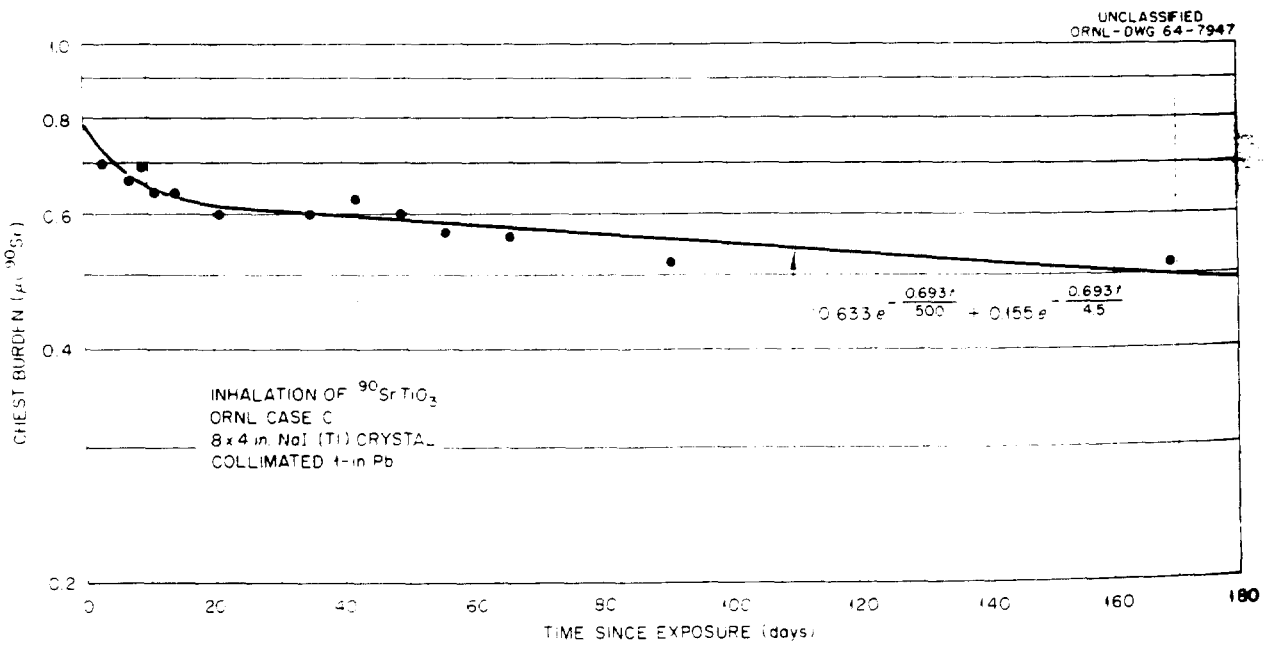

Fig. 27.3. Strontium-90–Yttrium-90 Chest Burden.

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In both of the other incidents involving exposures to strontium compounds, the whole-body scan counts detected measurable amounts of ^{90}Sr, 0.50 μc in the $^{90}SrTiO_3$ incident and 0.60 μc in the $^{90}SrCO_3$ incident. However, in both cases the chest counts with the collimated crystal showed only small indications of bremstrahlung, whereas profile scans show that the bulk of the deposit is in the abdominal region. These cases have adequately demonstrated that in vivo counting can give a rapid and positive indication of a serious intake of ^{90}Sr.

COMPUTER PROGRAM FOR IN VIVO COUNTING DATA

G. R. Patterson, Jr. M. T. Harkrider[6]
Barbara A. Flores[6] G. S. McNeilly

Program WBC,[7] designed to analyze routine whole-body count data on the CDC 1604 computer, has been in limited use since March 1963. Some few changes have been made to correct minor flaws that became apparent or to make the program more flexible; for instance, it is no longer necessary to separate whole-body scan counts and chest counts and process them as separate groups of data. By providing rapid analysis of routine body count data with accurate estimates of total-body potassium and ^{137}Cs, the computer can handle approximately 75% of the data analysis load.

Computer plotting of raw and smoothed spectra has been discontinued as uneconomical and not significantly better than a Polaroid photograph of the oscilloscope spectrum display. The practice of circulating a written weekly report of in vivo counting activities and results was begun in March 1963. In the fall of 1963 it was decided that, with a slight change in format, the IBM cards that identify each punched paper tape for the computer program could be utilized to produce a machine tabulated weekly report of in vivo counts. M. T. Harkrider wrote the necessary computer program, and report No. 1 for the first week of January 1964 was the first in vivo counting weekly report that was machine tabulated.

[6]Mathematics Division.

[7]B. R. Fish *et al.*, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, pp. 202–4.

DEVELOPMENTS IN THE ORNL IN VIVO GAMMA-RAY SPECTROMETRY FACILITY

W. H. Wilkie, Jr.

During July and August 1963, a second 512-channel analyzer was installed at the IVGS Facility. In addition, new preamplifier cables were run from the iron room to the control room, and the divider network was rewired. Both analyzers and the associated electronic equipment were mounted in an integrated control complex of matching cabinets. A tape-to-typewriter data converter was installed, permitting direct coupling of the paper-tape reader to the typewriter. A master switching panel was installed which permits quick switching of any of the data readout equipment to either one of the two analyzers. These equipment changes have made it possible to use the facility more efficiently by reducing maintenance downtime and by providing off-line equipment for data processing.

The Saran webbing of the rolling scan bed was replaced by a stainless steel top and a foam mattress.

Various methods for calibrating whole-body counters are being studied. Differences between the results of calibrations using several phantom-source configurations and the actual situations they approximate are being examined. Preliminary data indicate that a whole-body scan calibration using a simple point source of ^{137}Cs positioned in the central portion of the abdomen of a bottle mannikin is comparable with a calibration which a ^{137}Cs solution is uniformly distribu in the same type of phantom. Positioning of the point source in the single-source method is critical, and for this purpose a bottle mannikin was modified to allow reproducible positioning of the source. Neither method of calibration is expected to represent accurately all in vivo situations; in practice, distributions of source activity, and hence the overall detection efficiencies, vary considerably from case to case. It has been found also that nearly identical background spectra are obtained whether one uses a single container of distilled, demineralized water (size of the container determined empirically) or a multibottle phantom laid out under the crystal like a human subject. Here again, background for a human count is only an approximation of the actual perturbation of the radiation field as a result of scatter and absorption by the human body.

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IODINE-131 IN MILK AND IN CATTLE THYROIDS

P. E. Brown L. B. Farabee

In September 1963 the program for monitoring milk and cattle thyroids as indicators of the distribution of ^{131}I in this general geographical area was turned over to the Environmental Monitoring Section of Applied Health Physics. The program had been developed by the Health Physics Technology Section and tested during 1961–1963.[8–10] An average of ten thyroid samples per week were tested during most of this time, and one or more milk samples were tested weekly for over a year. Under the Health Physics Technology program, the prepared samples had been counted by using either a 4 × 2 or a 3 × 3 in. NaI(Tl) crystal and a 200-channel analyzer, the original in vivo counting equipment. Transfer of the program to Environmental Monitoring necessitated revision of the techniques to make them compatible with presently available equipment – a 4 × 4 in. crystal and a single-channel analyzer. Detection efficiency for counting thyroid samples with the single-channel system was found to give a lower detection limit of 10 pc per 20 g of thyroid at the 95% confidence level.

BIOASSAY AND ENVIRONMENTAL MONITORING

L. B. Farabee

During the past year, one member of the Health Physics Technology Section participated in an International Atomic Energy Agency (IAEA) program aimed at improving the reliability and comparability of the results of low-level activity measurements in environmental samples. A panel of representatives from 20 countries and worldwide organizations met in Vienna, Austria, October 28–31, 1963, for discussion and planning of an interlaboratory comparison program among nations associated with the IAEA. Representatives from the United States were: A. S. Goldin, U.S. Public Health Service, Bethesda, Maryland; M. F. Milligan, Los Alamos Scientific Laboratory; and L. B. Farabee, Oak Ridge National Laboratory. A paper entitled "Review of Existing Intercomparison and Interlaboratory Schemes in the United States" was presented by L. B. Farabee.

The need for greater accuracy in analytical data on the measurement of harmful radionuclides in the food supply of man was stressed, and guidelines for setting up an interlaboratory comparison program were discussed. Comprehensive reviews of interlaboratory comparison programs used in many countries were discussed by panel members. Statistical considerations for collection and analysis of environmental samples as a means for improvement in quality control were of interest. Although many radionuclides and many foods were suggested for use in the IAEA intercomparison program, it was expedient to limit the original program to only the more hazardous long-lived radionuclides. Therefore ^{90}Sr, ^{137}Cs, and ^{226}Ra were selected as having the most universal interest at present. A review of the panel discussion may be found in ORNL-CF-64-1-83, dated January 14, 1964.

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[8] B. R. Fish *et al.*, *Health Phys. Div. Ann. Progr. Rept. July 31, 1961*, ORNL-3189, pp. 232–34.

[9] B. R. Fish *et al.*, *Health Phys. Div. Ann. Progr. Rept. July 31, 1962*, ORNL-3347, pp. 148–52.

[10] B. R. Fish *et al.*, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, p. 206.

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