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Summary

PART I. RADIOACTIVE WASTE DISPOSAL

1. Fate of Trace Elements and Radionuclides in Terrestrial Environment

Fertilized and unfertilized soils, eroded material from unfertilized soils, river sediment, and sediment from a man-made pond were analyzed for their content and forms of phosphorus. Of the sought-for forms – free aluminum, free iron, calcium, organic, and occluded – the fertilized soils were characterized by the presence of all five forms. Sediment from the pond which had received commercial fertilizer differed from the fertilized soils by the very low content of both free aluminum and iron phosphates. The river sediments were characterized by low content of free aluminum phosphate. Unfertilized soil and material eroded from the unfertilized soil were characterized by the low content of calcium phosphate. Results suggest that calcium phosphate is associated with soils and sediments receiving commercial fertilizers and further suggest that eroding material can transport significant quantities of phosphate from soils which have not received commercial fertilizer.

An abundant growth of algae and wild celery, a rooted aquatic plant, occurred in a man-made pond after a generous application of commercial fertilizer. Subsequent reduction of the concentration of phosphate resulted in a decrease in the growth of both wild celery and algae. Results suggest that the form and concentration of phosphate in sediments play a key role in influencing the growth of rooted aquatic plants.

2. Disposal by Hydraulic Fracturing

A considerable amount of information on the underground behavior of hydraulically induced fractures is contained in the pressure histories of the injection and shut-in phases of the operation, particularly if a fluid like water is injected rather than a slurry. The analyses presented in this section represent an effort to develop a basis for interpreting such information in terms of the fracture orientation.

The first part is a theoretical and empirical evaluation of the pressure variations measured in the injection well during pumping. This analysis includes components due to turbulence at the entrance to the fracture, the viscous losses during laminar flow for circular and noncircular fractures, and the effects of vertical gradients, both in the confining rock stresses due to tectonism and in the fracture resulting from changes in elevation. These components were synthesized in various combinations to provide a predicted pressure history for almost any possible (idealized) fracture configuration.

The second part is an empirical graphic analysis of the pressure decay after the water injection was finished and the well shut in. This drop in pressure was very rapid at first but then went more and more slowly, so that after some 70 days the pressure was still over 200 psi. The drop in pressure is presumably principally due to two factors: (1) the continued lateral extension of the fracture due to recoil of the deformed rock cover near the injection well and (2) the water leaking out of the fracture. The first of these mechanisms will stop when the walls of the fracture make contact, which, if the fracture is horizontal, will happen when the pressure reaches that due to the weight of the overlying rock. A discontinuity in the time-pressure curve coming at this pressure would, therefore, be an indication that the fracture was horizontal. A log-log plot of the rate of change of pressure as a function of time did indeed show a change in slope at this critical pressure, suggesting that such an analysis of the pressure decay

data may be of value in determining the orientation of a water-filled fracture.

The so-called rock cover monitoring wells were constructed by cementing in a steel casing to a depth of 550 ft and then drilling an open hole to 650 ft. It was noted that, during a waste injection, the water level in these wells (which had been left full of water) in some cases rose, while, at the same time, it fell in others. An obvious explanation is that, where the grout-filled fracture passed underneath one of the wells, the shale adjacent to the open-hole section of the well was compressed and water was squeezed into the well. In the case of wells located in a direction other than that taken by the grout sheet, a shearing force would be developed in the shale adjacent to the open-hole section of the well, and the vertical load would be reduced, sucking a small amount of water into the shale. By equipping the rock cover monitoring wells with pressure gages and taking readings at intervals during an injection, it was possible to get indications of the direction taken by the fracture; however, these indications have not yet been correlated with the data from the gamma-ray logging of the observation wells.

3. Disposal in Natural Salt Formations

As reported previously (ORNL-4168), the radioactive phase of the demonstration disposal of high-level radioactive wastes was completed by the end of June 1967. By the end of December 1967, the remainder of the mine experiments had been terminated, and preparations were made to place the Carey Salt Company's Lyons, Kansas, mine on a standby basis, beginning February 1968. Arrangements have been made for periodic service of the equipment left on standby and for periodic collection of selected items of data.

During the 19-month operation of the radioactive demonstration, the average dose to the salt over the length of the main array holes was about 8×10^8 rads, and the peak dose was about 10^9 rads. The dose in the array in the old mine floor was about 10^8 rads.

Additional tests were run to determine the effects of water on container corrosion. The simulated-waste container cans have been removed for evaluation of the corroded conditions observed.

The laboratory pillar-model tests have been completed, and the effects of various variables have been analyzed. A completion report (ORNL-TM-2102) has been published.

The study of the economics of disposal in salt has been completed and was published as ORNL-3358. Using present worth considerations and a 4% interest rate, the money to be set aside annually at the time of waste generation ranged from about $2.4 million to $2.6 million for one-year-old waste to about $0.3 million to $0.4 million for waste 30 years old at the time of disposal. In terms of cost per kilowatt-hour of electricity generated, this corresponds to about 18 to 20×10^{-3} and 2 to 3×10^{-3} mill respectively.

To summarize, it may be said that most of the major technical problems regarding disposal in salt have been resolved. The results have been most encouraging. The feasibility and safety of handling highly radioactive materials in an underground environment have been demonstrated. The stability of the salt under the effects of heat and radiation was shown, as well as the capability of solving minor structural problems by standard mining techniques. The data obtained on the creep and plastic flow characteristics of the salt should make it possible to arrive at a suitable mine design for a disposal facility. A study of the economics of disposal in salt mines indicates that this method will be compatible with competitive nuclear power. All these items combined lead one to the conclusion that burial in salt mines is one of the better methods, if not the best, for the ultimate disposal of high-level solidified wastes.

Based on this conclusion, an examination is now being made of the desirability of establishing an actual prototype disposal facility in the immediate future.

4. Engineering, Economic, and Safety Evaluations

A hypothetical 6-ton/day nuclear fuel reprocessing plant was sited at ORNL, and estimates were made of dose equivalents to a "standard man" working at the Oak Ridge Gaseous Diffusion Plant and residing in Oak Ridge for assumed release conditions of ^{85}Kr and ^{3}H. Modes of exposure considered were ingestion, inhalation, submersion in contaminated air and water, and irradiation from a contaminated surface.

Tritium released to the environment will be expected to be predominantly in the hydrosphere and

will behave much the same as ordinary water. Although the oceans and seas represent the largest reservoirs for dilution of tritium, their long turnover times, even for the surface 75 m which mixes rather rapidly, will prevent attainment of uniform concentration throughout the circulating portion of the hydrosphere.

Laboratory studies have been made on the dissolution and movement of strontium that has been coprecipitated with calcium carbonate in mineral exchange systems. Mineral exchangers have substantial capacities for hydrogen ion buffering. In order to dislodge strontium from such a system, it was found necessary to add sufficient acid to overcome this buffering capacity. Hence, a sudden change in groundwater pH with a coincident release of ^{90}Sr in the waste seepage pit area is highly unlikely.

As a part of the long-range waste management study, projections were made of the nuclear power requirements and the reactor mix that may accommodate these requirements. Estimates were made of the volumes of waste and fission products accumulated to year 2000.

Factors that need to be considered for the long-term, on-site storage of solid high-level wastes are enumerated and discussed in a qualitative manner. On the basis of present information, salt offers the optimum combination of properties for a disposal formation. However, it seems possible that other formations may also be suitable.

A study has been completed of the potential disposal of brine effluents from inland desalting plants by deep-well injection. Present practice, costs, and requirements for a disposal operation were reviewed.

5. Earthquakes and Reactor Safety

A new technique, utilizing microscopic measurements of rock specimens, is being developed for utilization in reactor site evaluations. This technique is based on the premise that certain elastic properties of rocks that are indicative of present stress conditions can be determined and measured in the laboratory.

6. Dose-Estimation Studies Related to Peaceful Uses of Nuclear Explosives

The information requirements for estimating a population's radiation dose resulting from environmental releases of radioactive material have been categorized. Models have been developed and programmed to estimate expected radiation doses to populations in various hypothetical exposure situations related to peaceful uses of nuclear explosives. Radiation doses received from external and internal sources of exposure are estimated with computer codes entitled EXREM and INREM respectively. The modes of exposure considered for external sources are: submersion in contaminated air, submersion in contaminated water, and exposure to a contaminated surface. Ingestion and inhalation are the modes of exposure considered for internal sources. Steps are outlined for utilizing dose estimates obtained with these codes to identify the critical radionuclides in a given exposure situation. A study to determine the radiological-safety feasibility of using nuclear explosives to excavate a sea-level canal across the isthmus of Central America is under way, and those elements whose radioactive isotopes are of greatest radiological importance to that study have been tentatively identified on the basis of results obtained by completing the second step of the four-step critical radionuclide identification procedure. Use of the INREM code is demonstrated in detail with an example estimating the dose received by a population exposed to an atmosphere containing tritiated water vapor. The example illustrates evaluation of the age-dependent parameters and the influence of the age of those exposed upon the resulting dose estimate. The maximum variation among age groups, in this case, is a factor of 1.5.

7. Related Cooperative Projects

Representatives of other agencies continued to participate in the Radioactive Waste Disposal Section's studies. Two alien guests were in residence during the year. One member served as assistant news editor for the United States for *Health Physics* and as an assistant editor of *Nuclear Safety*. One member of the section participated in cooperative programs with the University of Tennessee and Vanderbilt University. Two members of the section participated part time at the Nuclear Safety Information Center.

PART II. RADIATION ECOLOGY

8. Responses of Animal Populations to Ionizing Radiation

Data from hematological and radiation effects studies on mammals were utilized for comparisons of radiation mortality in the hemopoietic death range as typified by $LD_{50(30\text{-}60\ days)}$ observations. An index was obtained by multiplying average interphase chromosome volume (ICV) of the lymphocyte by the elimination constant of ^{59}Fe (λ_b) for each species. This index gives close predictions of LD_{50} (rads) for the species for which data are available. Presently the relationship ICV (Fe λ_b) is linear, but it is anticipated that it will be sigmoid when data are obtained for the extremely radiosensitive and radioresistant species. An interesting observation during hematological studies of cotton rats was the rapid recovery of WBC counts after irradiation of individuals infected with a blood parasite, *Hepatozoon* sp.

It has been hypothesized that the increased life expectancy of animals exposed to low radiation doses was due to a reduction in the respiration rate. This hypothesis was tested by measuring the continuous oxygen consumption of crickets (*Achera domesticus*) exposed to doses of gamma radiation from 0 to 32 kilorads. A variety of statistical tests failed to demonstrate differences in oxygen consumption among the irradiated crickets. Thus the increased life expectancy of crickets exposed to low dose levels cannot be explained by a reduction in respiration rate.

Responses of honeybees to irradiation could be an important aspect of postattack recovery of agricultural landscapes. Irradiation experiments of samples of about 150 bees showed a reduction of 22% in life-span of bees exposed to 5000 rads of ^{60}Co gamma irradiation. On this basis five entire colonies of bees were exposed to 5000 rads of ^{60}Co irradiation in a large animal irradiator to determine effects of irradiation on pollen collection. Surprisingly, the irradiated colonies ceased to function as social units within two weeks. Pollen collections in the irradiated colonies were 61% of the controls the week before irradiation, but in the week following exposure this dropped to 28%; two weeks post irradiation, pollen collections were 7% of controls, and in the third week no pollen was collected. At the end of three weeks about 200 of the 87,000 irradiated adult bees survived.

The interaction of radiation and competition between two closely related species, *Drosophila melanogaster* and *D. simulans*, was tested in controlled laboratory populations. Although adults of *Drosophila* can survive several kilorads of gamma radiation and populations are easily maintained while receiving an acute or chronic dose of 2000 r every generation, a dose of 1000 rads given every three weeks to adult male *D. melanogaster* was sufficient to eliminate this species when it was in competition with *D. simulans*. In the control population *D. melanogaster* quickly became the most abundant species. These results illustrate the effects of competition interacting with an additional radiation stress.

The *Gambusia* population in White Oak Lake is exposed to a chronic radiation dose of about 11 rads/day. To determine whether White Oak Lake fish had acquired radioresistance, the $LD_{50/30}$ for these fish was compared with *Gambusia* from a nonirradiated population. The $LD_{50/30}$ of both populations was slightly greater than 3000 rads and not significantly different from each other. *Gambusia* as a species appears to be relatively radioresistant.

9. Responses of Plants to Ionizing Radiation

Studies of the effects of external beta radiation on higher plants were initiated to determine radiosensitivities of native plant species and to provide estimates of the ecological effects of postattack fallout on plants. Fallout simulant, consisting of sand (44 to 88 μ in diameter) containing approximately 5 nc of ^{90}Sr per particle, was applied to various organs of white pine, cottonwood, cocklebur, and bean plants. In all cases plant parts which received contact exposures were killed, while other plant organs survived with varying degrees of biological damage depending upon distance from the contaminated area of the plant. Contaminated white pine terminal buds receiving contact doses of 54 to 310 rads/hr started turning brown within five days and by ten days were dehydrated and brittle. Shoot growth from buds below contaminated buds received doses varying between 70 and 200 millirads/hr and showed an average growth reduction of 47% at 30 days. Browning of the

foliage occurred much earlier from the beta radiation than expected on the basis of previous experience with plant exposures to gamma radiation. Furthermore, translocation of ^{90}Sr to other parts of the plant occurred, and in the case of beans the fruit contained 0.4 to 5.0 nc of ^{90}Sr per gram.

Aquatic microcosms containing various combinations of physical components (soil, water, and container surface) and biological components (*Physa heterostropha*, *Najas flexilis*, and *Elodea canadensis*) were used to determine the effect of increased biotic complexity on ^{60}Co and ^{137}Cs cycling parameters and to test the stability of the cycling parameters under exposure to sublethal and lethal doses of fast-neutron irradiation. Generally, cycling parameters of ^{137}Cs and ^{60}Co were dissimilar in less complex microcosms and converged toward similarity as complexity increased. System stability was related more to the sensitivity of biological interactions than to the sensitivity of individual organisms. On the basis of mineral cycling parameters the simpler microcosms were less stable than the complex ones to a moderate stress (100 rads) but more stable than complex systems when subjected to an extreme stress (1000 rads).

10. Radionuclide Cycling in Terrestrial Ecosystems

Sampling of the tulip poplar forest tagged with ^{137}Cs continued to further define rate constants for the redistribution of ^{137}Cs through the ecosystem. The leaching of ^{137}Cs from leaves by rainfall has been decreasing gradually since 1962, but during July 1967 abnormally heavy rainfall caused more leaching than in 1966. There was no detectable increase in dispersal of radiocesium beyond 20 m outside the experimental plot. Radioactivity in 10 to 20 m from the plot is intermittent and appears to be related to the dispersal of tagged leaves, but within 10 m of the plot a severalfold increase in ^{137}Cs may be related to other botanical or zoological processes.

The weight loss from dogwood leaves during their first four weeks in litter is caused primarily by the leaching of water-soluble material. Weight loss was 37.5 ± 1.7% and energy loss was 27.7 ± 2.0% after 26 weeks, showing that the material removed from the leaves had a relatively low caloric value. Fungi are important in the mobilization of mineral elements in the litter. However, fungi can obtain mineral elements either from the solid substrate or the soil solution. With ^{137}Cs as a tracer, experiments showed that ^{137}Cs uptake by fungi isolated from solid substrates was proportional to the ^{137}Cs concentration in the soil solution. For fungi in direct contact with solid substrate the concentration appears to be proportional to ^{137}Cs availability in the substrate. Availability is a function of decomposability and leachability of ^{137}Cs in the substrate.

Studies of forest floor arthropods included chemical analyses of 37 species for Ca, K, and Na. The calcium content was most variable since some species have well-developed exoskeletons rich in calcium. Because of the variability of ash content, results were expressed as element content per gram of ash-free weight. Excluding Diplopoda, all other arthropods averaged 1.89 mg of calcium per gram. The distribution of sodium among species was symmetrical, with an average of 4.6 mg per gram of ash-free dry weight. The potassium frequency curve was positively skewed, due to the occurrence of high values in a few diplopod species. Generally potassium and sodium concentrations were consistent within and among major taxonomic categories. Whole-body concentrations of potassium and cesium decrease with an increase in trophic level, while sodium increases at the higher trophic level. In addition to trophic level differences, some species exhibited seasonal differences in their chemical composition. These differences were attributed to changing seasonal composition of the food base for the organisms. Food chain transfers of nutrients between trophic levels involve characteristic delays or time lag phenomena. Equations were developed to allow prediction of ^{137}Cs concentrations in successive trophic levels in the cesium forest. A 30-day biological half-life was obtained for the saprovore trophic level, which approximated the biological half-life determined previously for several of the species. This predictive equation may be applicable to the study of environmental pollutants in food chains.

Preliminary estimates of erosion and radiocesium redistribution on a fescue meadow were made to evaluate potential ^{137}Cs losses from this field to be tagged with ^{137}Cs-contaminated sand grains. The four experimental plots will contain 8 curies of contaminant, and estimated losses through

eroded soil and runoff water were 1.7 mc/year. Agreement between these results and those predicted by the Wischmeir and Smith soil-loss equation were relatively good, considering that the predictive model was derived for cultivated systems and that the topographic features of the experimental plots approach the lower limits for which the model is applicable.

11. Radionuclide Cycling in Aquatic Ecosystems

Fish from White Oak Lake are being assayed to determine the body burden of ^{137}Cs, ^{106}Ru, and ^{60}Co. Most emphasis is being placed on ^{137}Cs in bluegills and the comparative metabolism of cesium and potassium. The biological half-life of ^{137}Cs in White Oak Lake bluegills weighing 60 to 70 g (12°C) was 115 days and of 10-g fish 80 days. The excretion curves obtained are essentially linear and do not show the rapid initial elimination of ^{137}Cs ordinarily found in acutely tagged fish. These results suggest that most of the ^{137}Cs in bluegills is associated with the long component in biological half-life curves. Similar excretion experiments with snails and newts chronically contaminated with ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{65}Zn, and ^{60}Co showed similar results in that 80% or more of the radionuclide burden was associated with the long component in the excretion curve. The turnover of potassium by bluegills studied with ^{42}K-tagged fish indicated a biological half-life of about 16 days. This half-life is somewhat shorter than that indicated from potassium balance studies. The feeding habits of White Oak Lake bluegills are being studied to measure feeding rates of fish. About 71% of the diet consisted of chironomid larvae and pupae, 13% other insects and fish, and 16% detritus and algae.

Rates of uptake, sites of deposition, and the biological half-life of ^{60}Co in the black bullhead were determined. Uptake of ^{60}Co was rapid, and near-maximum levels of activity were obtained within one day, but uptake experiments continued for seven days. The most rapid uptake from water was by gills, gut, and stomach. The flesh had a lower concentration, but since it comprises 60% of the body weight, it was a major source of whole-body activity. Excretion of ^{60}Co by fish tagged in water and by those fed tagged food was compared. Fish tagged in water lost 28% of their initial dose in the first day of excretion, while those fed tagged food lost 95.5% of their initial activity. Otherwise the excretion curves were similar, with the first component having a biological half-life of 1.5 to 4 days, a second component of 35 to 48 days, and a third component having a long but undetermined biological half-life. Dissections of White Oak Lake bullheads showed that skin and bone contained proportions of ^{60}Co similar to those in the experimentally tagged fish, while blood, liver, and kidney were higher than in laboratory fish. The kidney of these fish contained a major pool of ^{60}Co; this was confirmed by stable cobalt analyses, which showed that the kidney had more cobalt per gram than any other tissue. Similar experiments are under way to define radiotungsten metabolism by the bullhead.

Feeding rates of snails were determined with a new and simple radioisotopic method. Rocks with their attached aufwuchs were tagged by placing them in a ^{60}Co solution containing 2 to 7×10^4 dis/min of ^{60}Co per milliliter. The rocks were rinsed and placed in clean water, and snails were allowed to feed on the rocks for 1 hr. After feeding, the snails were counted for ^{60}Co activity, samples of the aufwuchs were scraped from the rock and ^{60}Co activity per milligram was determined. Dividing the ^{60}Co activity in the snails by ^{60}Co per milligram of rock yielded the milligrams of aufwuchs consumed per hour. Movement of food through the gut of snails was slight within the 1-hr period and reached a maximum at 2 hr after the start of feeding. Feeding rates of snails determined by this method varied from 0.01 to 0.08 mg of aufwuchs per hour.

12. Watershed Aquatic Habitat Interactions

The paired Walker Branch watersheds were essentially completed as a research facility during the year, and experimental programs were planned and initiated. Two 120° V-notch weirs were constructed, and stage-height recorders and proportional water samplers were installed. Wong precipitation collectors were modified to serve as differential collectors of dry fallout and rain-scavenged materials and were installed at each of the five gage sites. The computer program for data reduction and processing is about 50% complete. An innovation in identification of storm events is through the use of rainfall intensity-duration records rather than the stream hydrograph.

Preliminary chemical analyses show the ionic content of the water is essentially normal for a stream in a limestone formation. The west fork of Walker Branch generally follows expected patterns of calcium and magnesium content as different springs flow into the stream. The east fork has some anomalies in dissolved calcium and magnesium content which may be the consequence of soil formations in area adjacent to the stream. A soft water spring is located in an area of Tarklin soil. This soil has a fragipan, and it was hypothesized that incoming precipitation percolates to the fragipan and then flows laterally to emerge at the spring without contacting the limestone bedrock. A more typical spring nearby is located in an area of highly permeable Bodine soils, and hard water occurs through contact with limestone.

Aquatic fauna of both forks was sampled to determine the biotic resources available for study. Approximately 60 different species were collected and preliminary identifications made. The snail *Goniobasis clavaeformis* is one of the most obvious components of the community. However, a large number of caddis flies, mayflies, and stoneflies are also present.

13. Systems Ecology

The developing systems approach to ecology received added support from a Ford Foundation Grant which permitted a number of visiting investigators to utilize facilities at the Laboratory. In addition to the ongoing programs these investigators were able to summarize data available from a number of ecosystems and in some cases develop models from the empirical data. These models are instructive in several ways, but perhaps most important is the potential for interaction of the model results with experimental programs.

14. Forest Management

The forest management program achieved its mean annual cutting rate of 1,500,000 bd ft of sawtimber for the first time. Thinning of the 20-year-old loblolly and shortleaf pine plantations yielded 3300 cords of pulpwood and fence posts. Forest management is conducted in cooperation with ecological research on the AEC reservation.

PART III. RADIATION PHYSICS

15. Theoretical Radiation Physics

A new evaluation has been made of mean excitation energies for some 36 compounds and mixtures of importance in radiation dosimetry. Dose buildup due to delta rays in a soft-tissue slab bombarded by normally incident high-energy protons has been studied. A simplified model for nuclear interactions has been used to obtain estimates of dose for neutrons and protons in the range from 0.4 to 2 Gev incident upon a soft-tissue slab. Numerical calculations have been made of ground-state energies and eigenvalues for an electron in the field of a stationary electric dipole. The quantum theory of a process in which a photon excites a normal surface plasmon which then undergoes radiative decay by collision with density fluctuations in the plasma has been formulated. The Coulomb excitation and radiative decay of spherical plasmons has been investigated theoretically. A fundamental quantum-theoretic study of the probability of multiple real transitions in an interacting fermion-boson system has been carried out. The mean free path of low-energy electrons in a free-electron gas has been calculated using a Bethe-Goldstone formulation. An explanation of Wood grating anomalies in terms of intermediate resonant surface plasmon states has been given. A Monte Carlo code for calculating the penetration of low- and intermediate-energy electrons in metal-insulator-metal diodes is described. The effect of surface plasmon decay in the spectrum of electrons ejected from a metal by energetic charged particles has been investigated theoretically. Progress in the calculation of internuclear potential energy as a function of nuclear separation in a diatomic molecule using Klein's semiclassical method is described. A theory of exchange and nonadiabatic polarization effects in low-energy electron scattering on an atomic system has been developed and is being applied to scattering on alkali atoms and noble-gas atoms.

16. Interaction of Radiation with Liquids and Solids

Reflectance and/or transmittance measurements have been analyzed to yield optical constants in

the vacuum ultraviolet for anodized and single-crystal Al_2O_3 and for evaporated films of Te, Pd, Ni, and Au. In addition, an ellipsometer has been constructed and used to determine the optical constants of Pd at long wavelengths. Photoemission measurements have been made on Cu, Pd, and Ni over the same energy range as the optical measurements.

The optical constants for Al_2O_3 show character-

17. Atomic and Molecular Radiation Physics

Electron transport in gases yielded the first experimental evidence for the minimum dipole moment necessary to bind an electron to a polar molecule. Studies on the scattering of thermal electrons from 41 pure polar molecules indicated for the first time the possible effect of a potential resonance on the scattering of low-energy electrons from polar mole-

These techniques have provided new information about many important fundamental collision processes and about the structure of the common gases (e.g., water, ammonia, hydrogen chloride, etc.), complex molecules (e.g., benzene, naphthalene, azulene, etc.), and molecules of biological interest (e.g., pyridine).

Employing SF_6 as a scavenger of low-energy electrons, the threshold excitation spectra of benzene, naphthalene, chloronaphthalene, nitrobenzene, azulene, pyridine, and xenon hexafluoride have been studied for the first time. The results exhibited considerable new information about the excited states of these molecules (especially the position of triplet states) and the existence of temporary negative ion states. The degenerate temporary negative ion resonance in benzene was found to be split into two resonances for pyridine – one at the energy of the benzene resonance and another approximately ½ v lower in energy. The temporary negative ion resonance in naphthalene was observed to shift to lower energy with the addition of chlorine or bromine onto one of the ring structures. Transient negative ion states were also studied in ethylene, carbon dioxide, and five fluorobenzene derivatives. The fluorobenzene data provided evidence for a linear increase in the electron affinity with increasing number of fluorines added to the benzene ring.

Nondissociative electron attachment to polyatomic molecules has been the focus of much fruitful study. An electron swarm procedure has been developed to measure electron diffusion and electron attachment rates under zero field conditions where the electron swarm energy distribution is most certainly thermal. Negative ion lifetimes have been determined very accurately for azulene and some eight aromatic and alicyclic fluorocarbon molecules. The latter results exhibited a very striking increase in lifetime with increasing number of atoms in the molecule. Furthermore, the experimental results qualitatively agreed with previous theoretical predictions.

Dissociative electron attachment processes have been studied in ammonia, deuterated ammonia, carbon dioxide, carbon monoxide, nitrogen dioxide, nitrous oxide, naphthalene, chloronaphthalene, bromonaphthalene, a series of fluorocarbons, and xenon hexafluoride. A rather complete study of ammonia and deuterated ammonia yielded the magnitude and energy dependence of the cross sections, the ions formed, kinetic energies of the negative hydrogen ions, and the electron impact excitation spectrum. Measurements of the appearance potentials of O_2^- and NO^- from nitrogen dioxide have provided interesting lower limits to the electron affinities of these molecules.

A new technique has been developed to study negative-ion–molecule reactions in the energy region from 0 to 2 ev. Reactions of H^- (and D^-) ions with H_2O (and D_2O) are shown to lead to OH^- (and OD^-) with a quite large cross section which increases with decreasing energy. A significant isotope effect favoring the $D^- + D_2O \rightarrow OD^- + D_2$ reaction was observed. Simple charge-exchange reactions of O^- with NO_2 leading to NO_2^- have also been studied as a function of the O^- ion energy.

19. Graduate Education and Vocational Training

In the Graduate Education and Vocational Training Program, five AEC Health Physics Fellows came to ORNL for summer training in applied health physics and research. Two came from the University of Rochester and one each from the Universities of California, Illinois, and Vanderbilt. The book *Principles of Radiation Protection: A Textbook in Health Physics* was published in December 1967. This book should be widely used in health physics education. Thirty-five colleges and universities were visited, and health physics research and career opportunities were discussed. Nineteen graduate students conducted thesis research in the Health Physics Division for the M.S. or Ph.D. degree during the year. A total of 38 university personnel ranging from undergraduates to professors spent last summer in the Health Physics Division. Several of our staff assisted the University of Tennessee Physics Department in writing research proposals to government agencies. The Division cooperated with Oak Ridge Associated Universities in screening of applicants for USAEC Fellowships; participation in an eight-week course in Health Physics for college teachers; and assistance in presenting a ten-week course and a three-week course for

state personnel. Lectures were given at several courses for M.D.'s entitled "Medical Radioisotopes." One member of the Division spent six weeks in Bombay, India, giving a series of lectures on radiation physics at the Bhabha Research Center.

20. Physics of Tissue Damage

In the Physics of Tissue Damage program the optical properties of liquid water in the vacuum ultraviolet spectral region were measured for the first time. Two methods were employed. In one the reflectance of water in equilibrium with its vapor was measured over the spectral region from 1050 to 3000 A for three angles of incidence. In the second method quartz or CaF_2 semicylinders were used with the liquid in contact with the flat portion of the semicylinder in a sealed cell. Light was incident and exited from the semicylinder on the curved surface at various angles. These two methods together yielded values of n and k, the real and imaginary parts of the index of refraction; ϵ_1 and ϵ_2, the real and imaginary parts of the dielectric constant; and $-\mathrm{Im}\,(1/\epsilon)$, the imaginary part of the reciprocal complexing dielectric constant. One possible interpretation of the optical data is that there is an exciton transition at 8.3 ev, an interband transition at 9.6 ev, and a band gap of 9 ev. Thus liquid water has optical and electronic properties very much like those of insulators, such as Al_2O_3. Other activities in this program included a recalibration of the electron spectrometer used to measure electron slowing-down spectra with the thought that the anomalously large electron fluxes found in low energy might be attributed to a spurious response of the instrument. However, the instrument was found to respond as expected, and the discrepancy between the Spencer-Fano theory and our results remains. Further measurements were taken on the transmission of very low-energy electrons (2 ev–3000 ev) through thin layers of aluminum and sodium. Approximately 28% transmission was found out to 20 ev, whereupon the transmission decreased to essentially zero at about 50 ev, became unity again between 400 and 1100 ev depending on film thickness, and went negative beyond this energy (more electrons were ejected from the films than were absorbed by the films).

PART IV. RADIATION DOSIMETRY RESEARCH

21. Dosimetry for Human Exposures and Radiobiology

The most significant accomplishment in the dosimetry study for survivors of the nuclear bombings of Hiroshima and Nagasaki was the determination of the energy yield of the Hiroshima bomb; the yield was 12.5 ± 1.25 kilotons. Progress in the evaluation of the shielding afforded by heavy concrete structures was made, and the development of techniques for calculation of doses for the in utero cases was commenced. Analysis of data from Operation HENRE was largely completed; an especially significant finding was a prominent but unexpected gamma-ray peak which was attributed to the reactions $^{16}O(n,\alpha)^{13}C^* \rightarrow n + {}^{12}C^* \rightarrow {}^{12}C +$ 4.43-Mev gamma ray.

22. Applied Research

A highly noteworthy development in these studies was the development of "thermally stimulated exoelectron emission" (TSEE) as a dosimetry system. The system incorporates great sensitivity (can be much less than 1 mr of gamma radiation), low Z (e.g., BeO), wide dose range (over ten orders of magnitude), and, by using different detectors, the capability to distinguish between radiations of different LET.

Another development has been the extension of the etch-pit method of identifying charged-particle tracks in insulating solids to permit alpha dosimetry (i.e., airborne activity such as in uranium mines).

Special "low-Z" glasses of silver-activated lithium borate were developed for gamma-ray dosimetry by the radiophotoluminescence technique; the glasses were found to have promise in neutron dosimetry as well.

23. Spectrometry Research and Development

Except for the reduction and analysis of data from Operation HENRE (see above), the emphasis of the study was on accurate and absolute calibration of both neutron and gamma-ray spectrometers. The 3-Mv Van de Graaff accelerator in the High Voltage Laboratory was used extensively in these studies.

Another important area of investigation was that of developing a good organic scintillator by use of zone-refined quaterphenyl. Thus far, the refining has produced crystals having light output for ^{137}Cs gamma rays as great as that of anthracene to within ±5%.

Calculations of recoil particle spectra in neutron-irradiated tissue have been completed for the cases of 3- and 14-Mev neutrons incident on 30-cm-diam cylinders.

24. HPRR and DLEA Operations

The HPRR was used extensively in dosimetry, radiobiology, and radiobotanical studies. There were no major modifications and no significant changes in operating procedures.

The DLEA was modified extensively to provide for precise energy selection of charged particles to be used in stopping-power studies.

PART V. INTERNAL DOSIMETRY

25. Internal Dose Estimation

The fluctuations of daily excretion levels of plutonium in hospital patients receiving a single intravenous injection of plutonium were studied and reported in 1967 (ORNL-4168, p. 273). Following a single intake to blood, subsequent significant increments of intake would be expected either to change the general trend of the excretion curve or to produce an excess of high values. During the past year a plutonium worker (not an ORNL employee) suffered a minor cut which was contaminated with ^{239}Pu. The excretion data were made available with a request for assessment of the body burden. The interpretation required some changes in the computer code because of the early excision of tissue and treatment with DTPA. Study of the trend of the data and of the distribution of the amounts excreted per day supports the conclusions that the treatment did enhance excretion and that translocation of plutonium from the wound to bone after the first day was minimal.

A Monte-Carlo-type computer program for estimating the absorbed fraction of gamma energy in a "target" organ from a uniform source in a "source" organ was reported in 1967 (ORNL-4168, p. 245). The program is based on a "phantom" and 23 "organs" which are defined by simple mathematical inequalities suitable for fast calculation by the computer. The entire phantom and each of the "organs" have a volume and shape approximating that of the whole body or of the corresponding organ. During the past year the computer program has been modified to allow each "organ" to have approximately the composition and density of the corresponding human organ. This permits one to estimate with more confidence the absorbed fraction for the skeleton and lungs. The results differ from those obtained using the homogeneous model principally in the low-energy region, say when the photon energy is below 0.2 Mev. Results are now available for the source in ten different organs and for 15 photon energies from 0.01 to 4 Mev.

The total dose received by an individual from ^{137}Cs present in a particular food (e.g., meat) will be proportional to the product of four factors: $C \times I \times T_b \times D$, where C is the concentration of ^{137}Cs in the particular food, I is the daily intake of that food, T_b is the biological elimination half-time of ^{137}Cs for the individual, and D is the dose received per disintegration of ^{137}Cs, which is assumed to be uniformly distributed in the body. The variation with age of each of the four factors is summarized, and the variation of total dose or dose commitment per day of exposure is considered for two cases: (1) where a single foodstuff is contaminated (e.g., milk, meat, fresh fruits, fresh vegetables) and (2) for fallout ^{137}Cs present in the Chicago diet as of January 1967 and January 1968.

When a radionuclide is present in an organ and produces daughter nuclides, some fraction of these daughter elements remains in the organ and will be eliminated at rates which may differ from the elimination rates of the parent. A mathematical model has been designed to provide estimates of the activity levels of the parent radionuclide and of the daughter nuclides present in the organ at an arbitrary time post intake of the parent. The model has been programmed for a digital computer for the case in which there is no significant translocation of daughter nuclides from other tissues and when the retention of each nuclide in the organ can be described as a sum of exponentials. Estimates of the total microcurie-days of residence for each radionuclide are provided also.

A model for estimation of the mean activity present in various segments of the gastrointes-

tinal tract following a single oral intake has been developed. The model assumes uniform mixing of the contents in the various segments of the tract or, equivalently, an exponential distribution of residence times for the contents in each segment, as has been suggested by some experimenters. The model is mathematically equivalent to a catenary compartment system, and thus the solution is a sum of exponentials. The model also provides estimates for the mean activity of daughters if they are present.

Studies related to eventual estimation of the hazard posed by ^{238}Pu dioxide particles with diameters in the range of 50 to 250 μ have been conducted to estimate the probability of inhalation of such particles. The theoretical formula takes the settling velocity of the particle into account as well as the breathing rate. Studies of the dose distribution in tissue near such a particle have been carried out.

Sorption of SO_2 gas on aerosol particles of lead oxides, aluminum oxides, platinum, and iron oxide has been studied. Sorption ranges from essentially 100% to 10% or less, depending on the type of aerosol and the conditions of the experiment. Evidence of subsequent release of sorbed SO_2 has also been found in the case of platinum.

26. Stable Element Metabolism

Dietary and excretion data on 25 elements have been obtained for two individuals eating ad libitum during a period of 250 days. The data have been analyzed to study the dietary trends and fluctuations and the resulting patterns of excretion. Regression coefficients relating the intake on a given day to excretion during the next 24 days have been obtained, with multiple correlation coefficients greater than 0.32 ($p < 0.05$) for 10 of the 25 elements.

A practical procedure has been developed for removal of phosphate from solutions of bone ash. This makes it possible to chemically concentrate the trace elements at pH 6.5 rather than at pH 5.2, substantially increases the number of trace elements that can be collected, and eliminates the undesirable coprecipitation of calcium phosphate.

Part V. Internal Dosimetry

W. S. Snyder

25. Internal Dose Estimation

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AN ESTIMATE OF THE SYSTEMIC BODY BURDEN OF ^{239}Pu RESULTING FROM A CONTAMINATED WOUND

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Knowledge of the retention and toxicology of plutonium in the human body is very limited, and techniques of total-body counting are not available for detecting a permissible body burden. Thus when exposure from an internal source of plutonium occurs at levels near or below the limiting organ burdens, it must be evaluated on the basis of an interpretation of excretion data. Excretion data, however, from intakes of known amounts of plutonium on which such an interpretation can be based are scarce and, in fact, are confined almost exclusively to data reported by Langham[3] on hospital patients who received intravenous injections of plutonium citrate. A method of analysis based on these data is used here to evaluate the body burden for a case of contamination via a wound.

The excretion data of the subject as made available for this study are shown in Fig. 25.1. The data are incomplete in that no fecal data are available, the exact time and amount of the first urinary sample are not known, and the urinary data extend only to 150 days after the injury. Also the interpretation is complicated by the use of DTPA (diethylenetriaminepentaacetic acid) treatment, for which neither the times nor amounts of the DTPA administered are known precisely.

A computer code reported in ref. 4 is used to estimate intake to blood. It is based on a power function model of excretion fitted to the urinary excretion data of the hospital patient.[3] The effect of

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

[4]W. S. Snyder, M. R. Ford, and G. Warner, "A Study of Individual Variation of Excretion of Plutonium by Man and Its Significance in Estimating the Systemic Burden," Proceedings of Thirteenth Annual Bioassay and Analytical Chemistry Meeting, Oct. 12–13, 1967, Berkeley, Calif. (in press).

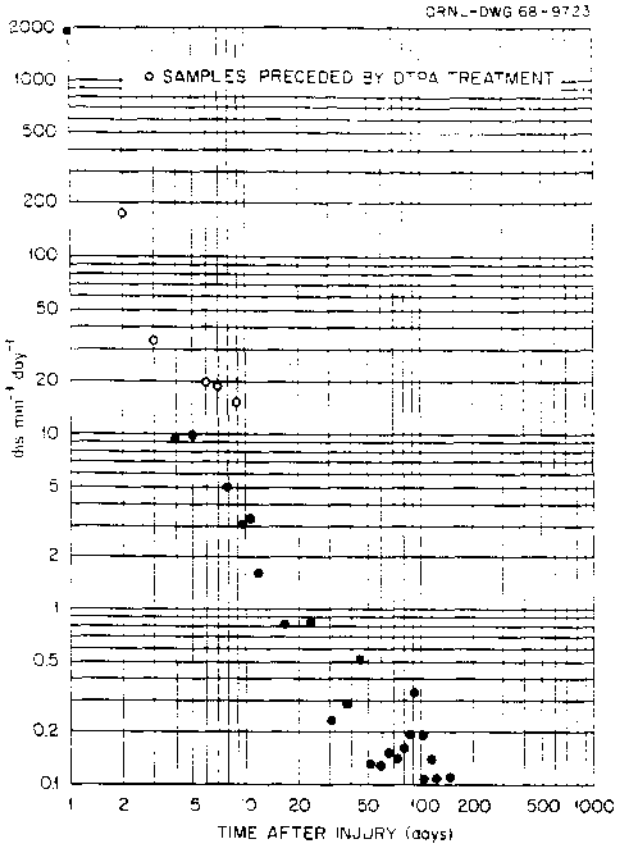


Fig. 25.1. Urinary Excretion Data of Subject.

DTPA treatment on rate of excretion is not allowed for in the computer program. There are conflicting accounts of the enhancement to be expected from treatment with DTPA, practically no enhancement[5] to enhancement by a large factor, perhaps 100-fold.[6] In the absence of information on the level of treatment, it does not seem possible to posit any quantitative correction for the effect of treatment in this case. Consequently, judgment concerning the effect of treatment has been based on the internal evidence of the data on this case.

The trend of the subject data is different in the early period, say days 1 to 10, than at later times, say days 20 to 150. This difference is confirmed by fitting power functions to the two portions separately by two methods (see Table 25.1): (1) the Point Fit method, minimizing the sum of absolute deviations, and (2) the Area Fit method, also minimizing the sum of absolute deviations, but which considers excretion as a continuous process. The curves fitted to the excretion data beginning at day 1 or day 17 produce steeper slopes and larger coefficients than do those fitted at later times. It is of some interest to establish the cause of steeper slope, for example, whether it indicates leaching of plutonium from the wound site or changing enhancement of the excretion due to the treatment. Leaching of plutonium from the wound site would be expected to increase the excretion rate, but it does not seem likely that this could account for a steeper slope of the excretion curve. If the power function correctly indicates the slope of the excretion curve of a single intake, this would plot as a straight line on log-log paper. No superposition of such lines at different times can produce a steeper slope for the curve which represents the sum of the

[5] J. R. Mann and R. A. Kirchner, *Health Phys.* 13(8), 877–82 (1967).

[6] C. R. Lagerquist, I. B. Allen, and K. L. Holman, *Health Phys.* 13(1), 1–4 (1967).

Table 25.1. Power Functions $at^{-\alpha}$ Fitted to Excretion Data of the Subject over Certain Periods of Time

Days post Exposure Included	By Point Fit–Least Deviations		By Area Fit–Least Deviations	
	Coefficient a (dis min^{-1} day^{-1})	Exponent α	Coefficient a (dis min^{-1} day^{-1})	Exponent α
1–10	1909	3.47	95.5	0.95
17–150	16.7	1.06	15.8	1.05
24–150	5.32	0.80	5.08	0.79
38–150	5.32	0.80	5.08	0.79
45–150	0.707	0.37	1.12	0.37

intakes. Thus, we ascribe the increased slope as an effect of treatment. There is no significant change detected whether one begins the fitting at day 24 or day 38, and the value of 0.8 obtained here for the exponent is well within the range of values reported for the hospital patients and is quite close to the value of 0.77 obtained by Langham.[3] The much lower exponent and coefficient when the fitting begins at day 45 is attributable to the fact that most of the measured values are close to the limit of detection. As the excretion levels approach background values, that is, the levels found to occur in "blanks" from "unexposed" individuals, the curve would be expected to become flatter. It seems reasonable that a value of about 0.1 dis min^{-1} day^{-1} (which is about the amount excreted per day by the subject after day 45) be taken as this level, the counting statistics as well as reported levels in "blanks" both making this plausible.

Analysis by methods described in ref. 4 of the day-to-day fluctuations ot the excretion data provides another indication that leaching from the wound site during the period from 17 to 150 days is not sufficient to alter the excretion pattern significantly and probably is small compared with the amount entering blood in the early period. The distribution of "high" daily urine outputs is not in excess of that predicted by the methods of ref. 4, as is indicated by the data in Table 25.2. While the procedure used here does not constitute a "proof" that no additional intake occurs after the

Table 25.2. Number of Intakes and Total Intake Estimated by Various Power Function Formulas

Model	Sample Days Used	k, Number of Samples	Fluctuation Allowance (%)	N^a	n^b	Total Intake (μc)
Subject's own formula[c] = $2.3 \times 10^{-3} t^{-0.80}$, PF-ALD[d]	24–150	16	0		12	6.8×10^{-4}
	38–150	14	0		10	6.6×10^{-4}
	24–150	16	10	1.5	2	3.8×10^{-4}
	38–150	14	10	1.3	1	4.2×10^{-4}
	24–150	16	20	3.0	4	5.1×10^{-4}
	38–150	14	20	2.6	3	5.1×10^{-4}
"Typical" formula = $3.2 \times 10^{-3} t^{-0.93}$, PF-ALD[d]	24–150	16	0		13	7.7×10^{-4}
	38–150	14	0		11	7.6×10^{-4}
	24–150	16	10	1.5	3	4.7×10^{-4}
	38–150	14	10	1.3	1	4.6×10^{-4}
	24–150	16	20	3.0	4	5.5×10^{-4}
	38–150	14	20	2.6	3	5.3×10^{-4}
Subject's own formula[c] = $4.8 \times 10^{-4} t^{0.79}$, AF-ALD[e]	24–150	16	0		13	3.0×10^{-3}
	38–150	14	0		11	2.9×10^{-3}
	24–150	16	10	1.5	3	1.9×10^{-3}
	38–150	14	10	1.3	2	1.9×10^{-3}
	24–150	16	20	3.0	5	2.0×10^{-3}
	38–150	14	20	2.6	3	2.1×10^{-3}
"Typical" formula = $1.5 \times 10^{-3} t^{-0.65}$, AF-ALD[e]	24–150	16	0		13	5.8×10^{-4}
	38–150	14	0		11	5.5×10^{-4}
	24–150	16	10	1.5	3	4.1×10^{-4}
	38–150	14	10	1.3	2	3.3×10^{-4}
	24–150	16	20	3.0	4	4.0×10^{-4}
	38–150	14	20	2.6	3	4.4×10^{-4}

[a] Expected number of intakes due to fluctuations = $(k - 1)$ times fluctuation allowance.

[b] Number of intakes found after first intake.

[c] The constant is normalized to Langham's power function fit to the data of the hospital patients.

[d] Point fit—absolute least deviations.

[e] Area fit—absolute least deviations.

first week or two, it does indicate that the excretion data are distributed in value as one might expect following a single or short-term intake to blood. Thus, we consider that the high values of the exponent seen when the early portion of the data is used for fitting are due to treatment, and the lowest values seen when only the late portion of the data is analyzed are due to an approach to background. We consider the value of 0.8 for the exponent as being the best estimate of the slope of the excretion curve for this individual.

The urinary excretion curves fitted to the data of all the hospital patients by the above methods, as reported in ref. 4, are $3.2 \times 10^{-3} t^{-0.93}$ and $1.5 \times 10^{-3} t^{-0.65}$ respectively. These formulas represent a fraction of the injected dose excreted per day. These formulas can be used, therefore, to make a first estimate of the intake of the subject under study. The excretion data of the subject are in units of disintegrations per minute per 24-hr sample. By comparing the formulas of the hospital patients mentioned above and those of the subject, one estimates the subject's intake to be $5.32/3.2 \times 10^{-3}$ dis/min $= 7.6 \times 10^{-4}$ μc using the "point fit" formulas and $5.08/1.5 \times 10^{-3} = 1.5 \times 10^{-3}$ μc if the "area fit" formulas are compared. Since elimination of a systemic body burden is negligibly small when uninfluenced by treatment, the total intake to blood may be taken as the estimate of the systemic burden on day 160 exclusive of any insoluble material which may remain at the wound site or which may be present in the lungs.

EFFECT OF SIZE, SHAPE, COMPOSITION, AND DENSITY OF BODY ORGANS ON THE ABSORPTION OF GAMMA RAYS FROM A UNIFORM SOURCE IN AN ORGAN

W. S. Snyder Mary R. Ford
G. G. Warner[7]

A code has been reported previously by Fisher and Snyder[8] for estimation of dose due to a source of photons within the human body. The phantom used in this code has approximately the form of the human body with a head and a leg region, and a trunk which is an elliptical cylinder. Twenty-two internal organs are mathematically defined as regions within the phantom, and they have approximately the right shape and size. In previous results reported from the use of this phantom, the phantom was taken as homogeneous. The present paper reports results obtained by modifying the code and the phantom in such a way that differences in density and composition of human tissues can be taken into account. The skeletal region is taken with a density of 1.5 g/cm^3 and with approximately the following composition by weight: 40% O, 23% C, 10% Ca, 7% H, 7% P, and 4% N. The lung region is given a density of 0.3 g/cm^3, with a composition as follows: 76% O, 10% H, 10% C, and 3% N. The remaining portion of the phantom is given a density of near 1 g/cm^3 and a composition of 63% O, 23% C, 11% H, and 2% N.

The code uses a modified sampling technique which permits one to estimate the path of a photon originating at an arbitrary point in the phantom but without determining entrance and exit points from the various regions. This avoids time-consuming checks for crossing of boundaries and therefore permits one to use substantially the sample size available previously. Sources distributed uniformly in the skeletal region and in the lungs have been programmed for initial photon energies of 0.01, 0.015, 0.02, 0.03, 0.05, 0.1, 0.2, 0.5, 1, 1.5, 2, and 4 Mev, and a sample size of 30,000 photons was used in each case.

Results obtained with the modified code, that is, using the nonhomogeneous phantom, are compared in Figs. 25.2 and 25.3 with values reported by Fisher and Snyder[8] and with values obtained by the first-collision effective radius method of ICRP Publication 2.[9] It will be noted that the absorbed fractions estimated by the Monte Carlo codes are significantly lower than those obtained by the methods used by the ICRP except for the skeleton and at photon energies below 0.1 Mev, where the increased importance of the photoelectric effect for bone as opposed to soft tissue generally produces higher estimates in the present calculation.

Comparing the two Monte Carlo calculations: at energies above 0.2 Mev the absorbed fractions change approximately in proportion to mass, and

[7] Mathematics Division.

[8] H. L. Fisher, Jr., and W. S. Snyder, "Distribution of Dose in the Body from a Source of Gamma Rays Distributed Uniformly in an Organ," to be published in *Proceedings of International Congress on Radiation Protection, Rome, Italy, September 5–10, 1966.*

[9] *Recommendations of the International Commission on Radiological Protection,* ICRP Publication 2, Pergamon 1959.

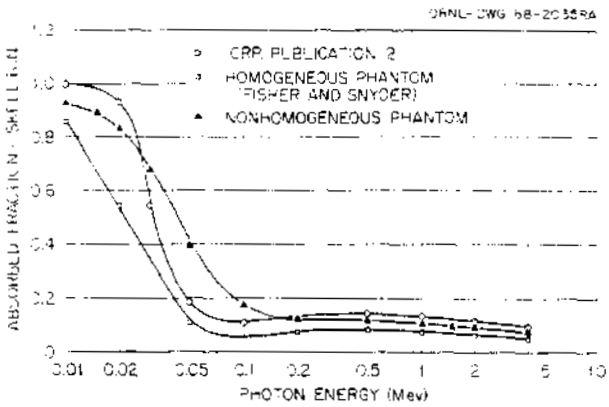


Fig. 25.2. Estimates by Various Methods of Absorbed Fractions of Photon Energy in the Skeleton. Source uniform in the skeleton.

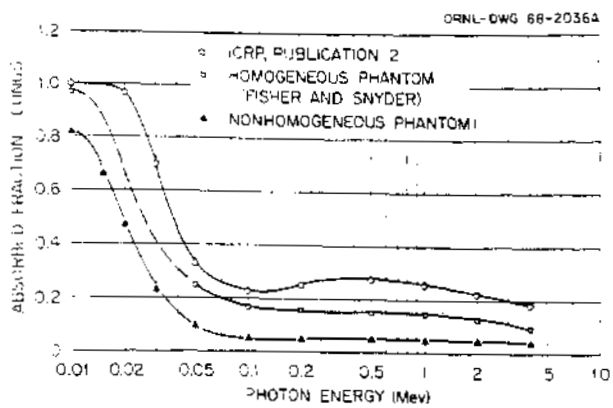


Fig. 25.3. Estimates by Various Methods of Absorbed Fractions of Photon Energy in the Lungs. Source uniform in the lungs.

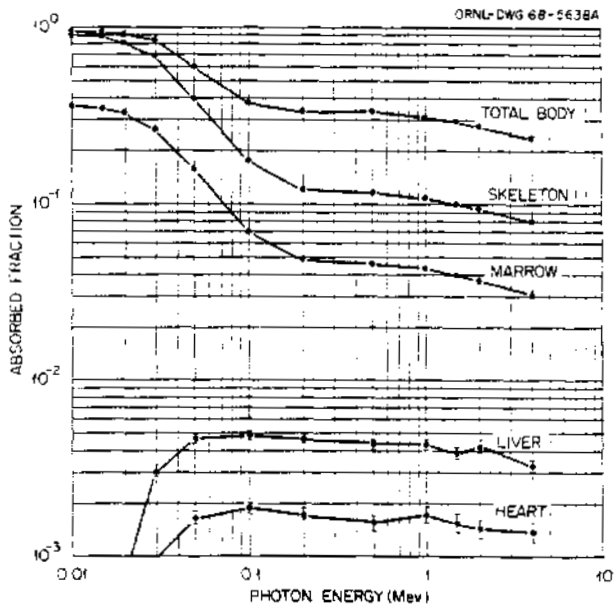


Fig. 25.4. Absorbed Fractions of Photon Energy in Various Organs. Source in the skeleton.

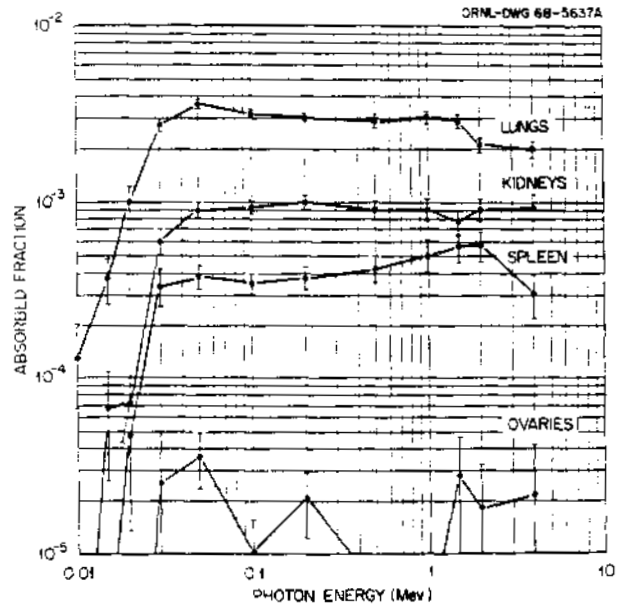


Fig. 25.5. Absorbed Fractions of Photon Energy in Various Organs. Source in the skeleton.

thus the doses are essentially the same; at photon energies below 0.2 Mev no detailed comparison with the earlier calculation for the source in the lungs is possible, because only a few cases were run with the homogeneous phantom. Results with the source in the skeleton are generally higher for the nonhomogeneous phantom than for the homogeneous. This is as would be expected, since not only is there greater mass in the nonhomogeneous case, but also the absorption coefficients are greater for bone than for tissue.

Figures 25.4 and 25.5 show the absorbed fraction in some organs other than those in which the source was located. The computed points have

been connected by straight lines, and the bars on the points represent one standard deviation on either side of the mean. Where no bars appear the deviations are less than 2% of the mean. These results illustrate what is, perhaps, the most novel feature of this code and mode of calculation in that it provides estimates of the absorbed fraction for all the 22 organ systems as well as for the whole body. Only the absorbed fractions considered to be statistically significant are shown in Fig. 25.4. In some cases, where the organ region is very small or located at a great distance from the source or both, the absorbed fraction is so small and the number of photon interactions occurring in the region is so small that the estimates are unreliable. Such a case is illustrated in Fig. 25.5, where for the absorbed fraction with the source located in the skeleton, one standard deviation is 30 to 95% of the mean. This could be remedied by running a larger sample of photons, but machine time becomes the limiting factor.

The estimates of absorbed fraction or, equivalently, of dose obtained by the method of this paper are sufficiently accurate to obtain the average dose in the organs represented here in all those cases where the statistics are sufficiently accurate, and we are trying, at present, various means of obtaining more reliable results for small organs located rather far from the source.

THE RELATIVE IMPORTANCE OF ^{137}Cs CONTAMINATION IN VARIOUS FOODSTUFFS IN TERMS OF TOTAL DOSE FOR VARIOUS AGE GROUPS OF A POPULATION

Mary Jane Cook W. S. Snyder

If a particular foodstuff, symbolized by the index i, is contaminated with ^{137}Cs, the total dose received by an organ of an individual consuming foodstuff i is the product of four factors – namely, $C_i \times I_i \times D \times T_b/0.693$, where C_i is the concentration of ^{137}Cs in the foodstuff, I_i is the daily intake of the foodstuff, T_b is the half-time for biological elimination from the body, and D is the dose rate to the organ in rems per day due to the presence of unit activity in the body. This is equivalent to assuming 100% absorption of ^{137}Cs into the blood and using a single-exponential model for elimination of ^{137}Cs from the body. Taking D as a constant amounts to assuming the distribution of ^{137}Cs in the body does not change much with time, and this seems to be the case for the forms of ^{137}Cs commonly encountered in the diet. Each of these four factors varies somewhat with age and other characteristics of the individual, and thus in determining the critical group of an exposed population, one must consider the variation of each factor. It is the product of the four factors which is proportional to the dose commitment per day of exposure and which will be indicative of the critical group so far as dose commitment is concerned. The variation of each of these factors with age will be discussed here briefly as an example of how this criterion can be applied in assessing the significance of exposure of a population to environmental ^{137}Cs.

The dose rate D due to unit activity in the body has been studied by Fisher and Snyder,[10] who give values of D appropriate for individuals of different sizes and for organs located at different positions in the body. A Monte-Carlo-type code was used to estimate the dose rate within the body due to a uniform distribution of ^{137}Cs within the body. Six phantoms corresponding to the newborn, to 1, 5, 10, and 15 year olds, and to an adult have been designed, and dose rate estimates are available for each of them. Fisher and Snyder assumed that 1 μc of ^{137}Cs was distributed uniformly within the phantom, and the absorbed fraction of energy for the whole body is presented in Fig. 25.6. A difference of about a factor of 2 is found in the fraction of energy absorbed when the same concentration of ^{137}Cs is present in the infant and in the adult. This applies only to the gamma dose rate, as the dose rate due to the beta radiation present does not vary significantly with body size if the concentration remains constant. It happens that the doses due to gammas and betas are approximately equal; so the total dose rate would only be about half as sensitive to changes in body size as is this gamma dose rate. The variation of the gamma dose rate within the phantom is shown in Fig. 25.7. There is approximately a difference of a factor of 2 in the gamma dose rate distribution as one proceeds from the central axis to the lateral surface of the phantom. The difference is more like a factor of 4 as one goes from the central axis to the head or feet.

We consider next the second factor, the biological turnover time, and its possible variation with age

[10] H. L. Fisher, Jr., and W. S. Snyder, *Health Phys. Div. Ann. Progr. Rept. July 31, 1966*, ORNL-4007, p. 221.

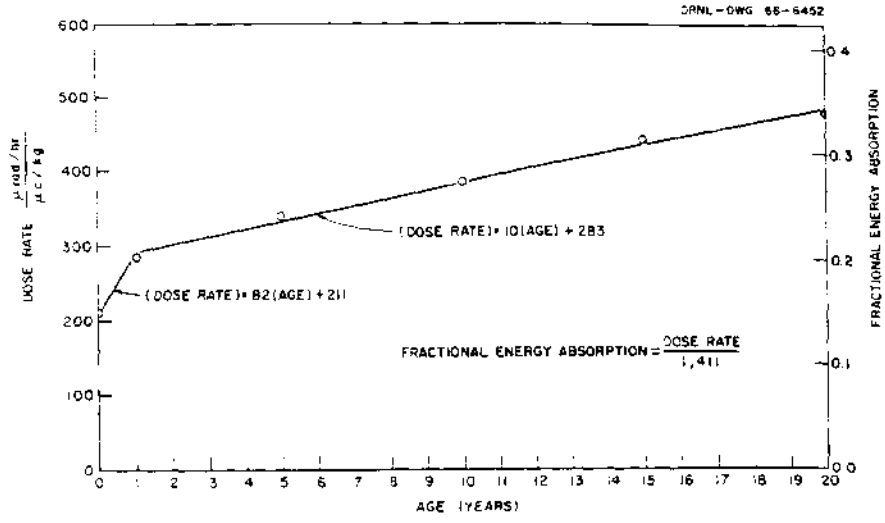


Fig. 25.6. Average Total-Body Gamma Dose Rate from a Uniformly Distributed ^{137}Cs Source.

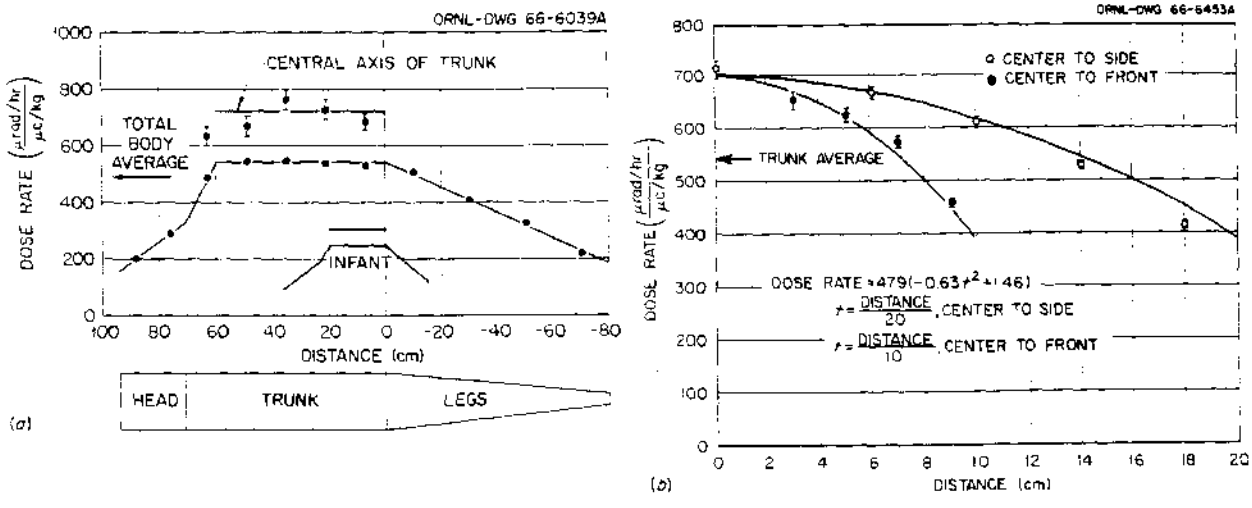


Fig. 25.7. Variation of Gamma Dose Rate in Phantom. (*a*) Average gamma dose rate in horizontal layers in the adult phantom from uniformly distributed ^{137}Cs source; (*b*) gamma dose rate from center to surface of adult phantom from a uniform ^{137}Cs source.

which is shown in Fig. 25.8. Several investigators have discussed the biological half-life of ^{137}Cs for different age groups, and various models have been proposed. Snyder and Cook[11] have offered one model (the dashed curve) based on stable cesium in the diet and the human body; the solid curve is obtained by Fisher and Snyder,[12] who observed that the experimental values of T_b were roughly proportional to body mass and thus related to age. For illustrating the use of age-dependent

[11] M. J. Cook and W. S. Snyder, *Health Phys. Div. Ann. Progr. Rept. July 31, 1965*, ORNL-3849, p. 190.

[12] H. L. Fisher, Jr., and W. S. Snyder, *Health Phys. Div. Ann. Progr. Rept. July 31, 1967*, ORNL-4168, p. 261.

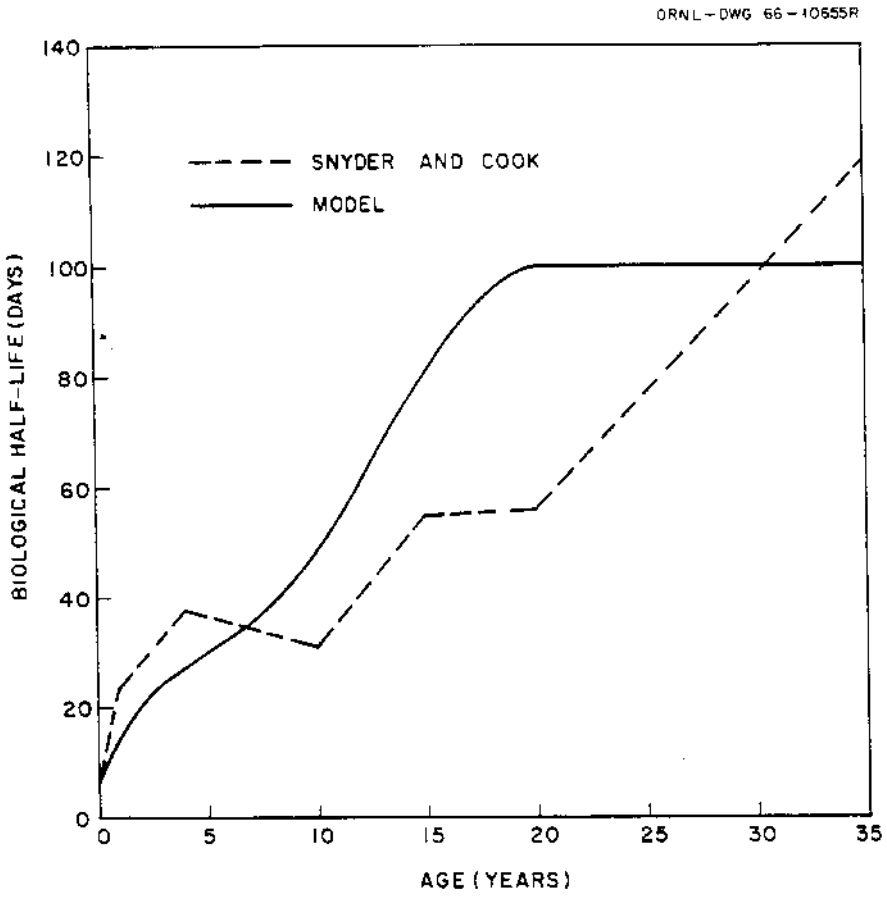


Fig. 25.8. Variation of the Biological Half-Life of Cesium with Age.

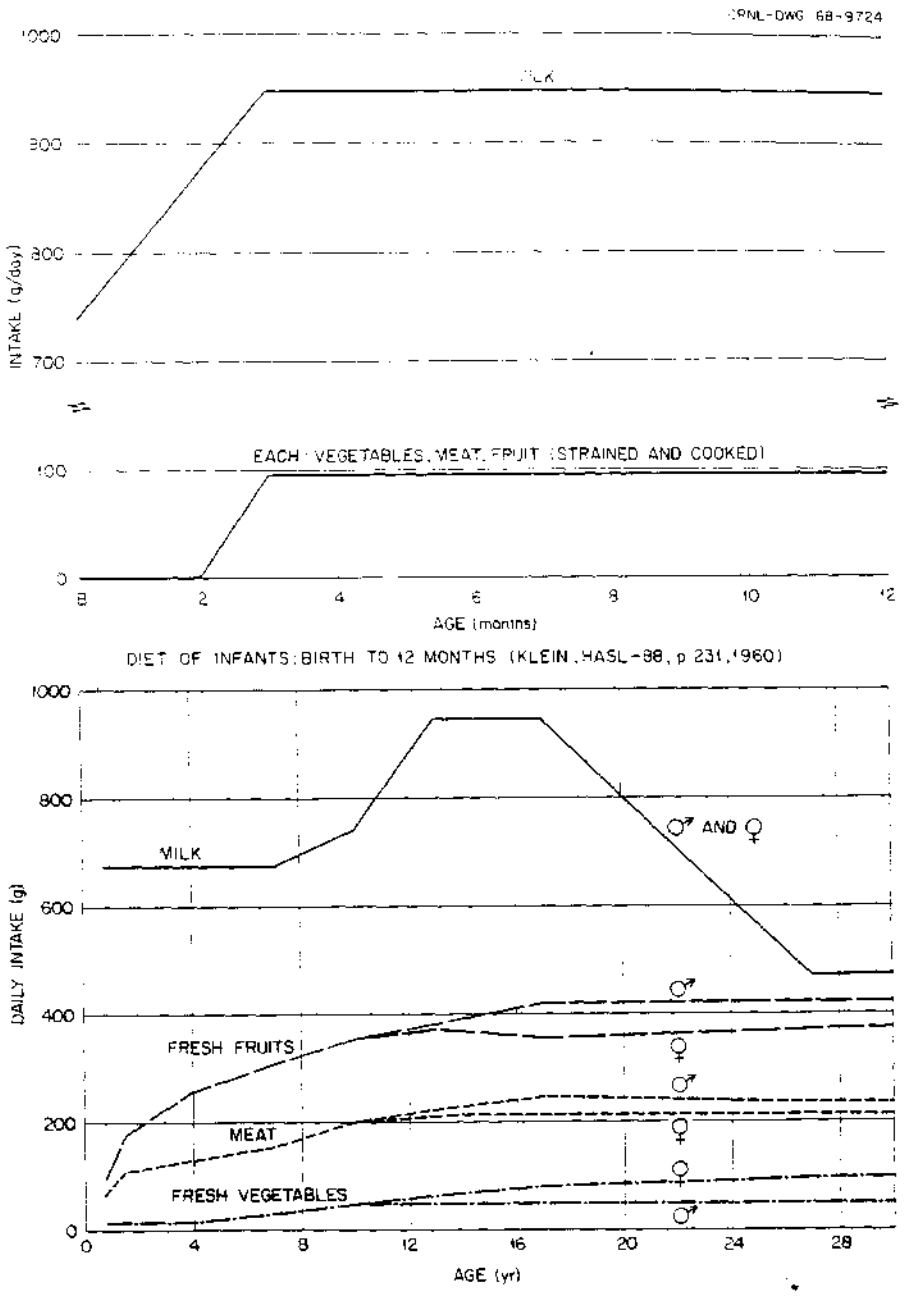
data, we have selected the model of Fisher and Snyder.

The third factor, I_i, of the original expression is the daily intake of foodstuff i, which is also a function of age. As an illustration, the intake of milk, meat, fresh vegetables, and fresh fruits has been considered. The data of Fig. 25.9 corresponding to diets consumed after the first year of life were obtained from ref. 13 and represent a moderate-cost family food plan of 1964. It will be noted that intake is significantly different for different ages as well as for males and females. The diet for the infant during the first year was taken from Klein.[14] The infant of one year or less presumably eats mainly cooked, strained, and canned meats, fruits, and vegetables. Because such an infant probably consumes quite small quantities of fresh fruits and vegetables, we have substituted intake of the cooked, strained, and canned fruits and vegetables for the fresh during the first year of life in calculating dose commitment for our illustrative cases. The estimated diets for the first year of life are from a different source than those for older individuals, and we have not attempted to reconcile the difference of the estimates at one year of age.

We have studied the product of these four factors – C_i, concentration of ^{137}Cs in the foodstuff; I_i, daily intake of foodstuff as a function of age; T_b, the biological half-life as a function of age; and D, the dose rate in rems per day due to the presence of unit activity in the body. In Fig. 25.10 we have

[13] Agricultural Research Service, *United States Department of Agriculture Family Economics Review*, Consumer and Food Economics Research Division, October 1964.

[14] S. Klein, *Health and Safety Laboratory Fallout Program Quart. Summary Rept. July 1, 1960*, HASL-88, p. 231.



Typical Intake of Various Foods by Different Ages for a Moderate Cost Food Plan – U.S. Department of Agriculture, Revised 1964.

Fig. 25.9. Typical Intake of Various Foods by Different Ages (Newborn to 30 Years).

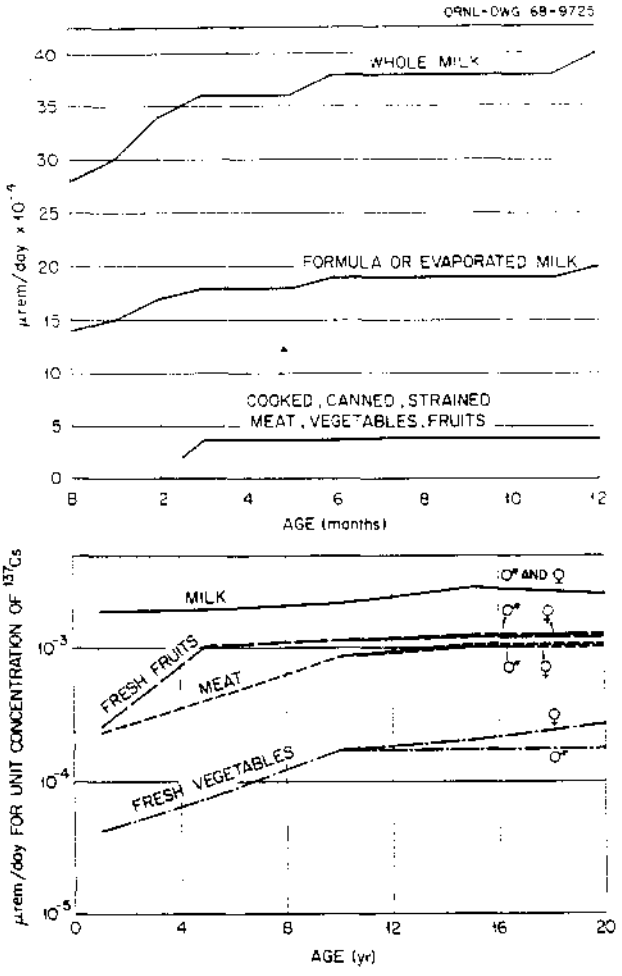


Fig. 25.10. Dose Commitment per Day of Exposure from Unit Concentration of ^{137}Cs in Various Foods (Newborn Through 20 Years).

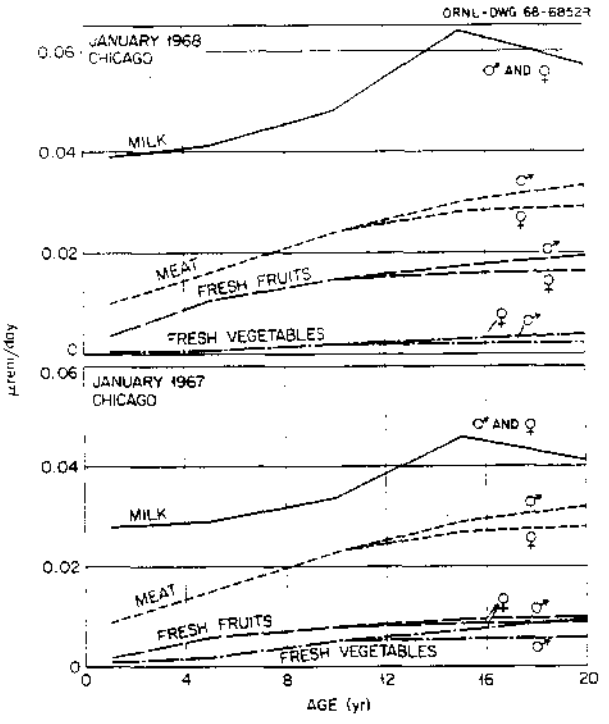


Fig. 25.11. Dose Commitment of Exposure from Fallout ^{137}Cs via Various Foods (1 Year Through 20 Years).

taken C_f as 1; that is, a unit concentration of ^{137}Cs is assumed in some one of the principal foodstuffs – milk, meat, fresh vegetables, and fresh fruits (canned vegetables and fruits during the first year of life) – which seem to be pathways for intake of ^{137}Cs. These basic data are applicable to any situation where data on concentrations of ^{137}Cs in the listed foods are available and for which the intakes are typical.

To illustrate its use for an actual situation and also to give a better idea of the relative importance of various foodstuffs in the case of contamination by fallout, values of C_f have been taken from the studies of Brar *et al.*[15,16] of Chicago foods in January 1967 and January 1968, and dose commitments per day of intake have been estimated for these levels of contamination in the four foods considered here. These estimates, plotted in Figs. 25.11 and 25.12, indicate that the relative importance of contamination in certain foods differs for different age groups, as might be expected. They also indicate how this variation may be taken into account in assessing the importance of such contamination in various foods and for the various segments of the population. For example, it appears that variation with age in the dose commitment per day of exposure due to a specified level of contamination of whole milk by ^{137}Cs varies by a factor of 2 or less if infants drank whole milk. If the infants consume formula milk or evaporated milk, which may be at quite a different level of ^{137}Cs concentration than whole milk, they may

[15] S. S. Brar *et al.*, *Health and Safety Laboratory Fallout Program Quart. Summary Rept. April 1, 1967*, HASL-181, p. III-134.

[16] S. S. Brar and D. M. Nelson, *Health and Safety Laboratory Fallout Program Quart. Summary Rept. April 1, 1968*, HASL-193, p. III-29.

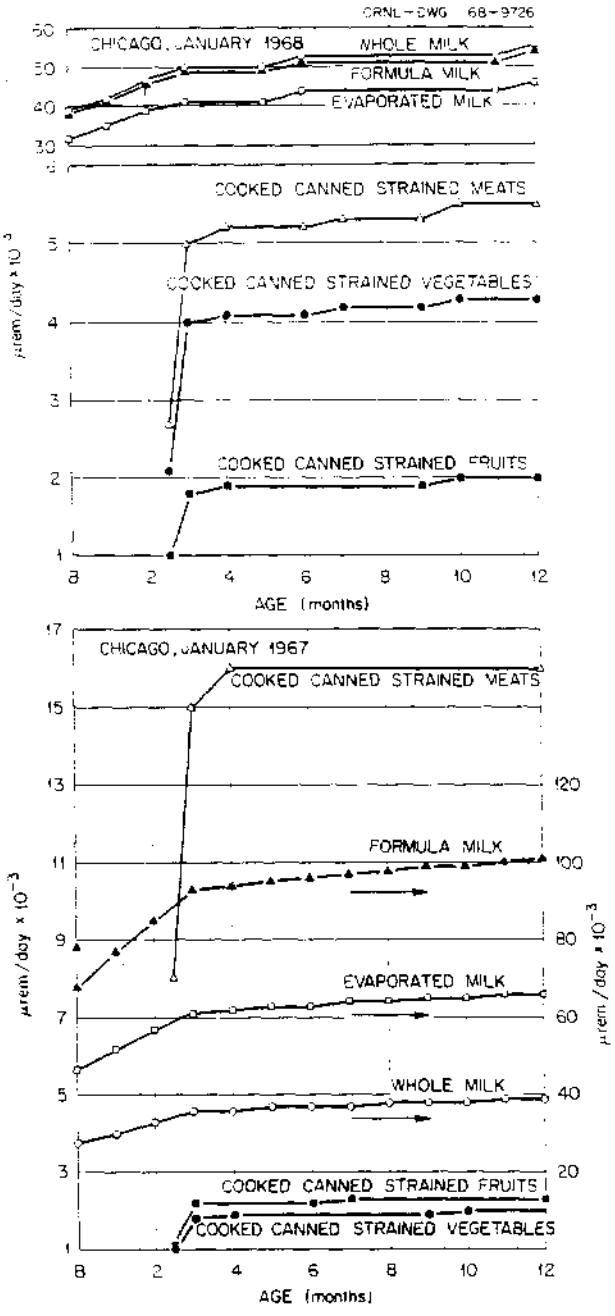


Fig. 25.12. Dose Commitment of Exposure from Fallout ^{137}Cs via Various Foods (Newborn to One Year).

have a higher dose commitment. The corresponding dose estimates due to ^{137}Cs present in meat vary widely with the age of the exposed individual. These data call attention to the rather great differences in dose commitment per day of intake with respect to age for these different foods. The trend is different when meat is the pathway for fallout ^{137}Cs intake than when milk is the pathway. Since the infant eats a very small amount of meat, his dose is quite small compared with the dose which an adult would receive, assuming that the meat was contaminated uniformly. If whole milk is contaminated, the newborn infant is low. Data are plotted in Fig. 25.12 for the infant drinking formula milk, evaporated milk, or whole milk and illustrate the wide differences to be expected.

These data are offered only as an example of the type of calculations that may be used for assessing population exposures from foods or fluids. The numerical values presented depend on estimates of intake and of contamination levels which may not be typical of other populations or other times, but the method applies for any population if the relevant data are available.

Before the age dependence problem is resolved completely, questions must be answered concerning (1) variability of the absorption of cesium into the blood when incorporated in various foodstuffs, (2) possible age dependence of this absorption factor, and (3) special diets of certain population groups.

CALCULATION OF MICROCURIE-DAYS RESIDENCE IN AN ORGAN RESULTING FROM A SINGLE INTAKE

S. R. Bernard

A computer program has been written to calculate microcurie-days, that is, the area beneath the retention curve when 1 μc of a radionuclide is initially taken into an organ or into the whole body. The microcurie-days of residence accumulated up to time t, $Q(t)$, arises in the equation for estimating the dose equivalent in rems accumulated up to time t. The daily dose rate at time t, $D_d(t)$, in rems per day received by an organ of mass m grams containing $q(t)$ microcuries of a radionuclide at time t having an effective energy of ϵ Mev per disintegration is given by the formula

$$D_d(t) = 51\ q(t)\ \epsilon/m\ , \qquad (1)$$

where 51 is the (gram-rems/microcurie-day)/(effective Mev/disintegration). The integral of Eq. (1) from time 0 to t,

$$D(t) = \int_0^t D_d(\tau)\ d\tau = 51 \int_0^t \frac{\epsilon q(\tau)}{m} d\tau\ , \qquad (2)$$

yields the dose equivalent accumulated up to time t, $D(t)$, in rems. In Eq. (2), ϵ will usually be a constant, and $q(t)/m$ is the concentration of the radionuclide in the organ at time t. For adult persons, m, the mass of the organ, is taken as a constant, and when it and ϵ are factored out, the integral (2) becomes

$$D(t) = \frac{51\epsilon}{m}\int_0^t q(\tau)\, d\tau = \frac{51\epsilon}{m} Q(t) . \qquad (3)$$

Here, $Q(t)$ is the area beneath the retention curve $q(t)$, that is, the microcurie-days residence up to time t.

When a parent of a decay chain of radionuclides is taken into the body, the daughter elements of the chain grow in with time, and the dose equivalent for each member of the chain must be estimated in order to estimate the accumulated dose or dose equivalent to an organ. For the ith member of the chain the microcurie-days of residence in an organ is given by

$$Q_i(t) = \int_0^t q_i(\tau)\, d\tau .$$

Assuming ϵ_i and m constant, as before, the dose equivalent $D(t)$ in rems due to the entire chain is given by

$$D(t) = \frac{51}{m} \sum_{i=0}^{n} \epsilon_i Q_i(t) . \qquad (4)$$

In order to calculate $Q_i(t)$ we require metabolic models, sometimes taken as sums of exponential terms, to approximate the retention of the radionuclide in the organ for extended periods of time. We list here the equations for the functions $Q_i(t)$ for the case in which the ith daughter element in the chain is only introduced into the organ by decay of its parent present in that organ. To this end let

$$R_i^s(t) = \sum_{j_i=1}^{n_i} a_{ij_i} e^{-\lambda^b_{ij_i} t} , \quad i = 0, 1, \ldots, n , \qquad (5)$$

denote the fractional biological retention in an organ (i.e., the retention equation of the organ considered, corrected for radiological decay and normalized to 1 at $t = 0$) of the ith radionuclide. That is, if a unit amount of the ith daughter enters the organ at time $t = 0$, the amount present t days later is $R_i^s(t)$. In (5), the constant

$$\lambda^b_{ij_i}$$

represents the rate of biological elimination of the j_ith component of the retention.

At present the retention function for an organ generally is estimated from experimental data in which a single injection is made initially into the bloodstream. For the parent of the chain,

$$q_0(t) = q_0(0) e^{-\lambda_0^r t} R_0^s(t) , \qquad (6)$$

where $q_0(0)$ is the microcuries initially reaching the organ and λ_0^r is the radiological decay constant. For the microcuries of the first daughter, $q_1(t)$, present in the organ, we assume linear superposition of inputs. The rate of production of daughter in $d\tau$ units of time is $\lambda_1^r q_0(\tau)\, d\tau$. The amount of daughter produced in the time interval $d\tau$ which is present at a later time t is then

$$\lambda_1^r q_0(\tau)\, d\tau\, e^{-\lambda_1^r (t-\tau)} R_1^s(t-\tau) ,$$

where $t \geq \tau$. We linearly superpose the incremental inputs, that is, integrate over $0 \leq \tau \leq t$, to obtain

$$q_1(t) = \lambda_1^r \int_0^t q_0(\tau)\, d\tau\, e^{-\lambda_1^r (t-\tau)} R_1^s(t-\tau) \qquad (7)$$

$$= \lambda_1^r q_0(0) \int_0^t R_0(\tau)\, d\tau\, e^{-\lambda_1^r (t-\tau)} R_1^s(t-\tau) ,$$

where

$$R_0(t) = e^{-\lambda_0^r t} R_0^s(t) = \frac{q_0(t)}{q_0(0)} . \qquad (8)$$

For the level of the second daughter in the organ, $q_2(t)$, we write

$$q_2(t) = \lambda_2^r \int_0^t d\tau\, q_1(\tau)\, e^{-\lambda_2^r (t-\tau)} R_2^s(t-\tau) ,$$

where $\lambda_2^r\, d\tau\, q_1(\tau)$ is the amount of the second daughter produced in $d\tau$ units of time and

$$\lambda_2^r\, d\tau\, q_1(\tau)\, e^{-\lambda_2^r (t-\tau)} R_2^s(t-\tau)$$

is the amount present at a later time t. In general for the level of the ith daughter of the chain present in the organ we would find

$$q_i(t) = \lambda_i^r q_0(0) \times \int_0^t R_{i-1}(\tau)\, e^{-\lambda_i^r(t-\tau)}\, R_i^s(t-\tau)\, d\tau \quad (8a)$$

as the expression for the microcuries of the ith radionuclide in the chain present in the organ at time t. Here $R_i(t) = q_i(t)/q_0(0)$.

When the $R_i^s(t)$'s are given by Eq. (5), $q_i(t)$ may be obtained in closed form, that is,

$$q_i(t) = q_0(0)\left(\prod_{k=1}^{i} \lambda_k^r\right) \times \sum_{j_0=1}^{n_0} \sum_{j_1=1}^{n_1} \cdots \sum_{j_i=1}^{n_i} \left(\prod_{h=0}^{i} a_{hj_h}\right) \times \left[\sum_{h=0}^{i} \frac{e^{-\lambda_{hj_h} t}}{\prod_{\substack{p=0 \\ h\neq p}}^{i} \left(\lambda_{pj_p} - \lambda_{hj_h}\right)}\right], \quad i = 1, 2, \ldots, n, \quad (9)$$

where

$$\lambda_{ij_i} = \lambda_i^r + \lambda_{ij_i}^b$$

is the effective elimination constant. Thus, the functions $Q_i(t)$ can be obtained from (6) for $i = 0$ and from (9) for all other i by straightforward integration.

In some cases it suffices to consider in the computer program only the special case when

$$R_i^s(t) = a_{ij_i} \exp\left(-\lambda_{ij_i} t\right),$$

that is, the longest-lived exponential (let it be the n_ith one). In this case, the sum of exponentials in (9) is the solution of the Bateman differential equations

$$\frac{d}{dt} q_i(t) = a_{in_i} \lambda_i^r q_{i-1}(t) - \lambda_{in_i} q_i(t), \quad i = 1, 2, \ldots, n, \quad (10)$$

$$\frac{dq_0(t)}{dt} = -\lambda_{0n_0} q_0(t).$$

After integration and rearrangement, we find

$$Q_i(t) = \frac{a_{in_i} \lambda_i^r Q_{i-1}(t) - q_i(t)}{\lambda_{in_i}}, \quad (11)$$

$$Q_0(t) = \int_0^t d\tau\, q_0(\tau) = \frac{q_0(0)}{\lambda_{0n_0}}[1 - R_0(t)].$$

In the above equations $q_i(t)$ is to be estimated from Eq. (9) remembering that we only take the n_ith term in the series. Thus, from (11) and (9) the microcurie-days can be estimated, sequentially. In the simplified single-exponential case just discussed, these fractions are analogous to the f_2's in the current ICRP Committee II handbook (*Health Phys.* 3, 1960).

We also mention that formula (8*a*) does not require that we restrict ourselves to exponential sums for $R_i^s(t)$. We can use a power function, $a_i t^{-b_i}$ (a_i and b_i are constants), for the $R_i^s(t)$. However, when we do this we do not obtain a closed form expression similar to (9). If the power function can be adequately represented by a finite sum of exponentials as suggested in the work of Bernard and Davis,[17] then the closed form expression, Eq. (9), will apply.

The computer code has been written by Sharron P. King (ORNL Mathematics Division) to evaluate the case of retention described by a single exponential term, Eqs. (9) and (11). Also, the extension of the code to handle Eq. (9) for multiexponential retention is presently under way.

A MODEL FOR ESTIMATION OF MEAN ACTIVITY PRESENT IN SEGMENTS OF THE GASTROINTESTINAL TRACT AT ANY TIME POST INGESTION

S. R. Bernard

In the papers by Eve[18] and Dolphin and Eve[19] an extensive review of the physiology of the gastro-

[17] *Health Phys. Div. Ann. Progr. Rept. July 31, 1966*, ORNL-4007, p. 232.

[18] I. S. Eve, *Health Phys.* 12, 131 (1966).

[19] G. W. Dolphin and I. S. Eve, *Health Phys.* 12, 163 (1966).

intestinal tract is carried out, and a new model for the dosimetry of the GI tract is presented. In their model they estimate the radiation dose to a small element of the gut walls in a segment of the tract. They compute a time average of this dose during the assumed residence time in the segment of the tract and consider this to be the mean dose throughout that section of the GI tract. Another model for residence of material in the GI tract is developed here and may serve as a basis for estimating the mean dose to segments of the tract. This other model is based on considerations of the random variable passage time through the tract.

In order to explain the basis of this other model, consider the case of passage of a stable element through the tract. Let T_1, T_2, T_3, and T_4 denote the random variable residence times of this stable element in the stomach, small intestine, upper large intestine, and lower large intestine respectively. The total time for the passage of the stable element is the sum of these random variable times, denoted by t, that is,

$$t = \sum_{i=1}^{4} T_i . \qquad (1)$$

If the residence times in the various segments are assumed to be independent and exponentially distributed, then the probability density functions of these residence times and of the total time t satisfy the same differential equations that arise from consideration of a catenary compartment model of the tract with the flows being unidirectional. We develop here the equations along the more usual lines of compartment theory and only point out that the same equations and estimates can be given a probabilistic interpretation. Figure 25.13 shows the model; $S_{ij}(t)$ denotes the amount (in microcuries) of the ith radionuclide ($i = 0, 1, \ldots, n$) in a chain of radionuclides present in the jth region ($j = 1, 2, 3, 4$) of the GI tract at time t. Thus, when the subscript j has values 1, 2, 3, or 4 the designated quantity relates to the stomach, small intestine, upper large intestine, and lower large intestine respectively. The symbol λ_{ij}^{a} ($j = 1, \ldots, 4$) denotes the fractional rate of loss from compartment j by absorption into the bloodstream, λ_j^b is the fractional rate of transfer from the jth compartment to the $(j-1)$th compartment, and λ_i^r is the radiological decay constant of the ith radionuclide in a decay chain.

The rate of change of the ith radionuclide in the jth compartment is given by

$$\frac{dS_{ij}(t)}{dt} = \lambda_i^r S_{i-1,j}(t) + \lambda_{j-1}^b S_{i,j-1}(t) - \lambda_{ij} S_{ij}(t) ,$$

$$i = 0, 1, \ldots, n, \; j = 1, \ldots, 4 . \qquad (2)$$

Here

$$\lambda_{ij} = \lambda_i^r + \lambda_j^b + \lambda_{ij}^a$$

is the effective elimination constant of the ith radionuclide from the jth compartment. In the differential equation (2) when $i = 0$ the term $S_{-1,j}(t) = 0$ for all t and all j, and for $j = 1$, $S_{i,-1}(t) = 0$ for all i and all t. The reader should be warned too that this equation (2) only applies when the units on the $S_{ij}(t)$ are microcuries. The values of the coefficients in the right members might be different if the $S_{ij}(t)$ are expressed in other units. We have used the Laplace transform method to obtain the general solution to these equations. The solution, assuming $S_{01}(0) = 1$, all other $S_{ij}(0) = 0$, for the level of the ith radionuclide in the stomach is

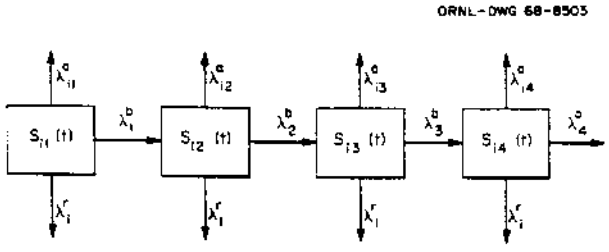


Fig. 25.13. A Catenary Compartment Model for GI Tract.

$$S_{i1}(t) = \left(\prod_{k=1}^{i} \lambda_k^r \right) \sum_{p=0}^{i} \frac{e^{-\lambda_{p1} t}}{\prod_{\substack{k=0 \\ k \neq p}}^{i} (\lambda_{k1} - \lambda_{p1})} , \quad i = 0, 1, \ldots, n .$$

For the level of the ith radionuclide in the small intestine

$$S_{i2}(t) = \lambda_1^b \left(\prod_{k=1}^{i} \lambda_k^r \right) \sum_{l=0}^{i} \left[\sum_{p=0}^{l} \frac{e^{-\lambda_{p1} t}}{\prod_{\substack{k=0 \\ k \neq p}}^{l} (\lambda_{k1} - \lambda_{p1}) \prod_{k=l}^{i} (\lambda_{k2} - \lambda_{p1})} + \sum_{p=l}^{i} \frac{e^{-\lambda_{p2} t}}{\prod_{k=0}^{l} (\lambda_{k1} - \lambda_{p2}) \prod_{\substack{k=l \\ k \neq p}}^{i} (\lambda_{k2} - \lambda_{p2})} \right] , \quad i = 0, 1, \ldots n .$$

For the level of the ith radionuclide in the upper large intestine

$$S_{i3}(t) = \lambda_1^b \lambda_2^b \left(\prod_{k=1}^{i} \lambda_k^r \right) \sum_{m=0}^{i} \sum_{l=0}^{m} \left[\sum_{p=0}^{l} \frac{e^{-\lambda_{p1} t}}{\prod_{\substack{k=0 \\ k \neq p}}^{l} (\lambda_{k1} - \lambda_{p1}) \prod_{k=l}^{m} (\lambda_{k2} - \lambda_{p1}) \prod_{k=m}^{i} (\lambda_{k3} - \lambda_{p1})} - \sum_{p=l}^{m} \frac{e^{-\lambda_{p2} t}}{\prod_{k=0}^{l} (\lambda_{k1} - \lambda_{p2}) \prod_{\substack{k=l \\ k \neq p}}^{m} (\lambda_{k2} - \lambda_{p2}) \prod_{k=m}^{i} (\lambda_{k3} - \lambda_{p2})} + \sum_{p=m}^{i} \frac{e^{-\lambda_{p3} t}}{\prod_{k=0}^{l} (\lambda_{k1} - \lambda_{p3}) \prod_{k=l}^{m} (\lambda_{k2} - \lambda_{p3}) \prod_{\substack{k=m \\ k \neq l}}^{i} (\lambda_{k3} - \lambda_{p3})} \right] , \quad i = 0, 1, \ldots, n .$$

For the lower large intestine the level of the ith radionuclide is

$$S_{i4}(t) = \lambda_1^b \lambda_2^b \lambda_3^b \left(\prod_{k=1}^{i} \lambda_k^r \right) \sum_{h=0}^{i} \sum_{m=0}^{h} \sum_{l=0}^{m} \left[\sum_{p=0}^{l} \frac{e^{-\lambda_{p1} t}}{\prod_{\substack{k=0 \\ k \neq p}}^{l} (\lambda_{k1} - \lambda_{p1}) \prod_{k=l}^{m} (\lambda_{k2} - \lambda_{p1}) \prod_{k=m}^{h} (\lambda_{k3} - \lambda_{p1}) \prod_{k=h}^{i} (\lambda_{k4} - \lambda_{p1})} \right.$$

$$+ \sum_{p=l}^{m} \frac{e^{-\lambda_{p2} t}}{\prod_{k=0}^{l} (\lambda_{k1} - \lambda_{p2}) \prod_{\substack{k=l \\ k \neq p}}^{m} (\lambda_{k2} - \lambda_{p2}) \prod_{k=m}^{h} (\lambda_{k3} - \lambda_{p2}) \prod_{k=h}^{i} (\lambda_{k4} - \lambda_{p2})}$$

$$+ \sum_{p=m}^{h} \frac{e^{-\lambda_{p3} t}}{\prod_{k=0}^{i} (\lambda_{k2} - \lambda_{p3}) \prod_{k=l}^{m} (\lambda_{k2} - \lambda_{p3}) \prod_{\substack{k=m \\ k \neq p}}^{h} (\lambda_{k3} - \lambda_{p3}) \prod_{k=h}^{i} (\lambda_{k4} - \lambda_{p3})}$$

$$\left. + \sum_{p=h}^{i} \frac{e^{-\lambda_{p4} t}}{\prod_{k=0}^{i} (\lambda_{k1} - \lambda_{p4}) \prod_{k=l}^{m} (\lambda_{k2} - \lambda_{p4}) \prod_{k=m}^{h} (\lambda_{k3} - \lambda_{p4}) \prod_{\substack{k=h \\ k \neq p}}^{i} (\lambda_{k4} - \lambda_{p4})} \right], \quad i = 0, 1, \ldots, n.$$

We have tested this model with the data of Hayes *et al.*,[20] who fed single intakes of ^{140}La (in a citrate) to patients and collected feces, which were then analyzed for the ^{140}La content. The radiological half-life of ^{140}La is ~40 hr, but Hayes *et al.* corrected the excretion data by the decay factor $e^{-0.693t/40}$ (where t is in hours), and thus their data correspond to a stable element. The mean residence times assigned by Eve for the different segments of the tract are $^1/_{24}$, $^4/_{24}$, $^{13}/_{24}$, $^{24}/_{24}$ days and consequently λ_j^b has been taken here to be 24, 6, 1.85, 1 day^{-1}, $j = 1, 2, 3, 4$, in testing the model.

Using these values and the further conditions that the stomach receives an initial input of 1 μc, all other sections have zero contents at time zero, taking $\lambda_a^j = 0$ for all j, then

$$S_{01}(t) = 1e^{-(24+\lambda_0^r)t}, \tag{5}$$

$$S_{02}(t) = 1.33\,(e^{-6t} - e^{-24t})\,e^{-\lambda_0^r t}, \tag{6}$$

$$S_{03}(t) = (+0.36117e^{-24t} - 1.92771e^{-6t} + 1.56653e^{-1.85t})\,e^{-\lambda_0^r t}, \tag{7}$$

$$S_{04}(t) = (-0.02905e^{-24t} + 0.71325e^{-6t} - 3.40952e^{-1.85t} + 2.725831e^{-t})\,e^{-\lambda_0^r t}, \tag{8}$$

while the fecal excretion is given by

$$\int_0^t 1 \cdot S_{04}(\tau)\, d\tau .$$

For $\lambda_0^r = 0$ this integral becomes

$$[-0.00121\,(1 - e^{-24t}) + 0.11887\,(1 - e^{-6t}) - 1.84298\,(1 - e^{-1.85t}) + 2.72531\,(1 - e^{-t})]. \tag{9}$$

Note that for this example, we assume no absorption from any regions of the tract into the bloodstream. Therefore (9) is applicable to the case of a single intake of a long-lived nonabsorbed substance, such as used by Hayes *et al.*[20] With their

[20] R. L. Hayes *et al.*, *Health Phys.* **9**, 915 (1963).

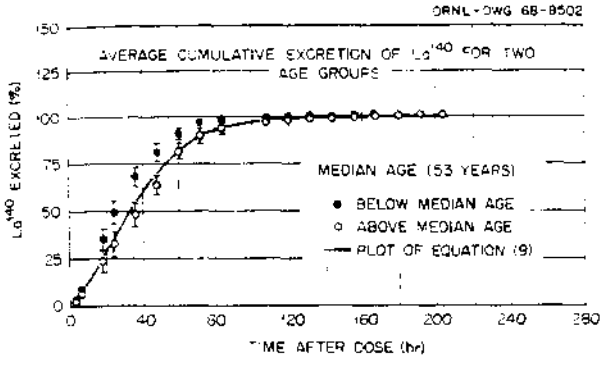


Fig. 25.14. Hayes *et al.* Average Cumulative Excretion of ^{140}La by Human Subjects Compared with Cumulative Excretion from Eq. (9).

permission, Fig. 25.14 is reproduced from ref. 20 and shows a plot of the cumulative excretion expressed in percent of that ingested at the various times after ingestion. Also shown are the estimates made by the authors from Eq. (9) [i.e., a plot of Eq. (9) as a function of time]. As can be seen, the agreement is fairly good. Some data on the levels in each region of the tract would be required to test this model adequately and see how good the estimate is in comparison with actual amounts observed at various times after ingestion of various soluble and insoluble substances.

This model permits one to estimate the mean contents of the various segments of the tract at any time following ingestion. Since an estimate of the levels of activity in the regions of the GI tract section is available, the energy deposition averaged over the entire section can be estimated rather than the peak dose or the time average over the region. However, the assumption of the exponential distributions of residence time and their independence in the various regions of the tract needs to be examined and tested with more experimental data. This model is not adequate for estimating the probability density that a given content of a radionuclide is present in a section at a given time for a given person. It only purports to give the average levels or expected levels to be found in the various sections at various times for a person having the assumed characteristics, that is, for a "Standard Man."

NUCLEAR SAFETY ENVIRONMENTAL STUDIES: BIOHAZARDS OF AEROSPACE NUCLEAR SYSTEMS

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Microspheres of $^{238}PuO_2$ having diameters of from 50 to 250 μ are used in space electric power sources. One of the greater potential hazards they might present in the event of their accidental release to the biosphere is to the respiratory system. The probability of entry into the body through the nose is a function of the difference between the intake velocity of the airstream into the nose and the falling velocity of the particle. The ratio of the concentration of inhaled particles to the ambient concentration as a function of particle size may be calculated as follows: Let

$$U_t = Q/A \ ,$$

where Q = air flow rate at a time t, cm^3/sec, A = cross-sectional area of external nares, cm^2, and U_t = flow velocity at time t, cm/sec.

If the particles are assumed to be uniformly mixed and suspended in the exposure volume, the rate at which they enter the nares will be nU_tA (particles/sec), where n is the number concentration (particles/cm^3). However, if, as in the usual case, the particles have a downward velocity $-V_s$, then only $n(U_t - V_s)A$ particles/sec would gain entry. An efficiency factor e_s can then be calculated:

$$e_s = \frac{n(U_t - V_s)A}{nU_tA} \ ,$$

which gives the fraction of particles entering compared with the number that would have entered if they had zero settling velocity. This factor, e_s, is then adjusted for the fraction of time during inspiration that U_t occurs.

Using the respiratory air flow curves of Silverman and Billings[21] for four types of inspiratory cycles and assuming a fixed intake area of 2.5 cm^2, the

[21] L. Silverman and C. E. Billings, "Pattern of Airflow in the Respiratory Tract," *Symposium on Inhaled Particles and Vapours*, C. N. Davies, ed., Pergamon, New York, 1961.

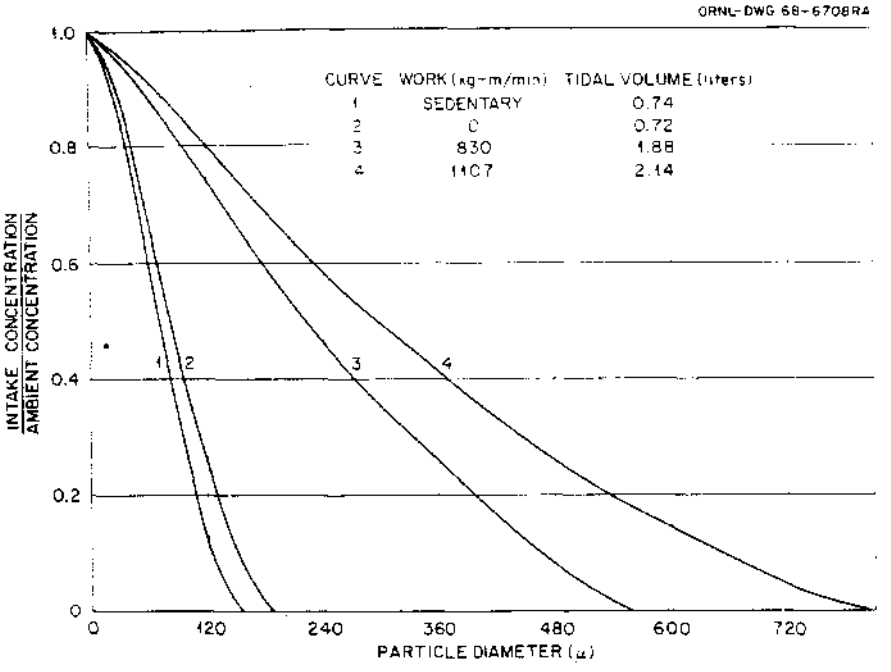

Fig. 25.15. Fraction of Plutonium Microspheres Inspired.

curves shown in Fig. 25.15 have been computed. Two factors which may be expected to give slightly lower values than those shown have been ignored: (1) the shadowing effect of the nose and (2) dilation of the nostrils and/or mouth breathing. However, these are at least partially offset by the assumption of a zero horizontal component in the path of the particle.

Using thin plastic scintillators[22] the alpha dose rate from $^{238}PuO_2$ microspheres has been measured as a function of depth in a tissue-simulating absorber (Mylar). Figure 25.16 shows average values found, and the interception of the dashed line with the abscissa is an estimate of the maximum range to be expected from a thin source. The average surface dose rate from five microspheres ranging from 123 to 225 μ diameter was 25,900 ± 2600 rads/sec.

INTERACTION OF AIRBORNE PARTICULATES WITH GASES

B. M. Smith[23] B. R. Fish

Studies are being conducted on the interaction of sulfur dioxide gas with airborne particles. Aerosols are generated by exploding wires in air, resulting in particles with diameters from about 0.01 to 0.1 μ (ref. 24). Aerosol is mixed with SO_2 tagged[25] with ^{35}S under controlled conditions in a gas-particle interaction chamber.[26] Samples for analysis are withdrawn from the chamber through diffusion tubes followed by membrane filters. The tubes are lined with lead foil coated with lead peroxide. The sample flow rate is selected so that SO_2 gas will diffuse to and be removed by the lead peroxide on the inside walls of the tubes while allowing aerosol particles to pass through the tube and be collected on the filters. For analysis, the lead-foil inserts are segmented and assayed for sulfur by radiochemical techniques. Particle samples are assayed for sorbed SO_2 in a similar manner.

[22] W. H. Wilkie and B. R. Fish, "Scintillation Extrapolation Dosimetry," *Symposium on Solid State and Chemical Dosimetry in Medicine and Biology*, IAEA, Vienna, 1966.

[23] Employee of National Air Pollution Control Administration (NAPCA), CPEHS, on assignment to ORNL.

[24] K. D. Duerksen *et al.*, *Health Phys. Div. Ann. Progr. Rept. July 31, 1967*, ORNL-4168, pp. 296–98.

[25] B. M. Smith *et al.*, *Health Phys. Div. Ann. Progr. Rept. July 31, 1967*, ORNL-4168, pp. 299–301.

[26] R. L. Walker, *Health Phys. Div. Ann. Progr. Rept. June 30, 1963*, ORNL-3492, pp. 187–89.

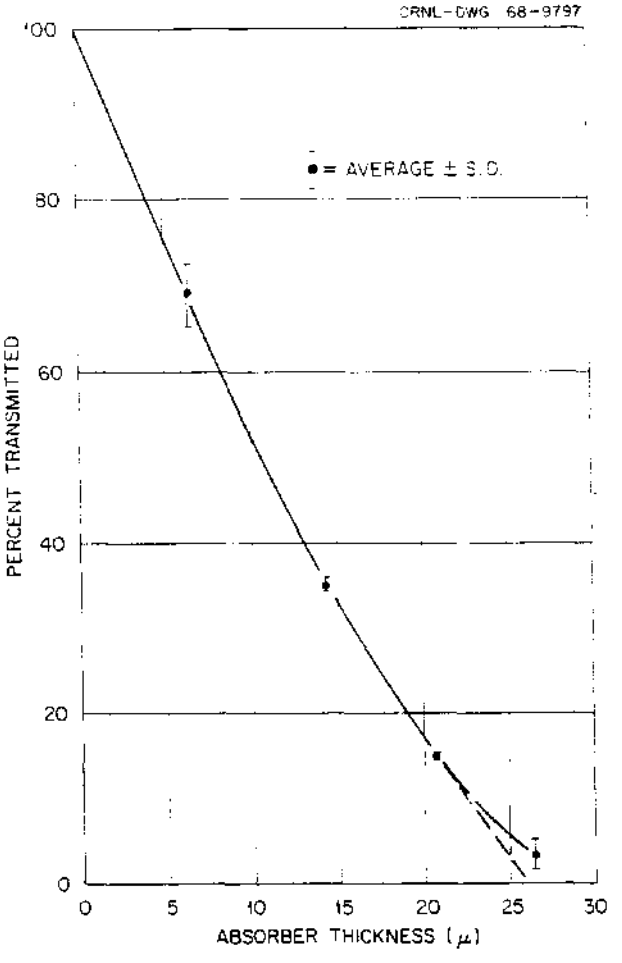

Fig. 25.16. Transmission of Alpha Rays from $^{238}PuO_2$ Microspheres Through Mylar.

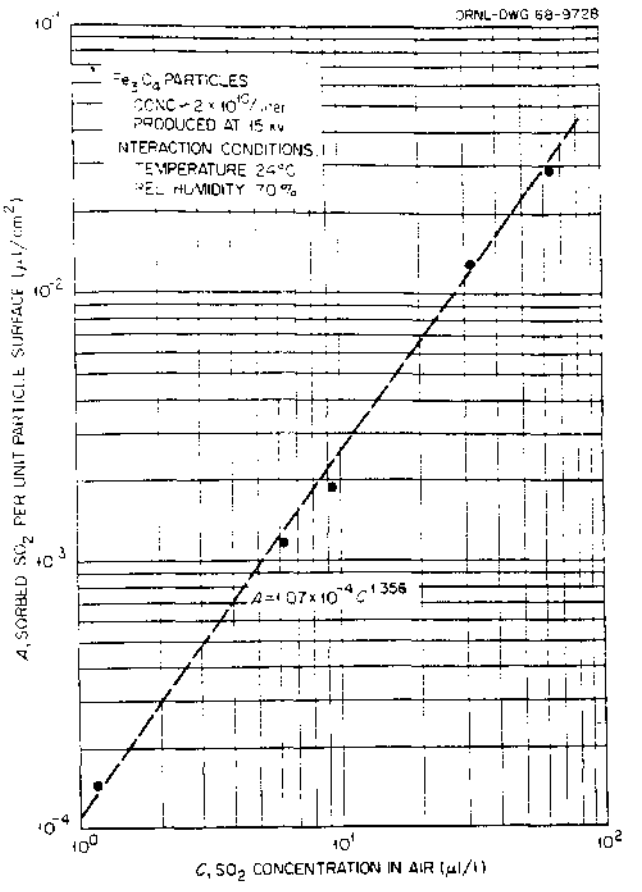

Fig. 25.17. SO_2 Sorption Isotherm. Particles dispersed in air.

Sorption of SO_2 gas on aerosol particles of lead oxides, aluminum oxides, platinum, and iron oxide has been studied. Lead oxide particles in concentrations of approximately 10^{10} particles per liter of air sorb essentially 100% of the SO_2 gas at 1 μl of SO_2 per liter of air under ambient conditions, whereas aluminum oxides and platinum particles sorb approximately 50% and 10% respectively. Preliminary indications are, however, that platinum reversibly sorbs SO_2 gas, which is subsequently released during sampling. Iron oxide (Fe_3O_4) particles sorb varying amounts of SO_2 gas in accordance with initial SO_2 concentration. A sorption isotherm for SO_2 gas on Fe_3O_4 particles in the disperse phase is presented in Fig. 25.17.

26. Stable Element Metabolism

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Mary Jane Cook
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Peggy L. Stewart[1]
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PATTERNS OF ELEMENTAL EXCRETION IN LONG-TERM BALANCE STUDIES

Isabel H. Tipton Peggy L. Stewart
Judith Dickson

In the program for analysis of total self-chosen diets and total excreta for trace elements, the material from two men for 50 weeks has been analyzed for 25 elements. Statistical treatment has been made for the 250-day period September 6, 1966–May 13, 1967.

Subject C was 23 years old, 6 ft 1½ in. tall, and weighed about 220 lb. Subject D was 25 years old, 5 ft 10 in. tall, weighing about 156 lb.

Differences in the dietary habits of the two individuals are indicated by the daily gross amounts of food and drink they ingested and their fecal and urinary output. Table 26.1 shows the mean and standard deviation of the total daily dietary intake, the water content of this diet, and the daily fecal and urinary output. It is obvious that subject C drank more than subject D, and this is also reflected in his greater urinary output. Subject D, however, drank more milk than subject C, and this is reflected in the values for calcium intake. The large standard deviation for fecal excretion comes about because of the number of days on which there was no fecal excretion at all. For subject C, on more than half of the days in the period there was no fecal excretion, and there were a few periods of four days when none occurred.

Table 26.1. Gross Daily Intake and Excretion

September 6, 1966 – May 13, 1967

Mean ± standard deviation in grams

	Subject C	Subject D
Total daily intake by ingestion	3100 ± 530	2700 ± 610
Water content	2600 ± 690	2100 ± 510
Total daily fecal excretion	110 ± 130	120 ± 110
Total daily urinary excretion	1300 ± 500	1000 ± 270

Table 26.2 shows the total dietary intake and fecal and urinary output of 25 elements for this period minus three days at the beginning. The errors of the method for most elements result in an uncertainty of about ±20% in the state of balance. Therefore balance has been expressed as the ratio total excreta/total ingested rather than as the more commonly used differences between total ingested and total excreted, so that the state of balance can be more conveniently observed. If this value is between 0.8 and 1.2, the subject may be considered to have been in balance over the period under study. If the ratio is below 0.8, the intake was greater than the output, and the subject was in "positive" balance. If the ratio is above 1.2, the output exceeded the input, and the subject was in "negative" balance. The ratio of the urinary output to total excreta or to intake by ingestion is sometimes used as the lower limit of f_1, the fraction of an element absorbed from the

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Table 26.2. Total Intake by Ingestion and Total Fecal and Urinary Excretion

September 9, 1966–May 13, 1967 (247 days)

Element	Subject	Total Ingested (g)	Total in Feces (g)	Total in Urine (g)	Total in Excreta (g)	Total Excreta / Total Ingested	Urine / Excreta	Urine / Ingested	f_1 (ICRP-59)
Aluminum	C	3.9	4.0	0.23	4.2	1.08	0.056	0.060	0.1
	D	3.8	2.8	0.19	3.0	0.79	0.063	0.050	
Barium	C	0.16	0.15	0.0028	0.15	0.95	0.018	0.018	0.05
	D	0.21	0.15	0.0034	0.15	0.73	0.022	0.040	
Beryllium	C	0.027	0.0018	0.00055	0.0023	0.086	0.24	0.020	0.002
	D	0.056	0.0012	0.00044	0.0016	0.03	0.27	0.0079	
Boron	C	0.28	0.067	0.30	0.36	1.27	0.81	1.04	0.9
	D	0.60	0.096	0.21	0.30	0.51	0.68	0.35	
Calcium	C	240	210	26	240	1.0	0.11	0.11	0.6
	D	530	290	34	320	0.62	0.11	0.067	
Cadmium[a]	C	0.0069	0.0020	0.0055	0.0075	1.1	0.74	0.79	0.0025
	D	0.012	0.0026	0.0057	0.0083	0.69	0.69	0.47	
Chromium	C	0.059	0.017	0.021	0.038	0.65	0.55	0.35	<0.005
	D	0.084	0.021	0.024	0.045	0.53	0.54	0.28	
Cobalt	C	0.083	0.0086	0.070	0.079	0.95	0.89	0.85	0.3
	D	0.12	0.014	0.064	0.078	0.67	0.82	0.56	
Copper	C	0.25	0.31	0.0055	0.32	1.28	0.018	0.022	0.28
	D	0.45	0.29	0.013	0.30	0.68	0.048	0.028	
Iron	C	3.7	3.4	0.23	3.6	0.98	0.064	0.062	0.1
	D	7.6	2.4	0.21	2.6	0.34	0.082	0.028	
Lead[b]	C	0.072	0.019	0.0074	0.027	0.37	0.28	0.10	0.08
	D	0.11	0.016	0.0084	0.024	0.22	0.34	0.075	
Magnesium	C	47	40	29	69	1.47	0.42	0.62	0.1
	D	92	46	25	71	0.78	0.35	0.28	
Manganese	C	0.79	0.67	0.0086	0.68	0.85	0.013	0.011	0.1
	D	1.3	0.68	0.0065	0.69	0.54	0.0095	0.005	
Molybdenum	C	0.068	0.027	0.035	0.062	0.92	0.56	0.52	0.8
	D	0.15	0.030	0.042	0.072	0.48	0.58	0.28	
Nickel	C	0.10	0.057	0.029	0.086	0.82	0.34	0.28	0.3
	D	0.20	0.092	0.031	0.12	0.61	0.25	0.15	
Phosphorus	C	410	110	310	420	1.02	0.74	0.75	0.75
	D	820	140	290	430	0.53	0.67	0.35	
Potassium	C[c]	410	63	660	720	1.75	0.91	1.60	1.0
	D	900	72	740	810	0.90	0.91	0.82	
Silver	C	0.046	0.0083	0.0018	0.010	0.22	0.18	0.039	0.01
	D	0.099	0.031	0.0019	0.033	0.34	0.059	0.020	
Sodium	C[c]	840	8.2	1200	1200	1.45	0.99	1.44	1.0
	D	1200	9.6	1200	1200	1.0	0.99	1.0	

Table 26.2 (continued)

Element	Subject	Total Ingested (g)	Total in Feces (g)	Total in Urine (g)	Total in Excreta (g)	Total Excreta / Total Ingested	Urine / Excreta	Urine / Ingested	f_1 (ICRP-59)
Strontium	C	0.41	0.34	0.028	0.37	0.91	0.081	0.088	0.3
	D	0.53	0.30	0.020	0.32	0.60	0.062	0.037	
Tin	C	1.5	1.1	0.026	1.1	0.72	0.024	0.018	0.05
	D	2.4	1.0	0.016	1.0	0.43	0.016	0.0068	
Titanium	C	0.20	0.11	0.096	0.20	1.01	0.47	0.47	10^{-4}
	D	0.51	0.22	0.088	0.31	0.61	0.28	0.17	
Vanadium	C	0.016	0.036	0.0052	0.041	2.48	0.13	0.32	0.02
	D	0.040	0.042	0.0073	0.049	1.23	0.15	0.18	
Zinc	C	2.7	3.6	0.38	4.0	1.48	0.094	0.14	0.1
	D	4.5	4.0	0.35	4.3	0.96	0.080	0.076	
Zirconium	C	0.12	0.036	0.024	0.060	0.50	0.39	0.20	$<10^{-4}$
	D	1.60	0.048	0.034	0.082	0.52	0.41	0.21	

[a] Cadmium data for weeks 28–36 only.

[b] Lead data for weeks 19–36 only.

[c] Subject C failed to add condiments to his duplicate meal. After he corrected this practice the apparent imbalance disappeared.

gastrointestinal tract of the total ingested. These ratios are given in Table 26.2. The ICRP Handbook 59 values for f_1 are included in the last column of the table for comparison. The agreement is good for those elements which are excreted almost entirely in urine, but it is obvious that for elements like strontium and calcium, the amount excreted in urine does not indicate the total amount absorbed, since some of the absorbed element is known to be reexcreted into the gut.

The daily variation makes it possible to study the correlation between elemental intake and excretion. Correlations between levels of excretion of an element and levels of intake by ingestion on previous days may be determined and multiple regressions obtained. If y represents the excretion of an element for any day and x_1, x_2, x_3, x_4, ... represent the dietary intake on the same day, the day before, two days before, three days before, etc., it is possible to write an equation which will predict the excretion in terms of the intake:

$$y = a_0 + a_1x_1 + a_2x_2 + a_3x_3 + \ldots ,$$

where a_0 is the amount excreted which cannot be accounted for by the intake on these days. The a's are regression coefficients which indicate the influence of the variability in a corresponding day's intake on the variability of the excretion. In using the equation to predict the elemental output on any day, these coefficients are just fractions of the intake on the corresponding day.

This kind of treatment has been useful in estimating reexcretion into the gut of certain elements, and thus has made possible a better estimate of f_1. Regressions were run on fecal excretion over a four-day period and the intake over the same period, the previous four-day period, two periods previous, and so on through five periods previous, or covering a total of 24 days.

The equation in this case is

$$y_{4\ \mathrm{day}} = a_0 + a_{1-4}x_{1-4} + a_{5-8}x_{5-8} + a_{9-12}x_{9-12} + a_{13-16}x_{13-16} + a_{17-20}x_{17-20} + a_{21-24}x_{21-24} .$$

The elements for which the multiple correlation coefficient R was greater than 0.32 and for which the probability that this value could have occurred

Table 26.3. Regression Equations for Estimating Four-Day Fecal Output from Intake by Ingestion

September 6, 1966—May 13, 1967

	Subject C					Subject D				
	Cr	Mo	Ag	Sr	Sn	Cr	Mo	Ag	Sr	Sn
Regression coefficient										
a_{1-4}	0.026	0.16	0.039				0.035	0.038		0.094
a_{5-8}		0.12	0.021	0.21	0.26			0.066	0.16	
a_{9-12}	0.041						0.039			
a_{13-16}									0.15	
a_{17-20}				0.24	0.14	0.08				0.071
a_{21-24}									0.15	
a_0 (μg)	220	310	100	980	4400	240	330	290	1700	3800
R^2	0.09	0.20	0.25	0.12	0.18	0.48	0.19	0.12	0.14	0.12
Mean four-day output (μg)	280	450	140	5700	15,000	330	480	510	6300	15,000
Standard deviation	220	490	110	4000	17,000	400	550	450		17,000
Standard error of estimate	210	450	97	3800	15,000	300	500	430		16,000
Four-day intake level (μg)	940	1100	790	6500	22,000	1400	2400	1600	8600	38,000

by chance was less than 5% ($p < 0.05$) are included in Table 26.3. It is probably safe to say that correlation between fecal excretion and dietary intake within the previous week reflects the passage of all of the unabsorbed element through the G.I. tract. Some reexcretion into the gut may also take place during this time, but it is impossible to estimate the fraction from these data. Since the regression coefficients are considered to be fractions of the amount ingested, tin and chromium showed less than 10% of the amount ingested to be reexcreted within 24 days. The value 0.08 for a_{17-20} for chromium for subject D was very highly significant ($p << 0.0001$).

The most interesting results were those for strontium. If it is assumed that fecal excretion within one week involved only the unabsorbed element, it appears that 24 to 30% of the amount ingested was reexcreted into the gut within 24 days. When this is added to the amount excreted in urine, the total fraction absorbed of the amount ingested is about 0.34 for both subjects, which agrees very well with the ICRP-59 value of 0.3. The distribution of the excretion of absorbed quantity between urine and feces appears to be the reverse of that reported by Fujita[3] from an experiment on the ingestion of a single dose of ^{85}Sr by one man and by Bishop *et al.*[4] after intravenous ingestion of a single dose of ^{85}Sr into two subjects. That is, the reexcretion into the gut for subjects C and D was over twice as great as the urinary excretion rather than one-half as great. Actually, the situations are not comparable, since subjects C and D ingested strontium continually instead of in a single dose, and the value of the fraction of strontium in the urine of the total excreted was always closer to the value for Fujita's subject during the first two or three days than to the value which this fraction became after 90% of the dose had been excreted.

The results for calcium were not included in the table because the multiple correlation coefficient R for subject C was less than 0.32. Since the correlation for subject D was sufficient to be included ($R = 0.48$), it should be pointed out that for both subjects 11% of the amount ingested appeared to be reexcreted into the gut during the four-day period 17 to 20 days after ingestion, which is close to the value proposed by Dolphin

[3] M. Fujita *et al.*, *Health Phys.* 9, 407 (1963).

[4] M. Bishop *et al.*, "Excretion and Retention of Radioactive Strontium in Normal Men following a Single Intravenous Injection," *Intern. J. Radiation Biol.* 2, 125 (1960).

and Eve[5] as a representative value. This value added to the ~11% excreted in urine gives an f_1 of about 0.22, which is closer to the value of 0.38 proposed by Dolphin and Eve than to the ICRP value of 0.6. The discrepancy arises from the difference in urinary output. Subjects C and D excreted in the urine about 11% of the amount ingested, whereas Dolphin and Eve suggest a value nearer to 20%. The latter value is also closer to the average value for three subjects reported by Harrison *et al.*,[6] although the value for one of those subjects was 13%. It is quite reasonable to suppose that calcium (and strontium as well) has components of endogenous excretion with much longer time lags than the 24 days included in this study. As more data become available longer periods of time will be included in the regression studies.

METHOD FOR REMOVAL OF PHOSPHATE FROM BONE ASH

Cyrus Feldman F. S. Jones

The removal of phosphate from bone ash facilitates the spectrographic determination of impurities in this material by making it possible to chemically concentrate the trace elements at pH 6.5, rather than the usual pH 5.2. This substantially increases the number of trace elements which can be collected and eliminates the undesirable coprecipitation of calcium phosphate.

A practical procedure was settled on for the removal of phosphate from solutions of bone ash. After ignition at 450°C, the ash is dissolved in nitric acid, followed by formic acid. This solution is placed in the cathode compartment of a two-part electrodialysis cell; the compartments are separated by an Ionac Ma 3475 x L anion-permeable membrane. Electrodialysis at 2 amp for a few hours transfers the bulk of the phosphate to the anode compartment; cations are retained in the cathode compartment and/or deposited on the carbon cathode. The latter is wet-ashed and added to the sample solution.

Trace elements are then collected from this solution by precipitation with 8-hydroxyquinoline + thionalide + tannic acid. The precipitate is examined spectrographically.

[5]G. W. Dolphin and I. S. Eve, *Phys. Med. Biol.* 8, 193 (1963).

[6]G. E. Harrison, W. H. A. Raymond, and H. C. Tretheway, *Proc. Intern. Congr. Peaceful Uses At. Energy* 11, 156 (1956).

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