CESIUM-137 AND OTHER GAMMA RADIOACTIVITY IN THE FLORIDA ENVIRONMENT - A STUDY OF SELECTED MEDIA

By
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Chairman: Billy G. Dunavant, Ph.D.
Co-Chairman: H. A. Bevis, Ph.D.
Major Department: Bioenvironmental Engineering

A study was performed of the kinds, levels, and distribution of gamma-emitting radionuclides in the Florida environment. The investigation was initiated because of the unusual levels and characteristic geographical patterns of cesium-137 (^{137}Cs) found in Florida milk and forage in earlier studies. Beef and vegetables were selected as the most important media for sampling.

Sampling was carried out in May and June, 1966, and during January through July, 1967. Analyses were performed by gamma spectroscopy on triturated whole samples. Complex gamma spectra were interpreted in terms of the individual contributing components by use of the simultaneous equations method.

The most significant gamma-emitting radionuclide present was ^{137}Cs, and the evaluation of the data was concentrated on this nuclide.

Levels of ^{137}Cs in both beef and vegetables showed geographical patterns of variation similar to those reported earlier by others in Florida milk. Maximum levels were found in the central and southern parts of the State, with intermediate levels in the northeastern and

north central parts of the State, and the lowest levels in the northwestern part of the State. In addition, vegetable samples showed a marked difference from southeast to southwest, with average levels in the southeastern part of the State as low as in the northwestern part.

The levels of ^{137}Cs in lower-quality meat from animals that had fed primarily on grass were much higher than those in high-quality meat from feed-lot animals. These levels were higher than any other levels reported in beef in the conterminous United States.

There was no apparent difference in ^{137}Cs concentrations between leafy, fruit, and root categories of vegetables; although certain crops in each category did exhibit consistently higher levels than the other crops in the same category.

Cesium-137 concentrations found in this study were compared to literature values, both to those reported for years previous to this study and to 1966-1967 values obtained by extrapolating published values to that time. Cesium-137 concentrations in both beef and vegetables in much of the State were considerably higher than the estimated national averages for the same period; average concentrations in the regions exhibiting the lowest average levels were found to be comparable to the estimated national averages.

The significance of the observed ^{137}Cs levels was evaluated in terms of the human intake of this nuclide. It was estimated that radioactivity intakes from locally produced food in Northwestern Florida would be similar to the national average. Three hypothetical cases of assumed diet composition and source of food were set up to evaluate levels in the remainder of the State. Consideration of diets with various combinations of Florida milk, beef, and vegetables having the average levels shown in much of the State resulted in predicted ^{137}Cs intakes and body burdens ranging from two to 20 times the projected national average. The intakes, body burdens, and resulting whole-body radiation doses estimated for the various cases considered were all less than the applicable radiation protection guides, but the guides could be approached by individuals constantly consuming food at the extreme levels found.

It was concluded that unusual environmental factors or mechanisms are involved in the levels of ^{137}Cs found in the Florida environment. The possibility of such mechanisms has important implications to waste disposal, hazard evaluation, and nuclear facility operation. The mechanisms were not identified but there is increasing evidence that the role of uptake from the soil is greater than that reported for most areas of the country.

CHAPTER I

INTRODUCTION

This research involved a study of the kinds, levels, and distribution of gamma-emitting radionuclides in selected segments of the Florida environment with emphasis placed on the nuclide cesium-137 (^{137}Cs) in animal and vegetable products.

This study was initiated because of unusual levels and characteristic geographical patterns of ^{137}Cs found in Florida milk and forage in earlier studies described in Chapter II. Milk monitoring programs of the United States Public Health Service (USPHS) and of the Florida State Board of Health have indicated that the average levels of ^{137}Cs in milk in much of Florida have been higher than the national average and among the highest in the nation for a number of years, while strontium-90 (^{90}Sr) levels have been consistently average or below. Further investigations have shown a consistent geographical pattern within the State, and it appears that these levels are primarily related to the intake of ^{137}Cs by cows through the medium of locally grown forage.

The major effort in this study was directed toward determining the extent to which these unusual patterns of ^{137}Cs are reflected in other elements of the human food chain, particularly lean beef and truck vegetables.

In addition, measurements were made of other gamma-emitting radionuclides in all samples.

Significance of this Research

First, it is important to the welfare of the people of Florida that the extent to which persons in the State are exposed to radiation from all sources and by all routes be known.

This study contributes to this knowledge by (1) investigating the extent to which the elevated levels of ^{137}Cs previously observed in milk and forage extend to other media, particularly other elements of man's food chain, (2) determining whether other gamma-emitting radionuclides show elevated levels in Florida, and (3) evaluating the potential influence of these nuclides and their levels on man's radiation exposure.

In Chapter II, it is pointed out that many routes of radiation exposure have been and are being evaluated in Florida. Gross alpha and beta radioactivity levels are measured in a variety of media in this state. Detailed studies have been performed in selected localities, and exposure to specific nuclides through the route of milk is being studied in great detail. However, this work represents the first extensive, statewide study of exposure to specific nuclides through food chain elements other than milk.

Second, the information developed here to estimate exposure levels should also contribute to the understanding of the mechanisms involved in the previously mentioned unusual levels of ^{137}Cs in Florida.

It also is important to predict and evaluate the impact on the environment--and, eventually, on human health--of nuclear activities such as the operations of nuclear power plants, nuclear laboratories, and nuclear powered vehicles. A thorough, systematic study of radioactivity in the State provides a baseline of radioactivity levels against which any future increases can be compared. This means determining not only average levels of various radionuclides for the purposes of comparison but also normal variations of these levels for use both in designing future surveys and in determining whether apparent increases are significant. In addition, knowledge of the mechanisms and rates of transfer of radionuclides through the environment provides a basis for predicting the consequences of accidental releases of radioactivity. At the same time, since intentional waste release is governed by criteria based on various assumptions concerning the normal behavior of radionuclides in the environment, it is important to be aware of any unusual behavior such as suggested by the Florida milk ^{137}Cs levels.

Finally, this study is significant in that the data collected will contribute to other environmental radioactivity study efforts in the State. For example, these data can be used to complement the study of ^{137}Cs in Florida milk being carried out by the Florida State Board of Health and to serve as an extension to the environmental monitoring being carried out in the Cape Kennedy off-site socio-economic impact area by the United States Air Force, the USPHS, and the Florida State Board of Health.

CHAPTER II

REVIEW OF THE LITERATURE

This study was undertaken as a direct result of the findings of earlier studies of radioactivity in Florida milk and dairy feeds carried out by this author and others. The USPHS began a program of sampling raw milk for radioactivity analysis in 1957, and out of these initial efforts grew the present USPHS Pasteurized Milk Network which has 63 stations, at least one in every state of the Union, the Canal Zone, and Puerto Rico.[1] A station representing Florida became operative at Tampa during August, 1960. In this network, a composite sample is taken from the pasteurized milk marketed in the community and shipped to a USPHS laboratory for radionuclide analysis.

Average monthly ^{137}Cs and ^{90}Sr levels at the Florida station of the network are compared against the corresponding national ranges and averages for the period 1961 through February, 1967, in Figure 1. Consistently, the average levels of ^{137}Cs at the Florida station of this network have been well above the national average; since mid-1964, they have been the highest in the sampling network. In contrast, ^{90}Sr levels have not been particularly high; with the exception of two months, they were consistently below the national average. As a result, the $^{137}Cs/^{90}Sr$ ratios have been particularly high.

Workers at the Florida State Board of Health initiated a program in 1961 for the statewide collection and regional compositing of raw milk from a 10 per cent random sample of the dairy farms in the state.[2-7]

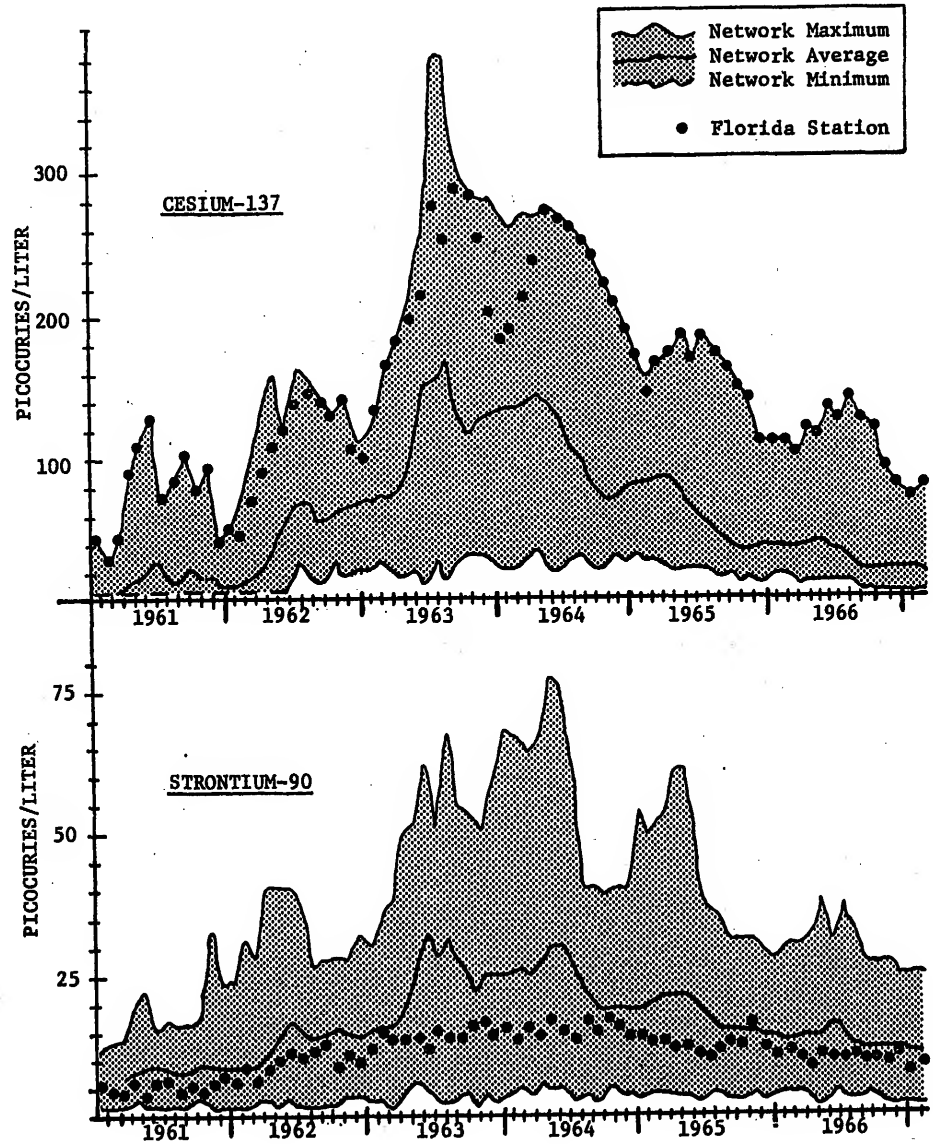


FIGURE 1. CESIUM-137 AND STRONTIUM-90 IN PASTEURIZED MILK--COMPARISON OF THE FLORIDA STATION TO NATIONAL AVERAGES AND RANGES[1]

Figure 2 summarizes ^{137}Cs and ^{90}Sr results for 1963 through 1966. It shows that the average results reported for pasteurized milk samples from the Tampa station of the national network are comparable to the average results for composite raw samples collected from the same general region. The figure also shows that the Tampa station results for both nuclides are reasonably well representative of the reported average of all milk produced in Florida, particularly the area east and south of Tallahassee. There does appear to be a gradual transition from lower ^{137}Cs levels in the northwestern part of the State to higher levels in the eastern, central, and southern parts of the State with the highest in the central part of the State.

Figure 2 also shows that ^{137}Cs levels in the northwestern part of the State are below the State average. These levels fall more rapidly with time than levels in the remainder of the State and fall from above the national network average in 1963 through 1965 to below it in 1966. Statewide average ^{90}Sr results are consistently below the national network average and remain fairly constant for the eastern, central, and southern parts of the State. These results show a greater fluctuation in the Northwest--even rising above the national network average in 1965 and 1966.

Porter _et_ _al_., in a special study of the Tampa milkshed during October, 1963, through February, 1964, observed considerable farm-to-farm variation within that area and concluded that the elevated milk levels were due to elevated levels of ^{137}Cs intake by the animals and not to unusual transfer of the nuclide from the feed to the milk.[8] They also identified pangolagrass (_Digitaria_ _decumbens_) as the principal contributor to the high ^{137}Cs intake.

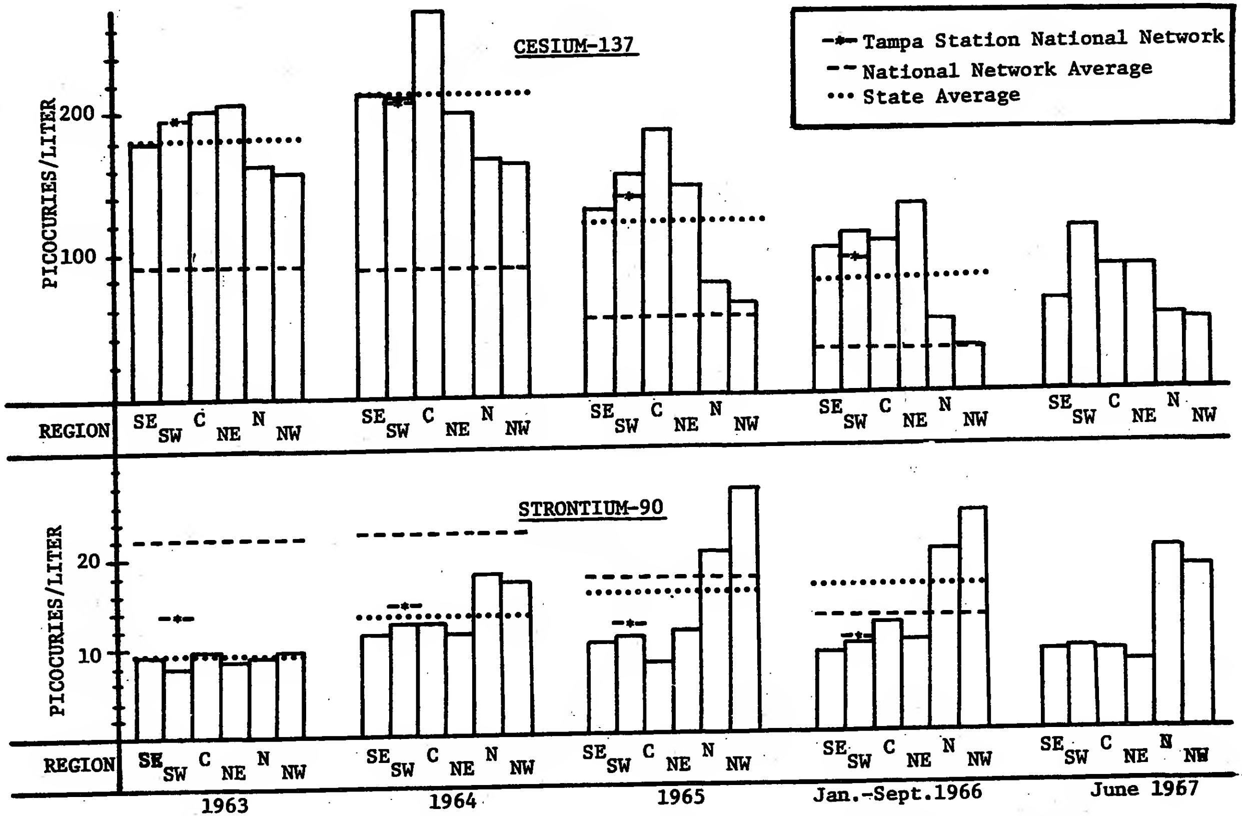


FIGURE 2. RADIONUCLIDES IN FLORIDA MILK--YEARLY AVERAGES BY REGION

In 1964, Roessler and Williams initiated a study to investigate more fully the factors influencing radionuclide levels in Florida milk.[9] Unpublished reports of their work also show a high farm-to-farm variability in milk radionuclide levels. These results suggest that when examining more farms on a wider geographic basis, equally high or higher intakes can be attributed to forages other than pangolagrass.[7]

Reviews of Environmental Radioactivity

The interest in environmental radioactivity since the early 1950's is evidenced by the large number of bibliographies, hearings, symposia, conferences, and books devoted to the subject.

Hoard, Eisenbud, and Harley prepared an extensive bibliography on radioactive fallout in 1956.[10] Because of growing public concern at this time about the possible effects of fallout from nuclear weapons testing, the Joint Committee on Atomic Energy of the Congress of the United States held hearings on radioactive fallout and its effects on man in 1957, and again in 1959.[11,12] The scientific community explored the question of radioisotopes in the biosphere in a symposium in 1959 and again in the First National Symposium on Radioecology in 1961.[13,14] The United States Atomic Energy Commission (USAEC) sponsored a conference on radioactive fallout from nuclear weapons tests in 1961 and again in 1964.[15,16]

Eisenbud authored a text on environmental radioactivity in 1963, and a collection of papers reviewing the ecology of radioactive fallout, soils, plants, foods, and man, edited by Fowler, was published in 1965.[17,18]

Natural environmental radioactivity was the subject of a symposium in 1964 and a 2,000 reference bibliography by Klement in 1965.[19,20]

The proceedings of a symposium on radiation and terrestrial ecosystems in 1965, edited by Hungate, review radioactive fallout phenomena and mechanisms, primordial and cosmic-ray produced radionuclides, soil-plant relationship, and radionuclide cycling as well as present results of individual research.[21] The Second National Symposium on Radioecology was held in 1967, at which time emphasis was placed on reporting individual investigations of both effects of radiation on the environment and behavior of radionuclides in the environment.[22]

Comar reviewed the literature up to January, 1965, relating to the movement of fallout radionuclides through the biosphere and man; Ellis reviewed and appraised the levels and biological significance of current fallout levels in the United Kingdom in 1965.[23,24] The fallout pattern in the Nordic countries was a topic at a proceedings of the First Nordic Radiation Protection Conference in Sweden in 1966.[25] About the same time Leistner, on behalf of Euratom, reviewed the reported research on the radioactive contamination of foods of animal origin, tabulating reported distributions, retention, and ambient levels.[26]

A book edited by Russell in 1966 is devoted entirely to radioactivity and human diet.[27]

In August, 1959, the President of the United States directed the Secretary of Health, Education, and Welfare to collate, analyze, and interpret data on environmental radiation levels. This assignment resulted in the appearance in April, 1960, of the publication, <u>Radiological Health Data</u> (subsequently changed to <u>Radiological Health Data and Reports</u>). This monthly publication contains regular reports of routine national and worldwide monitoring programs operated by agencies of the United States as well as frequent special reports of state monitoring programs, special studies, special evaluation of particular problems, and topical reviews of current research.[28]

Sources of Environmental Radioactivity

According to the best present knowledge, man has always been exposed to varying degrees of natural background radiation from cosmic radiation and from naturally occurring radionuclides.

Eisenbud points out that the world inventory of radioactive materials prior to World War II, both in the environment and in the laboratory, was confined to those which occurred in nature, with the exception of a relatively few millicuries of radioactivity produced in cyclotrons during the late 1930's.[17] Construction of large nuclear reactors during the war and the associated operations for extracting plutonium from irradiated uranium resulted in the first extensive occasions for contaminating the environment with radioactive substances. Then, in the late 1940's and continuing at an accelerated rate throughout the 1950's, there began a series of nuclear weapons tests that resulted in a worldwide distribution of radioactive materials in all segments of the environment--the atmosphere, the soil, food chains, and man himself.

Eisenbud lists the principal sources of environmental radioactivity as natural radioactivity, preparation of nuclear fuel through the stages of mining, concentrating, milling and fabrication, reactor operations and accidents, conventional radioisotope use, aerospace applications of radioisotopes and reactors, fuel reprocessing and radioactive waste disposal.[17] Comar identifies the major source to date as radioactive debris from the testing of nuclear weapons.[23] On the other hand, important future sources will likely be nuclear reactor operation, radioisotope use, and radioactive waste disposal.

Williams *et al*. pointed out that varying levels of natural radioactivity do occur in Florida and that these materials do have an opportunity to become relocated in the environment through the operations of phosphate mining, phosphate fertilizer production, and subsequent use of the by-products.[29] With regard to the other sources of environmental radioactivity just mentioned, there is no uranium mining, concentrating, or milling, nor fuel fabricating or reprocessing, nor large scale or commercial radioactive waste disposal in the State. Reactors are presently limited to the University of Florida Training Reactor, and there have been no reported contaminating incidents in the State.

The Pinellas Peninsular Plant of the USAEC is located near St. Petersburg, Florida, but from the regular published reports of the contractor's environmental monitoring in the vicinity, it may be inferred that radionuclides potentially reaching the environment from this plant are limited to tritium.[30] This means that, with this exception, at the present time any widespread radioactivity in the Florida environment must be naturally occurring radioactivity or must come from fallout injected into the atmosphere. At the same time, the environmental mechanisms affecting activity from these sources will affect in much the same manner any future radioactivity from local contamination sources.

With regard to the future, several nuclear power plants are definitely planned for the State of Florida and it is likely that radioisotope or reactor powered electrical generators or propulsion devices will be used in future vehicles launched from the missile testing and space flight complex at Cape Kennedy.[31]

<u>Ecology of Cesium-137 and Other Environmental Radioactivity</u>

It was pointed out in the preceding section that, with certain local exceptions, the major source to date of environmental radioactivity is radioactive fallout from the nuclear weapons testing. Cesium-137 and

^{90}Sr are considered biologically to be the most important radionuclides in long-range fallout because of their long physical half-lives, the high yield of these nuclides in fission, and their respective chemical similarities to the natural body constituents, potassium and calcium.[23,32] Indeed, the majority of studies of special nuclides in the environment appear to be concerned with these two nuclides.

The remaining examination of the literature emphasizes ^{137}Cs, but gives attention to other nuclides to the extent that they might also appear in samples examined for ^{137}Cs.

In 1959, Langham and Anderson reviewed biospheric contamination from ^{137}Cs. They estimated production by weapons tests and deposition up to that time, reported important exposure routes to man, and listed an extensive bibliography.[33] They proposed expression of ^{137}Cs levels as ^{137}Cs per gram of potassium because of the metabolic similarity of cesium and potassium. A later exhaustive review of cesium ecology was made by Davis in 1963.[34]

In 1965, Comar summarized and updated this information in a concise, yet comprehensive, review of the movement of fallout nuclides through the biosphere.[23] He observes that cesium and potassium are chemically similar. However, since they are not metabolically interdependent, the ^{137}Cs/potassium ratio is not as meaningful as the ^{90}Sr/calcium ratio. Nevertheless, ^{137}Cs concentrations are often expressed relative to potassium for two reasons: (1) naturally radioactive potassium-40 (^{40}K) and ^{137}Cs are often measured simultaneously by gamma spectrometry, with ^{40}K thereby serving as an internal standard, and (2) in man, the cesium/potassium ratio correlates better with lean body mass and hence with radiation dose than does the ^{137}Cs per kilogram (kg) of body weight.

The worldwide deposition of ^{137}Cs is usually estimated by applying a $^{137}Cs/^{90}Sr$ ratio to the values of deposition of ^{90}Sr since it has been studied more extensively. The $^{137}Cs/^{90}Sr$ ratios vary between 1.0 and 3.0 as these nuclides are produced, but this ratio will change markedly as the two elements pass through metabolic systems.

Radionuclide pathways to man

Fallout radionuclides reach man primarily through plants, either by his consumption of foods of plant origin or indirectly by his consumption of animal products.[33]

Plants become contaminated by radioactive materials from the atmosphere by either (1) indirect contamination which occurs when radioactive materials enter the soil and passes into the plant through the roots as do soil nutrients or (2) direct contamination in which the material is deposited on some portion of the plant and passage through the soil is bypassed.[23] Direct contamination reflects events of the recent past, varies with the rate of deposition, and is designated "rate dependent." Indirect contamination is governed by the total amount available in the soil and is designated "cumulative dependent." Indirect contamination has the characteristics that (1) material deposited before the plant develops can still enter the plant, (2) short-lived radionuclides that enter the soil will have a high probability of decaying to insignificant levels before reaching the plant roots, and (3) radioactive substances entering the soil may be diluted with soil substances, rendered unavailable to plants by fixation to soil materials, or discriminated against in plant uptake.

The concepts of cumulative and rate dependent contributions to radionuclide levels in an environmental component are illustrated in the generalized expression:

$$C = p_d F_d + p_r F_r$$

where C = concentration of the radionuclide in the sample on interest,

F_d = cumulative deposition,

F_r = current rate of deposition,

and p_d and p_r are the proportionality factors for cumulative deposition and deposition rate, respectively, for the particular system and nuclide under consideration.[23,35,36]

Once radionuclides are deposited on or incorporated into plants, the grazing animal effectively collects contamination from plant material and concentrates it in animal products; however various factors such as metabolic behavior of the specific nuclide and animal feeding and management practices influence the relationship between the amount deposited in tissues or transferred to secretions that are used for food.[23]

In order to select important media for further study and to consider possible movements of radionuclides in the Florida environment and possible geographic, media, and temporal variations, it is necessary to review briefly what is known about the passage of radionuclides, particularly ^{137}Cs, through various segments of the environment.

<u>Origin, transport, and deposition of radioactive fallout</u>

Klement and Langham both reviewed fallout phenomena and mechanisms in 1965.[36,32] Klement identified the sources of injection of radioactivity into the atmosphere as nuclear weapons tests, peaceful nuclear explosives tests or utilization, reactor operations, and space applications of nuclear energy. He placed weapons tests foremost from the standpoint of widespread environmental radioactivity. The radioactivity can include fission products (over 200 different nuclides of about 36 elements between atomic numbers 28 and 65) and activation products that come about as a result of neutron interaction with soil, air, water, or parts of the nuclear device at the time of the detonation. Radioactive debris is usually apportioned into three fractions: (1) fallout in the immediate vicinity, (2) material injected into the troposphere, and (3) material injected into the stratosphere.

Local fallout is not a concern in this Florida study, but tropospheric fallout can deposit short-lived nuclides in the weeks immediately following atmospheric weapons tests. Deposition from tropospheric contamination is governed by tropospheric air flow and should be highest at the latitude of injection with a certain amount of dispersion and displacement and with local low altitude variations due to terrain such as mountains and land-water contrasts.[17,36] One exception is the downward and equatorward movement along isoentropic surfaces of debris injected into the upper polar troposphere. A second exception is the poleward movement and sinking in subtropical regions of mid- and upper-tropospheric injections in equatorial regions.

Material injected into the stratosphere may remain there from several months to several years and provides the source of long-lived materials currently being deposited on a worldwide basis. Stratospheric materials are transferred to the troposphere, from which deposition is primarily due to precipitation with a small increment due to dry deposition.[36]

List _et al_. described the generalized worldwide deposition of ^{90}Sr up to late 1963.[37] The mean latitudinal distribution in the Northern Hemisphere showed a very low deposition in the vicinity of the north polar region, a rapid increase to a maximum between 40° and 50° N, and then a gradual decrease in the vicinity of the equator. Davis prepared a similar latitudinal deposition curve for average ^{137}Cs content of precipitation for 1955-1958.[34] Florida lies between 25° and 31° N, a location whose deposition on the mean distribution curve lies midway between the Northern Hemisphere peak and the equatorial minimum. Within latitude bands, differences in rainfall and possibly other meterological phenomena cause variations in deposition. It has been suggested that fallout over oceans might be higher than over land but List et al. report that comparison of soil from sites with maritime exposure to soil with sites from continental exposure shows no systematic difference either in total deposition or in amount of ^{90}Sr deposited per inch of rainfall.[37]

Two major techniques have been employed for the direct quantitative assessment of fallout deposition on the earth's surface: (1) deposition sampling (dry and precipitation) and (2) soil sampling. Based on soil sampling in 1963-1964, maps of ^{90}Sr deposition were prepared for the world and for the United States. The latter shows Florida lying between the 100-millicurie per square mile contour at the southern tip of the State and the 125-millicurie per square mile contour along the northern border.[37]

Stratospheric deposition is usually described by a simple exponential function; however, Machta has described shortcomings of such a simplified model.[38] Comar quotes an overall mean stratospheric residence time of two years, while data presented by Klement imply a value of one year.[23,36]

As an aid to predicting general fallout levels, the Federal Radiation Council divides the United States into "wet" and "dry" areas depending upon average annual rainfall (roughly separated by the 20-inch rainfall isoline).[39,40] It further denotes an intermediate area with slightly less than 20 inches of annual rainfall and, by superimposing the latitudinal effect on the precipitation effect, assigns most of the State of Florida to another immediate area with "an expected lesser fallout compared with the 'wet' eastern United States because of its subtropical location."

Estimates of ^{90}Sr deposition in the United States through 1966, derived from several sources, are shown in Table 1. Values for years beyond 1966 can be obtained by an extrapolation based on the assumptions that deposition is described by a single exponential function and that there will be no significant additions to the stratospheric content.

TABLE 1

STRONTIUM-90 DEPOSITION IN THE UNITED STATES
(MILLICURIES/MILE^2)

	Increment		Accumulated	
	Wet Area	Dry Area	Wet Area	Dry Area
Based on Federal Radiation Council Report No. 6:[40]				
1963	-	-	150	65
1964	30	12	180	77
1965	15	6	195	83
Total after 1965	25	10	220	93
1966 estimate[36]	7.5	3	203	86

Although air is the medium by which fallout is delivered to terrestrial environment, Klement emphasized that it is difficult to obtain explainable relationships between concentrations in surface air and other environmental media such as precipitation, soil, and biological systems.[36] Some qualitative information is being developed; for example, Pelletier, Whipple, and Wedlick have attempted empirically to fit measurements in the Lake Erie area with a model of the form:

$$\frac{D}{C_A} = H(a-e^{-br})$$

where D = deposition, picocuries per square meter, (pCi/m^2),

C_A = air concentration, pCi/m^3,

H = height of fall constant, m,

a = dimensionless factor to account for dry deposition into the precipitation collector,

b = elimination constant, (inches of rain)$^{-1}$, and

r = rainfall amount, inches.[41]

Further study is necessary before the various relationships can be clearly defined.[36] Air concentrations and deposition rates show a peak during the spring of each year which is related to entrance of debris from the stratosphere during late winter, spring, or early summer. After new tropospheric injections, peaks in concentrations are seen along the path of the cloud, and air measurements are particularly useful to determine arrival time of new debris at specific locations and thus anticipate the rise in levels in other media. Atmospheric contamination of fallout ^{137}Cs is not considered an inhalation hazard.[33]

Radionuclides in water

In his 1965 review, Klement also summarized some of the observations of fallout radionuclides in water reported up to that time.[36] He stated that short-term variations in deposition are probably not as detectable in fresh surface waters as they are in surface air, but that the same seasonal variations are seen. Relative concentrations of nuclides in surface waters vary with local deposition rates and characteristics of the local aquatic environment. Large differences can be expected within a major river or lake because of runoff and flow.

Langham and Anderson concluded that drinking water contamination with fallout ^{137}Cs does not constitute a health hazard.[33] However, water may have an importance in the food chain in that ^{137}Cs is more available to organisms living in aquatic environments than in terrestrial environments.[42]

Cesium-137 and other radionuclides in the soil

Schulz reviewed soil chemistry in 1965.[43] He listed fission products likely to be found in the soil, classified elements according to their mobility in the soil, and discussed the use of soil chemistry in predicting plant uptake of radionuclides.

Cumulative levels of radionuclides in the soil and their distribution in depth, important factors in plant uptake from the soil, are affected by weathering and other disturbing influences.[36] In most soils in the United States, ^{90}Sr does not appear to leach very rapidly (supporting the validity of soil sampling as a means of determining accumulated fallout deposition). Virtually all of the nuclide is found in the upper 8 inches of these soils. In medium to fine textured soils, about 75 to 80 per cent of the deposited ^{90}Sr is found in the upper 2 inches and even in medium textured soils similar proportions are found in the upper 4 inches.

A number of authors have cited the fact that cesium is readily retained by soils.[34,36,43,44] Its sorption is characterized by high adsorption onto mineral particles (the ion exchange properties of soil being largely associated with clay minerals in the soil complex), but there is a certain amount of disagreement over the mechanism of this fixation. Nearly all of the ^{137}Cs is found near the surface (the top 2.5 centimeters) of undisturbed soils in the United States.[34,36]

Downward movement of cesium in undisturbed soils is even less than strontium. After five years, 50 to 80 per cent of added ^{137}Cs remained in the upper 5 centimeters with little penetration below 10 centimeters.[23] It is pertinent to the Florida soil scene that, in batch experiments, sandy loams and clays absorbed 90 per cent of the available cesium, while sand absorbed only 50 per cent.[34]

Ranges of pH commonly encountered have little effect on cesium sorption since it is approximately constant above a value of 3.5 (though decreasing rapidly below pH 3.5).[34]

While cesium is desorbed by cations of neutral salts, more readily by K^{+} than by Na^{+} or Ca^{++}, it effectively competes with potassium in interactions in the soil.[34] Trace amounts of cesium adsorbed by the soil are held more strongly than strontium. Russell states that the extent of ^{137}Cs fixation in soil depends upon the clay content and is inversely related to the organic matter in which cesium remains less strongly bound.[35] Schulz states the chelation of nuclides by soil organic matter may cause them to keep some mobility in the soil.[43]

Complementary ions have a strong influence on the redistribution of nuclides in the soil profile.[43] For example, it is expected that the amount of soluble calcium will have a direct effect on the mobility of Sr^{++} and Ra^{++} in soils and consequently on the ease of uptake by plants.

Alkali cations present a more complicated case.[43] With regard to ^{137}Cs, complementary ions can be classed into two groups: (1) those having little effect on cesium, such as dilute H^{+}, the alkali cation Na^{+}, and the alkaline earth Ca^{++}, Mg^{++}, and Ba^{++}, which solubilize or release less than 10 per cent of added carrier-free ^{137}Cs and (2) those liberating moderate amounts of bound cesium, such as Cs^{+}, NH_4^{+}, K^{+}, and Rb^{+}, which free 20 to 80 per cent of the cesium to a labile form. Typical agricultural soil in the neutral pH range possesses primarily Ca^{++} and Mg^{++} as labile ions. These have little effect in releasing added ^{137}Cs for plant uptake.

<u>Movement of cesium-137 and other radionuclides from the soil to plants</u>

A number of experiments in nutrient solution indicate that cations apparently compete against cesium in uptake by plant roots.[34] In one experiment at low external cesium concentrations, potassium, rubidium, ammonia, and cesium were markedly effective in depressing ^{137}Cs uptake by barley roots, while sodium, lithium, calcium, and magnesium were not; at high concentrations, all inhibited ^{137}Cs absorption. Millet, oats, buckwheat, sweet clover, and sunflower discriminated slightly against rubidium and strongly against cesium in favor of potassium. In another experiment, an increase in the potassium of the solution decreased ^{137}Cs uptake by bean plants but resulted in an increased potassium content of the plants. At low potassium levels, barley showed high selectivity for potassium relative to cesium; however at high concentrations, uptake was less selective in favor of potassium. On the other hand, other investigators reported that adding potassium had only a small effect--a 100-fold increase in potassium reduced ^{137}Cs uptake by only a factor of two and non-radioactive cesium enhanced ^{137}Cs uptake.[45]

Uptake by plants from soil is influenced by both the availability of the element in the soil and the inherent behavior of the plant with regard to available elements. According to Menzel, plant concentrations of various radioactive elements, after they have been added to the soil in water-soluble forms, may be several orders of magnitude higher or lower than the concentration in the soil.[44] He classifies elements into five classes of uptake, ranging from strongly concentrated to strongly excluded by assuming that the behavior of radioactive elements in the soil-plant system is identical with that of stable nuclides of the same element. In general, those nuclides that are most readily absorbed are soluble in the soil or are isotopic with elements that have metabolic functions in the plant. Conversely, those that are least absorbed are quite insoluble in the soil. Notable examples are strontium, slightly concentrated; radium, not concentrated; and cesium, slightly excluded in uptake from the soil.

Removal of ^{137}Cs from the soil is inhibited by the strong fixation in the soil but uptake varies with soil type.[23,34,35]

In typical soils of the temperate regions, ^{137}Cs was taken up about one-tenth as much as ^{90}Sr, with this ratio decreasing with time to about one twenty-fifth or less after about three years.[23] This suggests more than one cesium storage compartment in the soil. Less strong binding in organic matter has already been mentioned, and observations suggest that a high level of organic matter in the upper layer of some permanent pastures might reduce binding of cesium by clays, thereby enhancing and extending its availability to plant roots to one or two years.[23,35] In certain tropical areas ^{137}Cs was found to be more available to plants than had been expected.[23]

Schulz reasons that complementary ions, in strongly affecting mobility of ions in the soil as outlined in the preceding section, will in turn influence uptake by plants.[43] In modern agriculture, nitrogen, potassium, and phosphorus are commonly added to soils. From the complementary ion effect, it would be expected that addition in the forms K^+ and NH_4^+ would liberate considerable amounts of ^{137}Cs into a labile form for subsequent plant uptake, while the application of nitrogen in the NO_3^- form would have little effect on the fixed ^{137}Cs. In an experiment by Schulz, both K^+ and NH_4^+ increased the ^{137}Cs uptake by romaine lettuce but the NH_4^+ had a much greater effect than K^+. This was explained by the hypothesis that after the NH_4^+ had released fixed ^{137}Cs, microbial oxidation of the excess NH_4^+ to NO_3^- removed it from competition with the now labile ^{137}Cs for plant uptake.

Davis agrees that uptake of ^{137}Cs from the soil is increased by the addition of cesium and cites this as evidence that the capacity to fix cesium in the unavailable form is actually very small.[34] He cites a number of experiments, however, where the uptake of ^{137}Cs by lettuce, grass, alfalfa, corn, millet, and wild plants was decreased by the addition of potassium or was inversely related to the exchangeable level present. In another experiment, added potassium resulted in a decrease in the ^{137}Cs uptake by Ladino clover and, as a corollary, the uptake increased as soil potassium was reduced by continued cropping.[45] In this latter experiment, stable cesium added to the soil, even to toxic levels, caused increased ^{137}Cs uptake in this crop.

The collected evidence from the studies cited in this section seems to indicate that complementary ions play a role in releasing fixed cesium from the soil, and that stable cesium added to the soil enhances the overall uptake of ^{137}Cs by plants, but that the effect of stable cesium on plant uptake from available ^{137}Cs and the effect of potassium on the uptake from the soil are not clearly understood.

The effect of soil moisture content is apparently not clear. In studies with beans in soil, limited soil moisture resulted in greater ^{137}Cs uptake than with ample moisture.[45] On the other hand, Pendleton speculated in 1962 and again in 1967 that because of the high availability of the nuclide to aquatic and emergent plants, milk and meat from grazing animals utilizing forage from wetlands may contain much more of the nuclide from fallout than such products from drylands, and that the geographic variations observed in ^{137}Cs content of milk may be, in part, a result of different levels of uptake by plants of wet and dry lands.[42,46]

Behavior of cesium-137 and other radionuclides in plants

Cesium and potassium follow similar paths in physiological processes but are not used in an equally competitive manner by plants.[34] Translocation of cesium is more rapid than that of other fission products. Tissue distribution of this element varies with species. When available during active vegetative growth, it goes to the leaves and flowers with smaller amounts going to the seeds. Accumulation of ^{137}Cs by plants under natural conditions varies considerably among species and with environmental conditions.

In 1959, Langham and Anderson speculated that ^{137}Cs was entering the biosphere via direct contamination of the vegetation rather than via soil contamination and plant uptake.[33] It was soon determined that trace amounts of cesium can be readily assimilated by direct foliar deposition.[34] Russell cites,as well documented, and other authors agree, that direct contamination of plants rather than absorption from the soil is the dominant uptake process for major nuclides, particularly during periods of high fallout.[35,23,34] Evidence for this includes the fact that vegetation ^{137}Cs levels follow seasonal variations rather than being accumulative, with amounts in plants being comparable to deposition during the period of foliage existence.[34] This observation is also consistent with a large discrimination against soil to plant transfer.

Comar refers to foliar absorption of ^{90}Sr.[23] Russell states that it is necessary to consider both direct foliar retention and "plant base uptake."[35] He cites evidence that much of this nuclide found on edible leaves is actually absorbed at the basal region in the mat of prostrate stems and surface roots and then transported upwards. In 1964 and 1965, Wykes observed a reservoir of radioactivity in the vicinity of the plant base in Upper Midwest grassland when he observed more ^{137}Cs and ^{90}Sr per unit mass in the root-mat fraction than in the soil or grass.[47]

According to Russell, the amount of foliar deposition is comparable for ^{137}Cs and ^{90}Sr, but absorption from the soil is responsible for a smaller portion of the ^{137}Cs reaching edible tissues than for ^{90}Sr.[35] He also states that in times of relatively low fallout, the ratio of ^{137}Cs to ^{90}Sr in herbage and milk has decreased. This is evidence for a greater rate dependence relative to the cumulative dependence for cesium than for strontium.

Comar suggests that since milk concentrations of ^{137}Cs (related to levels in pasture grass) have not followed the fallout rate in some areas, it must be assumed that there are also mechanisms for accumulation of previously deposited ^{137}Cs.[23] This would be consistent with the earlier mentioned less tightly binding compartment in the organic matter in the soil. Davis predicts that when the stratospheric load of ^{137}Cs becomes nearly depleted, soil will then become the primary route of uptake.[34]

Direct floral contamination is reported as a dominant process for ^{90}Sr absorption in cereals but is negligible with storage organs which grow beneath the soil since there is little downward translocation of strontium and other divalent cations in plants.[35] Comar reviewed reports that ^{90}Sr accumulates in cabbages mainly by the lodging of rain in cavities at leaf axils and in potato tubers by the downward leaching by rain over the stem surfaces. The latter represents a means by which underground tissues can become contaminated from deposition.[23] Neither author makes reference to mechanisms of ^{137}Cs accumulation in these crops.

Johnson and Ward reported the ^{137}Cs concentration in grain to be much lower than that in hay.[48] Elder and Moore and also Porter et al. reported that pangolagrass (Digitaria decumbens) and Spanish moss (Tillandsia usneoides) had ^{137}Cs concentrations much higher than any other plants in the Tampa, Florida, area.[49,8] They concluded that the nuclide was surface contamination or weakly bound on the grass but was actively bound on the moss.

A number of authors refer to the use of the equation:

$$C = p_d F_d + p_r F_r$$

with regard to radionuclide levels in plants. Klement cautions that it is an empirical formula and does not necessarily imply understanding of the mechanisms of transfer into and within a system.[36] He makes note of the fact that p_d approaches zero for some plants such as Spanish moss and some lichens and that p_r is near zero for plants sheltered from appreciable deposition on foliage. A modified form has been presented for ^{137}Cs in which F_{2c} and p_{2c} are substituted for F_d and p_d, respectively, thus relating concentration to deposition accumulated over the two previous years rather than to the entire history of accumulated deposition.[23] This form presumably relates to ^{137}Cs released by decay of organic material and to the less tightly bound deposition in the organic layer and considers as negligible any uptake of the nuclide tightly bound by the mineral fraction of the soil. Russell also cautions about the empirical nature of the expression and emphasizes that aside from the large errors normally inseparable from field investigation, deduced relations relate only to climatic conditions and patterns of deposition which prevailed during the surveys or field experiments from which the proportionality factors were derived.[35]

Cesium-137 and other radionuclides in animals

Pendleton et al. have briefly summarized literature relating to cesium and potassium metabolism by animals.[50] Absorption of both cesium and potassium by mammals is large and takes place through the digestive tract. These elements enter the body cells where cesium is more tenaciously retained; excretion from the body is urinary and in varying degrees, depending on species and diet, fecal. Increasing potassium intake has only slight effect in decreasing body cesium content.

According to Davis, the discrimation factor from foodstuffs to animals (or man) should favor cesium due to preferential assimilation of trace amounts of cesium relative to potassium.[34] Pendleton et al., citing a number of studies and authors, report that in many animal species the ^{137}Cs/potassium ratio in the body is about two to three times higher than the ^{137}Cs/potassium ratio in their normal diets (unlike the values observed for the ^{90}Sr/calcium ratio in animals which are less than one).[50]

Comar reiterates that ^{137}Cs is efficiently absorbed from the gastrointestinal tract for transport to muscle tissue and milk.[23] He cites literature values for concentration in the muscle tissue of cattle of about 4 per cent of the daily intake per kilogram and values ranging from 10 to 30 per cent for sheep and pigs.

Ward and Johnson quote literature values for cattle which indicate that 1 to 2 per cent of the daily ingested dose appears in each kilogram of muscle tissue.[51] Their own work showed ^{137}Cs levels per kilogram of edible meat leveling off at less than 1 per cent of the daily intake in mature dairy cattle, at 3 per cent in feed-lot cattle, and at 15 per cent in calves. These differences may have been due either to age or feed type. Kreuzer reported a close correlation between ^{137}Cs in meat of cattle and the methods of feeding and management in Southern Bavaria.[52] The muscle of grazing cattle was more heavily contaminated than that of housed cattle during the grazing season. Change from housing to grazing was reflected in a rapid increase in levels.

Kahn _et al._, in a study of the effect of feeding practices on ^{137}Cs concentrations in cow's milk, and Porter _et al._, in a study of the Tampa milkshed, reported that 12 per cent of the daily intake of ^{137}Cs was transferred to the daily milk production when the animal is in equilibrium with its diet.[53,8]

Estimates of the biological half-life of cesium in the cow, the goat, the pig, and the hen fall in the range of 20 to 30 days although some workers report a value as short as 2 to 3 days for the goat.[23] In any event, the turnover in these animals is rapid enough for tissue levels to show little lag in following dietary levels. Retention of ^{137}Cs in the body is affected by potassium and sodium levels as well as by diuretic action, but these effects appear to be variable and relatively small.

Russell noted that an error in the estimate of expected milk levels of ^{90}Sr was systematically related to the particular fallout history of the preceding year.[35] Taking into consideration the use of stored hay as feed early in the year, he introduced an additional set of terms into the concentration equation:

$$C = p_r F_r + p_d F_d + p_1 F_1$$

where F_1 is the strontium deposition during the second half of the previous year and p_1 the "lag rate" factor. With this three-term expression, he obtained a much better correspondence of predicted values to observed values.[35]

Pendleton et al. state that ^{137}Cs is even more available to organisms living in equatic environments than in terrestrial environments, and, because of the trophic level effect already cited, predacious fish may accumulate this nuclide up to 10,000 times the level in the water.[50] Comar reports that ^{137}Cs is concentrated in the flesh of aquatic organisms by a factor of 5 to 20 over that of the surrounding water and Gustafson reports high concentrations of ^{137}Cs in fresh water fish.[23,54,55]

Cesium-137 in man

What has been said about cesium metabolism and distribution and about the increase in the ^{137}Cs/potassium ratio with the trophic level in animals applies generally to man as well.[34,50] The data of Pendleton et al. indicate that the increase ratio is larger in adult humans than in children.[50]

Cesium-137 retention in humans has been described as multi-compartmental function, but many workers have found or believe that a single exponential is a satisfactory expression for most biological purposes.[56,57,58] Reported half-lives for the longer-lived component in adults are on the order of 80 to 140 days.

In 1962, Richmond et al. measured whole-body retention in four subjects and reviewed the literature.[56] Hardy et al. reported in 1965 on a study of retention of ^{137}Cs and ^{90}Sr following acute ingestion.[57] In this atudy, contaminated foods (^{137}Cs and ^{90}Sr, 20 and 60 times higher, respectively, than in the normal diet) brought back from Rongelap were consumed by a single individual over a seven-day period. In 1965, McGraw reviewed the collected measurements of the half-time of this nuclide in man and suggested a model for expressing this parameter as a function of age.[58] Eberhardt reported a different approach in 1967.[59] Using published data from a variety of sources, he derived an expression for ^{137}Cs biological half-life in humans as a function of body weight.

The maximum permissible body burden for ^{137}Cs for the occupationally exposed, as recommended by the International Commission on Radiation Protection, is 30 microcuries (μCi).[60] If one: (1) applies a factor of one-tenth for members of the general population, (2) assumes that cesium retention can be described with a single exponential function with half-life on the order of 100 to 150 days, (3) assumes an equilibrium situation of constant intake, body burden, and excretion, and (4) follows the Federal Radiation Council recommendation of assuming that the maximum exposure by an individual is no greater than three times the average measured for a population group, then one can derive a maximum permissible daily intake on the order of 4,000 to 7,000 pCi/day. A value of 4,400 pCi/day has been used in the literature.[61,62]

Equations have been presented relating dose to ^{137}Cs intake and a number of authors have attempted empirically to fit models relating measured body burdens to measured levels of ^{137}Cs intake and thus predict future body burdens from intake data.[59,63-66]

Arctic ecosystems

In 1961, Baarli *et al.* and Liden found unusually high ^{137}Cs body burdens in people in Norway and Sweden, respectively.[67,68] These initial findings led to further studies which suggested that unique ecological situations exist with regard to radionuclides in the fallout-food-man chain in arctic regions.[69-72] Although these regions have less fallout than many other areas of the world, levels of ^{137}Cs in arctic inhabitants are among the highest documented.

High human body burdens have been traced primarily to the influence of ecological factors upon fallout accumulation in the lichen-caribou(or reindeer)-man chain.[73,74] Lichens represent a most important reservoir of ^{137}Cs and other fallout radionuclides because of their longevity (decades, even one century), persistence of aerial parts, and their dependence upon nutrients dissolved in precipitation. This effective retention of ^{137}Cs by lichens, the importance of lichens as a winter food for caribou, and the dependence upon caribou and reindeer for food by many northern peoples has resulted in this important ^{137}Cs concentration process.[75,76,77]

In the United States, studies of radionuclides in the arctic food chain were addressed initially to questions associated with the proposed Project Chariot site located near Ogotoruk Creek in northwestern Alaska.[78] Since 1963, surveillance activities have continued and expanded in Alaska.[79-84] The Battelle-Northwest Laboratory has studied the body burdens of ^{137}Cs in northern Alaskan residents (especially in Anaktuvuk Pass) since the summer of 1962.[85]

In April and May of 1965, the USPHS expanded the geographic area investigated by measuring ^{137}Cs body burdens throughout Alaska.[73]

Radiation dosage received by the average adult Anaktuvuk Pass Eskimo from ^{137}Cs body burdens during 1964, the year of highest values, was estimated at 135 to 150 millirem (mrem).[73] This value is about 30 per cent less than that amount serving as a Radiation Protection Guide for population groups during normal peacetime.

Environmental Radioactivity Measurements in Florida

Environmental radioactivity sampling and measurement within the State of Florida began with the collection of samples in 1957 by the Florida State Board of Health, Bureau of Sanitary Engineering.[86] This soon led to a statewide program of sampling and gross alpha and beta radioactivity analyses of surface and underground water resources, sediments, raw and finished water from water supplies, and sewage plant influents and effluents.[2-5] Some soil, aquatic organisms, and rainfall were also included in this early sampling. Gilcreas, Morgan, and Vreeland reported the establishment in 1957 of a routine program involving regular monitoring of gross alpha and total radioactivity of air and rainfall at the University of Florida and of surface and underground waters in Alachua county.[87,88,89] The results of these sampling programs are reported in a number of publications.[2-5,86-90] The University of Florida program also included twice yearly sampling and gross alpha and total-activity analyses of plants, animals, and soil.[87,88,89]

As a consequence of an agreement with the USAEC, the USPHS established the Radiation Surveillance Network in 1956 with nationwide stations for sampling of dry deposition (later discontinued) and air.[91,92] In 1957, the network was expanded and precipitation sampling was added. One station was designated at Jacksonville under the operation of the Florida State Board of Health with five to seven continuous precipitation and 24-hour air samples collected per week. In 1961, a second station was established at Miami.

In 1960, the Florida State Board of Health established similar air and precipitation sampling stations at Orlando, St. Petersburg, Tallahassee, and Pensacola; the six stations became known collectively as the Florida Radiation Surveillance Network.[2] Florida data from the state and national networks are reported in the various issues of Report of Florida Radiological Data.[2-5] In 1963, air sampling stations were also established at Titusville, Cocoa, and Melbourne in the vicinity of Cape Kennedy.[3]

In 1953, the USPHS established the National Air Sampling Network--a survey of airborne particulate pollution, including gross beta radioactivity, in urban and non-urban areas in the United States.[93] One 24-hour sample is ordinarily taken every two weeks; sampling frequency was increased at many stations during time of anticipated increases in airborne radioactivity due to fallout. Stations are designated for operation either yearly or during alternate years on a staggered schedule. During the period 1959-1964, stations operated every year both at Tampa and in the Florida Keys and on more selective schedules in Jacksonville, Miami, Orlando, and St. Petersburg. Results were regularly reported in Radiological Health Data.[28] Precipitation sampling was added at designated stations with the cooperation of the United States Weather Bureau in 1959. These included a station at Tampa beginning July, 1960.

A station of the 80th Meridian Air Sampling Program was established at Miami in June, 1957.[94] This network of stations near the 80th meridian (west) was established in 1956 by the United States Naval Research Laboratory with the assistance of the USAEC, the United States Weather Bureau, and interested groups in Canada and South America. The stations sampled ground-level air for measurement of radioactivity using a variety of experimental collecting devices. In 1957, all stations were supplied with filter samples and blower units, and the network was expanded to include locations of interest to the United States Air Force Cambridge

Research Center. It was operated in 1958 and 1959 as part of the International Geophysical Year Program on Atmospheric Nuclear Radiation and continued from 1960 through 1962 with a reduced number of sampling sites.[94,95] Individual samples were analyzed for gross beta radioactivity and samples were composited monthly for analysis for a variety of specific nuclides. Reports of results and 80th meridian concentration profiles from 1959 on appeared regularly in Radiological Health Data.[95] Lockhart et al. summarized the results for 1957-1962 and included a complete list of reports (26 references) resulting from this program during that time period.[94,96]

The USAEC Health and Safety Laboratory (HASL) deposition sampling program, in operation since 1958, employs two methods of collection for ^{90}Sr analysis.[97] In the "pot" method, precipitation and dry fallout are collected for a period of one month in an exposed stainless steel pot and transferred to a bottle for shipping, while in the "column" method precipitation and dry fallout are collected in a polyethylene funnel connected to an ion exchange column which is capped for shipment after a one-month collection. A station utilizing a pot collector is in operation at Coral Gables.

Operational responsibility for the 80th Meridian Air Sampling Program was transferred from the Naval Research Laboratory to HASL as of January 1, 1963, to be run in conjunction with the fallout sampling program.[98] Concurrent with this transfer, gamma analyses, both total-gamma activity and activity above 1 million electron volts (MeV) energy, were substituted for gross beta analysis as the initial means of sample evaluation. Ion exchange collectors for deposition sampling were added to the stations not having them. This resulted in the initiation of a column collection station at Miami in July, 1963, in addition to the Coral Gables pot station. Results of the 80th Meridian Program since 1963 and of the deposition sampling are reported in HASL technical reports, and summaries have been published periodically in <u>Radiological Health Data</u>.[28]

The USAEC and the United States Department of Agriculture have been conducting soil sampling, first individually and then jointly, since 1955 to determine world-wide distribution of ^{90}Sr.[99,100] During this course of sampling, collections have been made at both Miami and Jacksonville.

The Appalachicola River at Chattahoochee and the Escambia River at Century have both been sampled under the USPHS National Water Quality Network (currently the Federal Water Pollution Control Administration's Water Pollution Surveillance System).[101] This program was established in 1957 for sampling surface waters of major United States river basins for physical, chemical, biological, and radiological analyses. Weekly grab samples are collected for analysis at a central laboratory for gross alpha and gross beta activity in suspended and dissolved solids. Analyses are performed on either weekly samples or monthly composites, depending upon the amount of radioactivity expected. Strontium-90 analyses have been performed on selected quarterly compositíes from each station since 1959. Results have been reported for the Appalachicola River for 47 one-month periods between April, 1960, and June, 1965, and for the Escambia River for 39 such periods between May, 1961, and May, 1965.

In 1961, the Water Supply Activity, Interstate Carrier Branch, Division of Engineering and Food Protection of the USPHS, established a Drinking Water Analysis Program to gather extensive data on the radioactivity content of water supplies on interstate carriers.[102] Results have been reported for two samplings each at two water plants in Fort Lauderdale, two water plants in Miami, and a plant in Tampa during the period, 1960-1963.

In addition to the studies of radionuclides in milk already mentioned, a special study conducted by Consumers Union in July through August, 1958, included Miami among 50 North American stations from which milk samples were collected for ^{90}Sr and stable calcium analysis.[103]

In describing shellfish radioactivity research, Lackey reports the total background activity of oysters collected at two locations in Florida in March, 1958.[104] Gross alpha and gross beta determinations and gamma spectra were reported for analyses performed by the USPHS on oysters, fish, shrimp, crabs, and other biota, as well as on salt water and silt collected periodically during January, 1962, through January, 1963, from several locations in Brevard County, Florida.[2]

In summarizing results for a variety of food samples, Straub _et al_. reported ^{90}Sr and stable calcium levels for one sample each of radishes, turnips, and turnip greens from Florida, all collected from Cincinnati, Ohio, markets in 1959.[105]

In 1959, 1961, and 1962, the Miami area was included in the USPHS-sponsored Consumers Union studies of ^{90}Sr, ^{137}Cs, and other radionuclides in a typical diet of teenagers.[106,107] These studies were carried out periodically between 1959 and 1964 in a number of cities in the United States.

Florida samples have been included in the United States Food and Drug Administration's studies of radioactivity in domestic and imported food and animal fodder.[2,3,108]

Fifty-six Florida vegetable and citrus samples, collected January through March, 1964, by the Florida Department of Agriculture, were analyzed for specific nuclides by the Florida State Board of Health.[4]

The USPHS Institutional Diet Sampling Network, established in January, 1961, with 8 locations, was extended to 21 cities, including Tampa, in October, 1961.[109,110] Twenty-one meals plus snacks (three meals per day for seven days) are collected each month at a participating school or other institution in the respective cities. Solid food, dairy products, and wastes are analyzed as separate portions at USPHS regional laboratories. Results are reported in terms of weight consumed per day and concentration and daily consumption of calcium, potassium, phosphate, strontium-89 (^{89}Sr), ^{90}Sr, iodine-131 (^{131}I), ^{137}Cs, barium-140 (^{140}Ba), and either total radium or radium-226 (^{226}Ra). These results appear periodically in _Radiological Health Data_.[28]

In the previously mentioned study of background radiation in Florida, Williams _et al_. reported levels of natural radioactivity in the air from three Florida cities during 1963-1965, levels of gross alpha and gross beta radioactivity in food collected in 1964 at various places in the State, and gross alpha activity in ash from the 1963 samples of the state-wide milk sampling network.[29]

In addition to the special studies in Florida milksheds already mentioned, Cromroy _et al_. evaluated the exchange characteristics of soil from a dairy farm near Tampa and measured ^{137}Cs uptake from this soil by different grasses.[111] They reported: (1) a higher ^{137}Cs level in pangolagrass growing at the Tampa farm than in samples from the Agricultural Experiment Station at Gainesville, (2) that the clay fraction of this soil was kaolinite, which exhibits ion exchange and absorption but has a layer spacing not favorable for "struttural absorption," (3) that the uptake of ^{137}Cs by this soil in a slurry test was much less than that reported for other soils, (4) significant uptake from the soil by grasses transplanted to this soil, (5) different rates of uptake with Bahia-grass, reaching an equilibrium level more rapidly than pangola, and (6) that the more slowly equilibrating pangola showed an indication of progressing toward an equilibrium level about 30 per cent higher than that of Bahia. The first sampling of a third grass, Bermuda, showed levels that indicated either a much lower equilibrium level than the other grasses or a slow rate of uptake. However, retarded growth prevented further sampling. Unfortunately these authors did not report radioactivity levels in these grasses prior to transplanting.

Reports of monitoring sewer effluent, air, surface water, and milk for tritium in the environs of the USAEC's Pinellas Peninsula Plant near St. Petersburg have appeared periodically since 1960.[30] In 1965, tritium levels were also reported for precipitation collected at Ocala during 1961 and 1962 as part of a global tritium distribution study.[112]

Hartgering described a worldwide sampling program in 1955 and 1956 to estimate human exposure by analysis of 24-hour urine specimens for isotopes of iodine, strontium, and cesium.[113] Participants were five to ten personnel at each of a number of military installations, which included McDill Air Force Base, Tampa.

Radioactivity of humans has been measured at a number of installations. In some of the published reports of ^{137}Cs measurements in humans at the Walter Reed Army Institute of Research, average concentrations have been tabulated by states.[114,115] The average of six determinations on Florida individuals during 1958-1959 was not noticeably different from averages in other parts of the country at that time.[114] In addition, the average ^{137}Cs levels and number of subjects were reported by quarter for Florida subjects for six quarters during 1960 and 1961 (a total of eight Florida subjects).[115] Seven Florida subjects were also identified as having been included in the United States averages for July, 1963, through August, 1964.[116]

Cesium-137 measurements have been made in humans at the University of Florida, but results have not yet been published.[117] It was announced to the press that a whole-body counter survey was to be made of residents in the Tampa institution participating in the Institutional Total Diet Sampling Program, but no published reports were found to date.[118] Undoubtedly, subjects from Florida have been counted at other installations even though specific Florida results have not been reported in the literature.

Current environmental radioactivity measurements in Florida

Current sampling in Florida for radioactivity analyses is shown in Figure 3.

At the present time, gross alpha and total radioactivity measurements continue to be made regularly at the University of Florida on samples of air, precipitation, sewage, water supplies, and surface water.[119] Monitoring also continues routinely on a statewide basis.[114] The Tampa Pasteurized Milk Network Station collects weekly milk samples; the two Radiation Surveillance Network Stations at Jacksonville and Miami continue regular operations. The Florida Radiation Surveillance Network Stations continue to collect air and precipitation samples, and the State laboratory analyzes these samples for gross beta activity. The statewide grab sampling of surface and underground waters and of water supplies continues

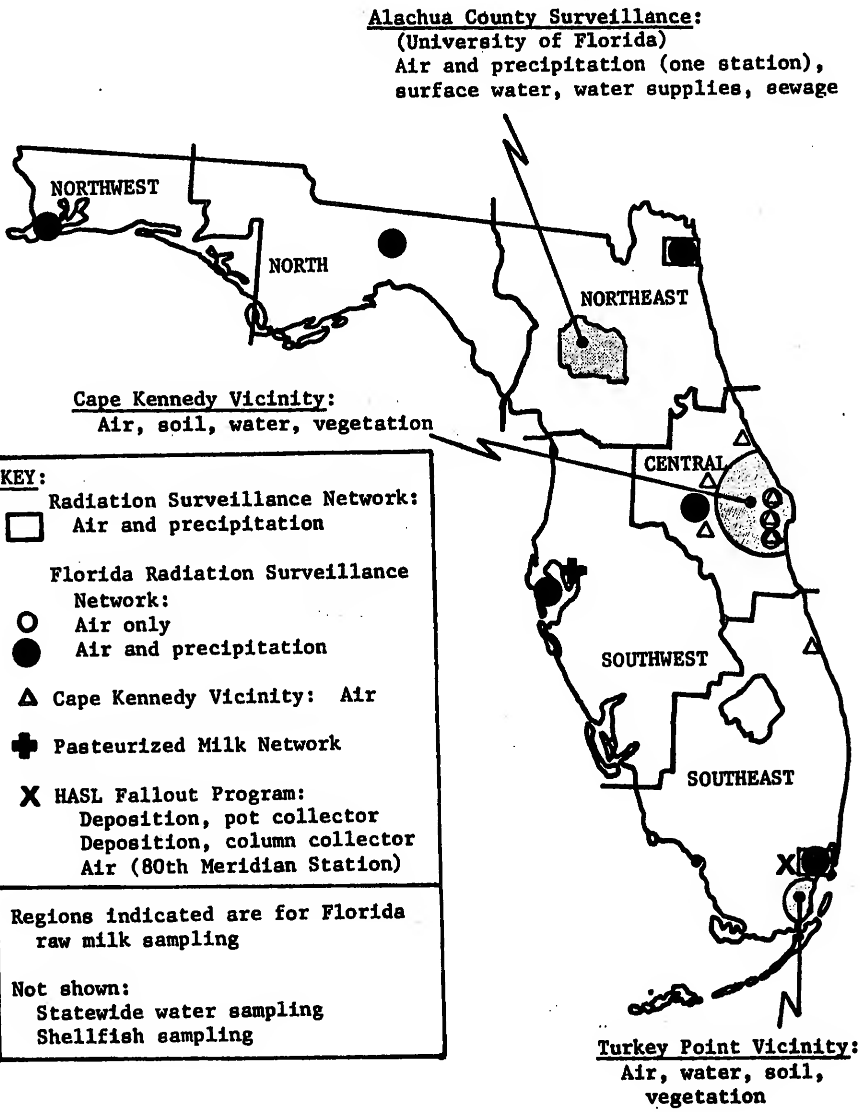


FIGURE 3. CURRENT SAMPLING PROGRAMS IN FLORIDA FOR ENVIRONMENTAL RADIOACTIVITY ANALYSIS

for gross alpha and beta analysis. Grab samples of shellfish are submitted to gamma-spectrum analysis, and then they are ashed and analyzed for gross alpha and beta activity. Raw milk samples are currently being collected on a statewide basis for combining into six regional composites for gamma spectrum and ^{90}Sr radiochemical analyses.

In addition, several special monitoring projects are underway in the State.[120] A cooperative plan has been drawn up by the United States Air Force, the USPHS, and the Florida State Board of Health to collect environmental samples in the vicinity of Cape Kennedy for radioactivity analysis. Air is being sampled for gross alpha and beta analysis at Fort Pierce, Orlando Air Force Base, McCoy Air Force Base, and New Smyrna Beach; the three previously mentioned state-operated stations at Titusville, Cocoa,and Melbourne function as part of this program. Soil, edible vegetation (including citrus), and non-edible vegetation are being sampled regularly and submitted to gamma spectral analysis, while water is collected and submitted to gross alpha and gross beta analyses of both the dissolved and suspended solids. Samples from the 12 sites within the first 10-mile radius of the area are analyzed by federal laboratories; samples from the 10 stations in each of the two 10-mile annuli beyond this are analyzed by the Florida State Board of Health Radiological Laboratory.

A special sampling program is also being carried out by the Florida State Board of Health near the site of the planned nuclear power plant at Turkey Point, a facility due to go into operation in the early 1970's.[120] Eighteen sites are sampled on a rotating basis; six are sampled per month so that each site is sampled quarterly. Analyses are similar to those for the Cape Kennedy off-site area.

Results from the Miami and Coral Gables stations of HASL's air and fallout sampling programs were included in the latest reports of these programs, and results were reported for the environment of the Pinellas Peninsular Plant of the USAEC as recently as February, 1967, implying that both these programs are currently in operation.[30,95,97]

Cesium-137 and Other Radionuclides in Selected Environmental Media - Methodology and Findings

As will be discussed in more detail in Chapter III, the earlier observations on radionuclides in Florida milk and feeds and the preceding review of cesium ecology and Florida radioactivity measurements led to the selection of meat and vegetables as the major items of study with nuclide emphasis on ^{137}Cs.

Although this study is intended to investigate levels and patterns of radioactivity in specific media, the ultimate significance of the findings lies in the influence of the observed levels on human radioactivity intake. Methods of evaluating dietary intake as reported in the literature can be grouped into three general categories: (1) evaluation of individual food items and, if all components of a total diet are evaluated, subsequent computation of total dietary intake, (2) analysis of the composite total diet of a real or hypothetical individual, and (3) analysis of urine and feces.[121,122,123,50]

Cesium-137 in individual food items

Meat sampling usually involves selection of some particular muscle, portion, or product for analysis. It has been reported that the concentration of ^{137}Cs can vary from muscle to muscle.[124] Fredrikson *et al.* reported considerable variation in the rate of cesium uptake between different muscles with the most active muscles taking up cesium most readily and the inactive muscles ultimately reaching a higher content,[125] Ekman reported that the half-time of ^{137}Cs varies between tissues, being longer, for example, in skeletal muscle than in most other organs or tissues.[126]

On the other hand, in a study of uptake and fate of ^{137}Cs in rats, cattle, sheep, swine, and poultry, Hood and Comar report that the most striking feature of cesium distribution in tissues was the constancy of the pattern, both between individual and species and between the various tissues of the body.[127]

Data reporting environmentally occurring levels of ^{137}Cs in meat or animals in the United States are limited; some of the first data was from animals maintained in the vicinity of nuclear testing sites. Cesium-137 levels were reported for unspecified muscle from cattle sacrificed in 1957 and 1958 from herds maintained in the vicinity of the Nevada Test Site, and another reference was made to cattle sacrificed before and after the 1961 "Gnome" underground nuclear test in New Mexico.[128,129]

The United States Department of Agriculture has reported ^{137}Cs and ^{90}Sr levels of beef rib meat surveyed in 1960 and beef soup stock surveyed in 1961.[130] The following year bologna and frankfurters were surveyed for ^{90}Sr only, and the results were not significantly different from those reported earlier in rib meat.

Plummer recently reported very little difference in concentrations of naturally accumulated ^{137}Cs between various muscles of the Georgia while-tailed deer; this prompted him to use the tongue as an estimator of general body levels.[131] Miettinen reported 1960 and 1961 ^{137}Cs levels in reindeer, potatoes, and certain other food items of Finnish Lapps.[72] He reports that there is an error of only a few per cent when analyzing the shoulder of the reindeer as representative of all consumed tissues. He estimates losses of activity in meal preparation to be about 10 per cent, but he does not make a corresponding correction in his calculations.

In a highly detailed study in the United States, Ward and Johnson reported ^{137}Cs levels in beef from dairy and feed lot cattle at Fort Collins, Colorado, in 1963.[51] They found no differences, within limits of counting statistics, in content between 12 retail cuts from the same animal as measured on four different beef animals. They concluded that, while greater precision in counting may have shown statistically significant differences between areas of the carcass, these would be small compared to the effect of differences in the ^{137}Cs contamination of the various animals' diets.

Straub reported surveys in which individual vegetable crop samples collected from markets in 1958 were analyzed for ^{90}Sr, ^{137}Cs, and other gamma-emitting radionuclides.[105] Food categories reported include fruits, fruit juices, meats, leafy vegetables, root vegetables, legumes and corn, and rice.

Setter _et al_. reported an elaborately planned survey of individual food items.[132,133] Market sampling was performed simultaneously with a food consumption survey for six quarters beginning July, 1962. Selection of samples was based on varieties and amounts of food available at the time of sampling as indicated by the United States Department of Agriculture marketing reports and local marketing data. Categories included meat, vegetables, fruit, and potatoes. During the six calendar quarters, ground chuck beef was sampled for four quarters and lean pork for two, lettuce for four and cabbage for two, and apples for four and oranges for two; potatoes were sampled for four quarters. Results for

^{137}Cs and ^{90}Sr were reported for each of seven regions and the whole of the United States. The lowest ^{137}Cs levels were found in lettuce, cabbages, potatoes, apples, and eggs. These authors stress that identification of food from production areas is difficult because of factors such as: (1) centralized marketing, (2) raising of the majority of poultry feed in one region, and (3) raising of beef in one region for fattening in another on feed from a third.

Highlights of the United States Food and Drug Administration's program of measuring radioactivity in individual domestic and imported human and animal food items included: (1) a call in 1954 for food packed prior to the nuclear era, (2) a subsequent program of gross radioactivity analysis, (3) gross beta activity measurements on a broad basis since 1958, and (4) emphasis on ^{90}Sr, ^{137}Cs, and ^{131}I since 1960.[108,134-138] Under this program, results of one ^{137}Cs analysis and about 100 ^{90}Sr analyses were reported for Florida samples collected in 1962 and 1963.[2,3,108]

Laug reported that in this program samples were mostly raw agricultural products, usually unwashed and unpeeled, and generally in the condition in which they were found in the warehouse or store.[138] For the purpose of data summarization, samples were assigned to classifications such as vegetables, brassicae, root vegetables, white potatoes, corn, and fruits (including tomatoes, cucumbers, pumpkins, and squash, as well as the berries, citrus, and other tree fruits). Cesium-137 levels were generally five times as high as ^{90}Sr levels. Vegetables ranked in ^{137}Cs content from highest to lowest as follows: leafy vegetables, dairy products, brassicae, root vegetables, fruits, corn, and white potatoes. Laug also states that, although the first year following the resumption of atmospheric tests was not marked by dramatic increases of ^{90}Sr in foods, sporadic higher concentrations were noted in leafy crops. Fallout concentrations were reported to be higher in the eastern and central United States than in the West, an effect believed to be associated with rainfall. In an experiment with a leafy vegetable and one kind of brassicae, commercial processing (washing and canning or freezing) reduced ^{137}Cs by 20 per cent.

The Florida State Board of Health determined and reported ^{137}Cs and zirconium/niobium-95 ($^{95}Zr/Nb$) contents of 56 leafy, non-leafy, root, brassicae, and miscellaneous vegetable samples and fruit samples that had been collected by the Florida Department of Agriculture in 1964.[4]

Bruce reported the levels of ^{137}Cs and ^{90}Sr found in milk, meat, root vegetables, and leafy vegetables in the United Kingdom during 1962-1964 and in milk and meat during 1965.[139,140] Cesium-137 levels in meat from cows, sheep, and reindeer in Norway were reported by Hvinden for the period 1959-1965 and by Madshus for 1964-1965.[141,142] Madshus took the precaution to collect all samples from the semi-tendinous muscle when sampling meat.[142] He also reported average ^{137}Cs levels by zone for potatoes and carrots for the fall of 1964. These two items were chosen for sampling because of universal production. DeRuyter and Aten estimated intake by humans in the Netherlands by sampling urine and feces.[122] Between November, 1964, and March, 1965, they also sampled some individual food items, including milk, grain products, kale, and brussels sprouts.

Uptake of ^{137}Cs by some leafy vegetables should be similar to grasses and legumes. Porter <u>et al</u>. and Cromroy <u>et al</u>., in the studies reviewed earlier, reported ^{137}Cs levels in grasses and animal feeds collected near Tampa, Florida.[8,111] Csupka published ^{137}Cs levels for grass and alfalfa in Western Slovakia during 1962-1965, and Wykes reported levels of this nuclide in grass in Minnesota in 1965.[143,47]

The highest ^{137}Cs levels in lean tissue samples in the United States are those reported by the National Center for Radiological Health in a program of monitoring unspecified muscle from Alaskan caribou and reindeer during 1962-1966.[82] Pendleton et al. reported ^{137}Cs levels that were measured in mule deer meat by gamma counting the hip to knee portion of skinned hind legs.[50]

Cesium-137 in the total diet

A number of total diet studies conducted in the United States are pertinent to this research because they report nuclide levels in individual food items or categories, illustrate how food items have been classified into categories, or report effect of food on total intake for the area and time represented.

In the Consumers Union total diet study, diets representing the total diet of teenagers for a week in the particular sampling region were prepared by home economists from produce purchased in local markets, and the composites were submitted for analysis.[144]

Reference has already been made to the Tampa station of the National Institutional Diet Sampling Network.[109] Cesium-137 intake levels at this station have been among the highest in the network, a condition which is not inconsistent with high milk levels in that area and the fact that dairy products are one of the major contributors of this nuclide. Reports are available for the January through June, 1966, portion of this study. Baratta and Williams have made comparisons between these two studies.[145] They traced differences in observed levels of intake to differences in amounts of food consumed in each study.

In addition to the program of sampling raw food items, the Food and Drug Administration has a total diet study.[146,147,148] Sampling in this program includes 82 food items in the 11 categories of the food plan recommended by the United States Department of Agriculture as nutritionally adequate at a moderate cost level for boys 16 to 19 years of age (the highest intake group of the United States population). Items are sampled in amounts proportional to consumption as reported in the 1955 Household Food Consumption Survey. Samples are collected at designated cites (none in Florida) in the United States. The categories include leafy vegetables, smooth vegetables, root vegetables, potatoes, dried beans, fruits, and meat and eggs. The most recent report includes ^{137}Cs levels in the total diet at nine cities for February through November, 1965.

In March of 1960, the USAEC instituted a diet study in a single city.[149] This program was later expanded to three stations and it became known as the Tri-City Diet Study. In this study of the typical adult diet, foods are grouped and analyzed in 19 categories with ^{90}Sr and ^{137}Cs levels reported in each category for each city. Categories include meat, poultry, fresh fish, shellfish, eggs, fresh vegetables, canned vegetables, root vegetables, potatoes, dried beans, fresh fruit, canned fruit, and fruit juices. At Chicago, analyses of these samples for ^{137}Cs have been performed quarterly since 1961. At the other two stations, these analyses were performed at the end of each year through 1964 and then quarterly in 1965. Reports for each city include yearly quantity intake, nuclide concentration, and yearly nuclide intake for each food category and daily and percentage of total ^{137}Cs intake attributable to the five overall categories of milk, meats, cereals, fruits, and vegetables.[150]

Thompson and Lengeman draw attention to the fact that estimates of quantity of food consumed do not match the precision of the laboratory radionuclide measurements.[123] By use of different available consumption estimates, they show how estimates of intake based on the same laboratory data can vary as much as 75 per cent.

In spite of this potential difficulty, Rivera reported that ^{137}Cs body burdens calculated from intake data were in relatively close agreement to measured body burdens.[64] He suggested that diet analysis was a useful means of predicting body burden. He also stressed the need for better analytical techniques and more frequent food measurements.

Using ^{137}Cs/potassium ratios as a basis, Gustafson examined the relation between diet and human body burdens.[65] He concluded that diet corresponded to _in vivo_ data when the former was transformed by a factor of 3.0 and advanced in time by four months. In the same report, he extrapolated the human body burden to the end of 1966.

The factor of 3.0 used by Gustafson to transform diet ^{137}Cs/potassium ratios to correspond with body burden levels is consistent with the trophic level effect discussed earlier.[50] The four-month time advance used in this transformation is probably related to time required by the human body to equilibrate with changes in the diet and may also be related to storage of food before consumption.

The Federal Radiation Council developed predictions of the expected radionuclide levels in the diet in 1963-1965.[39,40] Since then, measurements of radionuclides in milk, the total diet, and humans have been reviewed for the years 1963-1965 and compared to the Council prediction estimates for these years.[61,62] In addition, levels for 1966 were predicted using the approach proposed by the Council. The observed values were in agreement with the predictions; the peak of intake by population groups appears to have occurred in 1964.

Other gamma-emitting radionuclides

In addition to ^{137}Cs and ^{40}K, food and vegetable samples are commonly analyzed in the various surveillance programs in the United States by gamma spectroscopy for any or all of the nuclides ^{131}I, cerium-144 (^{144}Ce), manganese-54 (^{54}Mn), ruthenium/rhodium-106 ($^{106}Ru/Rh$), zinc-65 (^{65}Zn), and $^{95}Zr/Nb$; by gamma spectroscopy or radiochemical methods for ^{140}Ba or barium/lanthanum-140 ($^{140}Ba/La$); and by chemical or emanation methods for total radium or ^{226}Ra.[78,105,106,109,110]

The Institutional Diet Sampling Network samples are analyzed for potassium, ^{137}Cs, total radium or ^{226}Ra, ^{140}Ba, and ^{131}I.[106] However, in reports for 1966 the latter two were reported to be below detectable levels. Zirconium/niobium-95 was reported for reindeer and caribou muscle in 1964.[82] Manganese-54, ^{65}Zn, $^{106}Ru/Rh$, and cerium/praseodynium-144 ($^{144}Ce/Pr$) were reported in lichens, mosses, sedges, and other plants in Alaska during 1959-1961.[78] As was noted earlier, the Florida State Board of Health reported levels of $^{95}Zr/Nb$ found in Florida vegetable samples in 1964.[4]

The presence of ^{65}Zn, a neutron activation product, was reported in a variety of foods collected in 1958-1959 and attributed to high altitude fallout by Murthy et al.[151] It was reported by Van Dilla in 1960 in beef liver, beef muscle, hamburger, and milk.[152]

Manganese-54 was reported in human and bovine liver in 1965 and in freshwater clams in 1966 by Sax and Gabay, and in the Netherlands diet in 1964-1965.[153,154,122]

Antimony-125 (^{125}Sb) was reported by Svensson and Liden in forest moss in 1965 and by Johnson et al. in Colorado forage in the years 1962-1965.[155,156] DeRuyter and Aten tentatively identified trace amounts of both ^{125}Sb and beryllium-7 in leafy vegetables in the Netherlands in 1964-1965.[122]

Cesium-134 was identified in 1960 by Krieger and Groche as a neutron activation component of fallout from some weapons tests.[157] During the period 1960-1964, it was found in air samples, elk, milk, wheat, and beef in the State of Washington and in caribou and reindeer meat, fish, and human inhabitants in the arctic.[72,81,158,159] This nuclide was thought to have wide distribution and to have been produced prior to 1961. It was present at the level of about 1 to 2 per cent of the ^{137}Cs levels.

Traces of sodium-22 (^{22}Na) have been reported in vegetables in the conterminous part of the United States in 1964; in fallout, herbage, milk, and total diet samples in Italy in 1962-1964; in elk, bass, milk, wheat, beef, and human urine collected in the State of Washington during 1963-1964; and in moose, caribou, and reindeer meat as well as human urine and Eskimos in Alaska in 1963-1964.[158,160,161,85] Perkins and Nielsen suggest that a considerable amount of ^{22}Na was generated during 1961-1962.[158] DeBortolli et al. show evidence that this nuclide is taken up from the soil rather than being deposition rate dependent.[161]

The activation product, iron-55, has been reported in arctic and subarctic food chains and residents, and in the blood of residents of the State of Washington.[76,85] The low energy X-ray, 5.9 thousand electron volts (keV), emitted by this nuclide does not interfere with the spectra of other nuclides previously discussed and probably would not be detected by a conventional counting system used to analyze for these others.[85]

Nuclear testing will add fresh fission products to those observed in environmental samples. Klement reported that transient increases in air and precipitation radioactivity were observed following the French nuclear tests of February and April, 1960, but that debris from these events added not more than 0.1-0.2 per cent to the total worldwide fallout of long-lived fission products.[162] Iodine-131 and ^{140}Ba were found in air, precipitation, forage, and, in some cases, milk following the Chinese nuclear tests of October, 1964, May, 1965, and May, 1966.[163,164,165,156,166] The Florida State Board of Health reported a very pronounced increase of several days duration in the gross beta activity levels of air in seven Florida cities following the 1964 test.[4] Following the 1965 test, gamma analysis of air samples in the United States showed ^{131}I, tellurium-132, molybdenum-99, and neptunium-239 in addition to ^{131}I and ^{140}Ba/La.[165]

Naturally occurring radionuclides of the uranium and thorium series may also be present in environmental samples. In 1960, Muth et al. reported the radium content of various foods in Germany.[167] Either total radium or ^{226}Ra is also reported for several of the total diet sampling programs in the United States.[110,168] In reporting a study of gamma activity in a variety of plants in the states of Kerala and Madras in India, Mistry et al. published vegetable ash gamma spectra that very clearly showed activity from thorium series nuclides as the principal component.[169] Klement reviewed the literature in 1965 and reported sources and typical levels of natural radionuclides in foods.[170]

Analytical methods

Analytical methods vary from laboratory to laboratory and are characterized by a combination of innovation and standard physical, chemical, and radiochemical procedures.

Two of the laboratory systems handling large numbers of environmental samples, the Health and Safety Laboratory of the USAEC and the laboratories of the National Center for Radiological Health of the USPHS have issued analytical reports and manuals in a readily available form.[171-182]

Boni has developed a number of procedures for analysis of ^{137}Cs and other radionuclides in environmental samples.[183,184] Other papers on the analysis of environmental samples by gamma spectroscopy have been published by Ward, Johnson, and their associates.[185,186] In 1966, Mercer reviewed analytical methods and corresponding references for measuring radioactivity in food and other biological materials.[187]

CHAPTER III

EXPERIMENTAL APPROACH

The programs of monitoring radionuclides in Florida milk have indicated that ^{137}Cs levels in Florida milk have characteristic geographic patterns with unusual levels in much of the State. These patterns and levels are particularly striking when compared to those of ^{90}Sr.

Reports of additional investigations of this situation indicated that the levels of ^{137}Cs in Florida milk are directly related to levels of intake of this nuclide by the animals and that high levels in milk are due to elevated levels in one or more types of locally grown forage.

The findings reported for milk suggested that an unusual radioecological situation exists in Florida. This suggestion in turn prompted a study of (1) the extent to which these unusual levels and characteristic patterns of ^{137}Cs extend to other media and involve other nuclides, (2) the influence of environmental radioactivity on human radiation exposure in Florida, and (3) some of the reasons for unusual environmental radionuclide levels in this state.

Selection of Media for this Study

Various media were examined with respect to the following characteristics: (1) their role in the food chain, (2) their distribution in the State, (3) the importance of their economic role, (4) their capacity for concentration of radionuclides, and (5) their likelihood of becoming contaminated. As a result, lean beef and vegetables grown for human consumption were selected for sampling.

Since cesium follows metabolic pathways in animals similar to those of potassium and is preferentially concentrated by animals, animals and animal products are important sources of exposure to ^{137}Cs. Beef and milk are among the most important animal products produced in Florida. Milk is an important source of human exposure to radioactivity; dairies are distributed throughout this state, and the dairy animals receive a large portion of their feed from local pastures and locally grown forage. However, milk was not considered in this investigation because it is already being routinely sampled on a statewide basis and is currently being studied in detail at the farm level.

In contrast, there have been no reported studies of specific radionuclides in Florida meats. In fact, Ward and Johnson referred to the paucity of any information on ^{137}Cs levels in beef.[51] Since high levels of ^{137}Cs have been reported in forage in parts of the State, beef animals deriving a portion of their diet from these forages will have correspondingly high intakes of this nuclide. Since ingested ^{137}Cs is distributed throughout the tissues, particularly the muscle, and because beef is an important part of the human diet, this commodity is potentially a significant source of ^{137}Cs intake in this state.

Other animal products produced in Florida for human consumption include poultry products and pork. However, in this state chickens and other poultry are commonly raised in batteries off the ground and are fed grains and supplemental feeds largely brought in from outside the production area. Consequently these animals receive little feed from the immediate environment. Hogs receive a variety of prepared feeds as well as refuse and scraps, but, since Florida is a "grain-poor" state, much of the hog feed is also imported. Because of the feed sources and management practices, poultry, egg, and pork radioactivity levels would probably show little relationship to the factors influencing the milk levels that originally instigated this study and therefore were not studied in this investigation.

From the fact that certain animal forages grown in Florida appear to have unusual levels and distinctive patterns of ^{137}Cs, it might be predicted that other vegetation grown under similar conditions of exposure to deposition and uptake from the soil would have similar patterns of this nuclide. Vegetables are an important commercial crop in this State (400,000 acres, production valued at over $250,000,000 in the 1965-1966 production year), they are consumed directly by man, and some vegetable crop or another is produced statewide.[188,189]

Although occasional ^{137}Cs analyses of Florida vegetable samples have been reported in the literature, there have been no systematic, statewide studies.

Selection of Beef Sampling Stations

A program was drawn up to sample animals raised at the Florida Agricultural Experiment Stations with supplementation where necessary by samples purchased directly from slaughter houses or butcher shops. Beef sampling stations are described in Table 2 and shown in Figure 4.

The beef sampling program was based on information obtained by consultation with the director of the University of Florida Meat Laboratory.[190] Within the Florida Agricultural Experiment Station system, beef animals are fed at seven sites within the State. Five of these sites are identified in Table 2 as Stations 1, 2, and 3; Station 4--Source 1; and Station 7. Beef animals are also fed at the Animal Nutrition Laboratory in Gainesville but none were killed during the course of sampling in this study. In addition, beef research is getting underway at the West Florida Station at Jay, but no animals were available for slaughter during the course of this study.

TABLE 2

BEEF SAMPLING STATIONS

Station Number	Location	Source of Animals	Collection Point
1	Belle Glade	Everglades Station	Meat Laboratory
2	Ona	Range Cattle Station	Meat Laboratory
3	Brooksville	West Central Florida Station	Meat Laboratory
4	Gainesville	1. Beef Research Unit 2. Local Farms*	Meat Laboratory Local Packing Plant*
5	Pensacola	Local Farms, Escambia and Santa Rosa Counties	Local Butcher Shops and Slaughter Houses
6**	Graceville	Local Farm	Farm where Raised
7	Quincy	North Florida Station	Local Packing Plant with Assistance of Meat Laboratory Director

*Special sampling.
**Special sampling, one sample only.

All of the beef feeding experiments culminate in slaughter and grading to determine the marketability (quantity and quality) of the meat raised under the various feeding programs.[190] Experiment station animals are usually slaughtered at the Meat Laboratory in Gainesville. In the

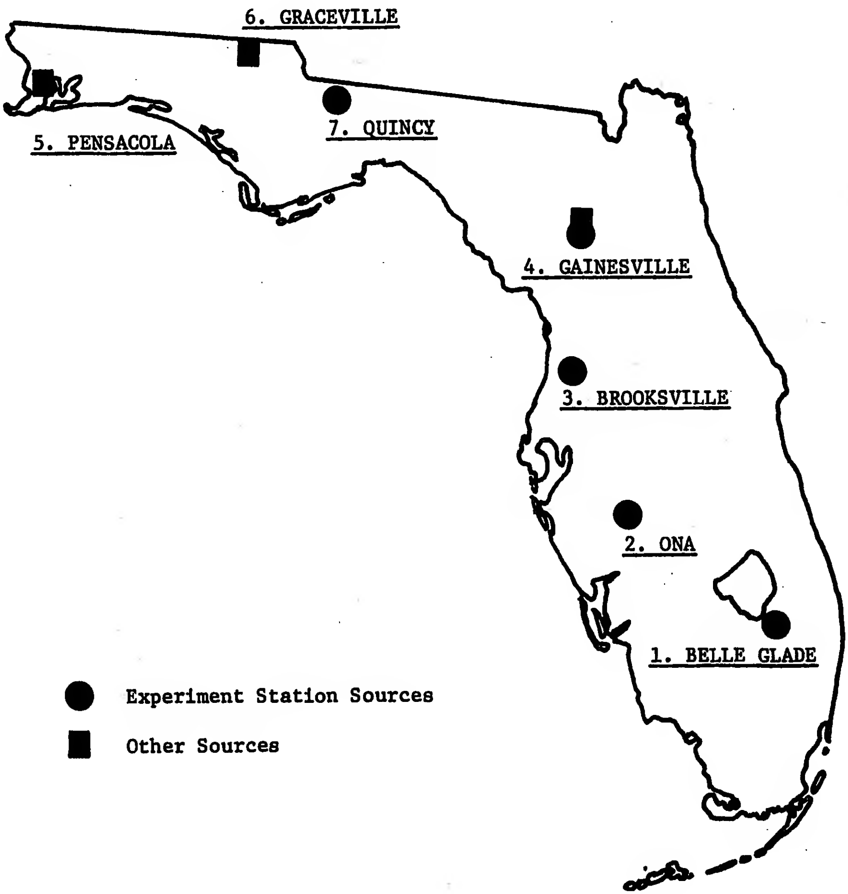


FIGURE 4. BEEF SAMPLING STATIONS

case of the station near Quincy, animals are slaughtered at a local packing plant in that city, but even these are graded by the director of the Meat Laboratory.

The sampling of experiment station animals has the advantages that: the majority of the samples are available from a single source, the identity of the carcass is maintained through the slaughter house, and detailed records are available if needed. These records identify the history and description of the animal, the feeding program, and the weight and grade of the carcass. In contrast, slaughter house animals are frequently bought at auction and mixed together in holding pens and carcass identity is not maintained, both of which make it difficult to identify the source and history of any sample collected. This is not of concern if the sole object of the study is to determine the average radionuclide exposure to the public from meat marketed at a particular point, but it does present a problem when it also is desired to relate meat activity levels to specific locations and feeding practices.

Beef Sampling

Since random sampling from certain feeding experiments could have resulted in the sampling of bulls far in excess of the small number appearing in retail cuts on the market, the restriction was placed that bulls were to be eliminated from sampling. A further restriction was placed that samples were to be from "control" animals or from those on the experimental diets most closely resembling the feeding practice of the particular area.

In sampling from butcher shops or commercial slaughter houses, samples were taken from meat on hand at the time, and special care had to be exercised to receive samples from animals that had been locally grown.

This was no problem in the several cases where samples were collected from country butchers who fattened their own beef.

Sex, breed, and age were not considered as variables in this experiment, but these and other factors were recorded where available for possible future use in investigation of any unusual or particularly interesting results. It was assumed that the distribution of different samplings from a station (often different experiments in the case of experiment station samples) and the method of selection would produce samples representing the average and range of radioactivity levels in fattened beef raised under various "typical" Florida conditions.

Sampling in May, 1966, of two carcasses from each of two experiment stations confirmed the feasibility of the mechanism of sample procurement, indicated the sample size necessary for analysis, and tested the sample preparation, the analytical, and the computational procedures.

The main beef sampling took place during the period January through June, 1967. After part of the samples were collected, the ^{137}Cs/potassium ratios were examined to help determine the total number of samples to collect. Inspection of the data suggested a trend with highest values in the southern and central parts of the State and lowest in the northwestern part. It was assumed that the "within stations" variance for all 1967 samples would not be greatly different from that computed for all samples collected up to that time (1966 and 1967) and that the final 1967 station means would not be greatly different from those observed up to that time. Estimations were then made of sample sizes necessary to show selected statistically significant differences.

Although inspection of the available data suggested differences between stations, it was found that, with the assumed variance, at least six samples per station would be necessary to conclude that any of these differences is statistically significant.

Vegetable Sampling

To minimize possible bias introduced by crop differences, stratified sampling was employed with samples collected within each of three categories (strata): (1) leaf and stem, (2) fruit, seed, and pod, and (3) root and tuber. This categorization is generally consistent with the one used by the United States Food and Drug Administration to summarize results of analyses of individual vegetables and with the food groupings used in the total diet studies described in the literature review. If sufficient numbers of samples are collected in each category, an examination of category effect is also possible.

Sampling was further stratified within these categories by specifying a total of 12 crop classifications to be sampled wherever available. These classifications, shown in Table 3, were set up by selecting and combining from the approximately 20 different vegetable crops grown in significant quantities in Florida.[188,189]

Selection was based on the amount of production and availability in the State, following consultation with a vegetable crops specialist from the Florida Agricultural Extension Service. Acreage figures taken from published reports are also included in Table 3.[189] The only high-acreage crops not included in the sampling were sweet corn and watermelons. The corn presented technical problems in the collection and preparation of sufficiently large samples of the edible portion, and the watermelons are seasonal in nature and represent only a small portion of a person's yearly intake.

The principal harvest season for most vegetables is between October and June.[188,189] Preliminary sampling was carried out in May and June, 1966, to test the feasibility of the sampling mechanism, to develop and test sample preparation, analytical, and computational procedures, and to determine the size of portion and counting time necessary to detect existing levels of radioactivity. Full scale sampling took place between January and July, 1967.

TABLE 3

VEGETABLE SAMPLING

Category	Crop	No. of[188] Areas Where Grown	Acreage[189] Harvested in 1965-66 Season
Leaf, Stem	Lettuce, endive, escarole, romaine	8	12,100
	Celery	5	12,200
	Cabbage	11	14,500
	Greens	many	not reported
	All Leaf, Stem	13 without greens	
Fruit, Seed, Pod	Beans, peas	12	43,500
	Pepper	10	16,800
	Tomato	10	51,600
	Cucumber	16	16,300
	Squash	13	10,000
	All Fruit, Seed, Pod	19	
Root, Tuber	Potato	9	43,500
	Turnip, Rutabaga	many	not reported
	Other (such as carrots and radishes)	2	not reported
	All Root, Tuber	10 without turnips	

In the case of vegetable sampling, sampling "stations" were identified by vegetable production areas as adopted from the designation by the vegetable crops specialists.[188] All samples from fields within one of these production areas were identified with that particular station number. In order to perform statistical analyses of the data and keep the number of samples to a reasonable level, the State was divided into regions and each station was assigned to an appropriate region. Initially the State was arbitrarily divided into four regions of approximately equal area and designated as Southern, Central, Northeast, and Northwest. The data itself, however, began to suggest that more homogenous regions would result if the lower east and west coasts were assigned to separate regions. This resulted in the regional designation shown in Table 4 and Figure 5 and identified as Southeast, Southwest, Central, Northeast, and Northwest.

Sequence of Study

Following sampling, the work was pursued in five stages:

1. Determination of kinds and levels of gamma radioactivity in the selected media,

2. Determination of variations in media, geographic location, and points in time,

3. Identification of unusual levels of gamma radioactivity in these media and development of hypotheses concerning causes and mechanisms involved,

4. Investigation of the interrelationships of the kinds and levels of radioactivity in various media, and the relationships with other available parameters,

5. Estimation of the effect of these nuclide levels on the radioactivity in food products, and in turn, on human radionuclide intake and radiation exposure.

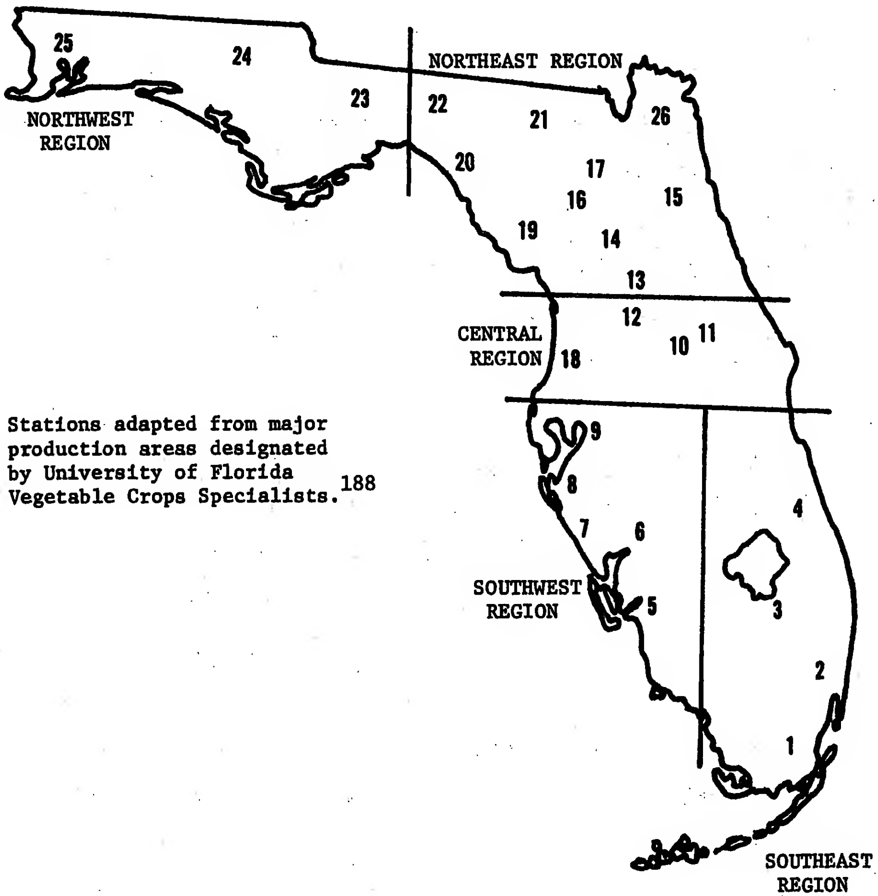
TABLE 4

IDENTIFICATION OF VEGETABLE SAMPLING REGIONS AND STATIONS*

Station** Number	Region	Area Represented (county or part of county)	Soil Type
1	Southeast	Dade	Marl, Rockland
2	Southeast	Broward, E. Palm Beach	Sands
3	Southeast	W. Palm Beach, N. E. Hendry, E. Glades, Highlands	Mucks, Peats
4	Southeast	Martin, St. Lucie, Okeechobee, Indian River, Brevard	Sands
5	Southwest	Collier, Lee, Hendry, Charlotte, Glades	Sands
6	Southwest	Hardee, S. Polk, DeSota, E. Manatee	Sands
7	Southwest	Sarasota	Mucks
8	Southwest	Manatee, S. Hillsborough, Sarasota	Sands
9	Southwest	E. Hillsborough, W. Polk	Sands
10	Central	N. W. Orange, S. E. Seminole, E. Lake	Mucks
11	Central	N. Seminole, S. Volusia	Sands
12	Central	Sumter, Lake	Sands
13	Northeast	Marion (less Weirsdale)	Mucks
14	Northeast	S. Marion, S. E. Alachua	Mucks
15	Northeast	Flagler, St. Johns, Putnam, Clay	Sands
16	Northeast	Alachua, Union	Sands
17	Northeast	Bradford	Sands
18	Central	Citrus, Hernando, Pasco	Sands
19	Northeast	Gilchrist, Levy	Sands
20	Northeast	Taylor, Lafayette, Dixie	Sands
21	Northeast	Columbia, Hamilton, Suwannee	Sands
22	Northeast	Jefferson, Madison	Sands
23	Northwest	Gadsden, Leon	Sands
24	Northwest	Calhoun, Washington, Holmes, Jackson	Sands
25	Northwest	Escambia, Santa Rosa	Sandy Loams
26	Northeast	Duval County	Sands

*Adapted from University of Florida Vegetable Crop Specialists.[188]

**Corresponds to station numbers as shown in Figure 6.



Stations adapted from major production areas designated by University of Florida Vegetable Crops Specialists.[188]

FIGURE 5. VEGETABLE SAMPLING STATIONS

CHAPTER IV

ANALYTICAL PROCEDURES AND EQUIPMENT

Beef Sampling

The standard procedure for beef sampling was to sample two animals from each group of animals. The most notable exception was the sampling of six animals at one time at Quincy, where there was only one slaughter of experimental animals during the course of this study. Three pounds of lean muscle were requested per sample; a total of 5 pounds was requested when the sample contained untrimmed bone and fat.

In sampling experiment station animals, samples were selected from the first two carcasses hanging in the cooler which met the restrictions of the sampling design (Chapter III). Samples in this case consisted of the portion associated with the 13th rib (3-4 inches of short loin) from either side of the carcass. This portion could be collected without seriously affecting the marketability of a hanging carcass.

In sampling other animals, it was not always possible to sample this portion or identify the portion sampled, particularly when samples were obtained through cooperating individuals.

Special samples from a packing plant at Station 4 (discussed in Chapter V) were collected in the cutting room from animals selected by packing plant personnel. When picked up in the shipping department, the samples were not identified as to the portion of the animal.

Other samples were cut from unspecified lean portions of carcasses of locally fed animals on hand at the butcher shop or packing plant at the time of sampling.

Vegetable Sampling

Samples were collected both directly from farms and from markets. A large proportion of the samples were obtained through the cooperation of the Chemistry Division of the Florida Department of Agriculture, which has an extensive program of sampling and analyzing agricultural products for pesticide residue. The remainder of the samples was obtained directly by the investigator, who visited both farms and markets, and through the assistance of volunteers, who collected and shipped in samples.

Vegetable samples were requested to be market samples that had gone through a washer or were in a condition suitable for washing in the laboratory. Portions of at least 3.5 kg of edible portion (8 pounds) were requested. If a large enough single portion were not available, a sufficient number of smaller portions from the same area were composited to make up a sample of the required size. All samples were identified by area where grown and, if possible, by farm or nearest community.

Vegetable samples received from the Florida Department of Agriculture laboratories were collected according to standard procedures of that department.[191,192] Briefly, field samples were composited from five portions, each collected from different parts of the field. Market or packing shed samples were composited from portions taken from a number of crates in order to represent the lot or originating field. These samples were then sub-sampled at the Department laboratory for analytical purposes, and the excess portions after sub-sampling were made available for this study. Some of the Department procedures call for quartering heads of leafy samples such as cabbage and lettuce with analysis of opposite quarters. When the original sample did not include sufficient heads to provide unquartered heads for this study, unused quarters were provided. In the case of a small number of samples, the entire sample was sliced before sub-sampling and the sliced excess provided for this study. In these cases, the possible effect of unwashed samples had to be considered in evaluating the data.

Field samples collected directly by the investigator or by volunteers were collected in essentially the same manner as those received from the Florida Department of Agriculture. Market samples collected by these individuals consisted of 8 to 10 pounds of produce selected from that on display at the market at the time of sampling. Market samples were collected only if the area where grown could be identified.

Beef Sample Preparation

Beef samples were prepared for analysis by boning, trimming the excess fat, and grinding the lean portion twice through a 3/16 inch (in.) grinder plate.

Vegetable Sample Preparation

In general, analysis of vegetable samples was performed on the whole of the portion corresponding to the particular category being examined (leaf and stem; fruit, seed, and pod; or fleshy root and tuber); samples were not peeled, cored, or shelled. Tips were not removed from bean pods, and seeds were not removed from squash and peppers. The entire portion analyzed was edible although not always eaten.

Unless already washed when received, vegetable samples were washed under running tap water, using a stiff bristled brush where necessary to remove clinging dirt. They were then drained dry or blotted with a turkish towel to remove excess moisture. After washing, samples were chopped into smaller pieces if necessary and triturated with either a meat grinder or a blender. About 1 milliliter (ml) of 40 per cent formaldehyde per 100 grams of finished sample was added as a preservative. The tap water used for washing was checked periodically for radioactivity to assure that washing did not introduce significant radioactivity.

More specifically, tops were cut off turnips, rutabagas, and other root vegetables at the top of the fleshy root, and stems were removed from hard skinned squash. These vegetables were washed and cut where necessary and then ground in two passes through a meat grinder with a plate with 1/8 in. holes.

Stems were removed from cucumber, peppers, soft skinned squash, and tomatoes; they were then washed, dried, chopped, and blenderized.

The outer leaves were removed from the cabbage head, the base of the head was cut off, and the remaining portion was then chopped and blenderized. Some cabbage was received as quartered heads; these quarters were trimmed along the cut faces before chopping to remove dirt and dehydrated material.

On the few occasions when untrimmed bunches of celery were received, they were topped near the center of the leaf cluster, and the upper portion was discarded; a portion of the base of all bunches was cut away. Individual stalks of celery, collards, and turnip, mustard, and other greens were separated from the bunches and washed, drained, blotted, chopped, and blended. It was necessary to scrub with a brush to remove clinging dirt at the base of the stalk. Collards presented a particular problem in that they were very tough and required considerably more distilled water for blending (up to 1,000 ml per 3.5-liter (l.) sample) than did other vegetables.

Heads or bunches of salad greens such as lettuce, romaine, escarole, and endive were cut in halves or quarters which were then further separated into clusters of a few leaves for washing, blotting, chopping, and blending.

The equipment used in sample preparation is identified in Appendix A. Several different pieces of power equipment were tested for first stages of sample reduction, but, since these devices had to be borrowed and returned for each batch of samples, it was deemed more convenient to prechop by hand with a butcher knife and cutting board.

In grinding, samples were precut only to a size that would pass into the grinder, ground material was collected in tared containers, the required amount of preservative was added and hand mixed, and the amount of preservative and the net weight of the sample were recorded.

The first step in blending was to weigh the washed, dried, and prechopped samples in a suitable container. A portion of the weighed, prechopped sample was then introduced into the blender along with the least amount of distilled water necessary to get blending action (none in the case of tomatoes and cucumbers; about 200-500 ml in the case of other vegetables). Once blending action started, larger pieces of the sample were introduced and additional distilled water was added only if necessary. The net weight of sample added to the blender was computed and recorded after taking the tare weight on the container. Any added water was measured and recorded by volume; unit density was assumed. The weight of blended material (sample plus water) was calculated and the required amount of preservative was added to and blended with the sample.

Preserved samples were transferred to labeled wide mouth plastic jars and held for analysis. Dilution factor due to added water and preservative was calculated and recorded for future use in calculating nuclide concentrations.

Gamma Radioactivity Counting

Individual pieces of equipment are described in Appendix A. Gamma-radioactivity analyses were performed with a multichannel scintillation spectrometry system. The detector was a stainless steel clad 4-in. by 4-in. right cylindrical sodium iodide (thallium activated) crystal coupled to a phototube. The detector was located in a shield 20 in. by 20 in. by 24 in. high (inside dimensions) with 2-in. thick lead walls, floor, and cover and with a 30 mil cadmium and 5 mil copper lining. Signals are analyzed by a 400-channel pulse height analyzer calibrated to 10.0 keV per channel.

Samples were loaded to the full mark in preweighed polyethylene Marinelli beakers, and the loaded beakers were weighed and placed over the detector in the shield. Two counting configurations were used; a 3.5-liter beaker was used for vegetable samples, and a 1.0-liter beaker was used for meat samples and vegetables of less than 2 liters in quantity. Samples between 2 liters and 3.5 liters in quantity were diluted to 3.5 liters.

Normally, samples were counted for 50.0 minutes; some of the samples at the beginning of the study were counted for 100 minutes.

At least two analyses, usually on the same or consecutive days, were performed on each lean beef sample. All the vegetable samples were given an initial count, and about two-thirds of these samples were recounted after 50 to 100 days. The delayed second count helped confirm the tentative identification of any short-lived activity and permitted recounting of the longer-lived activity in the presence of much reduced levels of the short-lived activity.

Gross (background plus sample) spectra for individual samples and a background spectrum for the particular counting period were digitally recorded for computer analysis and future reference. Backgrounds were also subtracted from the individual counts in the analyzer memory, and the oscilloscope displays of net spectra were inspected visually for the presence and relative size of peaks. The net spectra of all original counts and selected recounts were also recorded with the x-y plotter for future visual reference.

Interpretation of Gamma Spectra

A gamma spectrum, as developed on a multichannel gamma spectrometer, is a frequency distribution of counts registered in particular energy increments. Gamma rays are emitted by radioactive atoms at one or more discrete energies characteristic of the particular nuclide. However, because of the nature of the interaction process and the limitations of the instrumentation, gamma-ray photons of a single energy do not show a "line" spectrum at the output of the spectrometer system but rather show a continuous spectrum with an approximately Gaussian-shaped distribution (photoelectric peak) corresponding to absorption of the total energy of the photon plus a lower energy portion (Compton continuum) corresponding to smaller fractions of the total photon energy. In addition, the spectrum may show effects related to the configuration of detector and shield, such as backscatter peaks or escape peaks (peaks lower than the principal peak by the amount of energy carried away by X-rays escaping from the detector) and may also show secondary peaks corresponding to additional photon energies emitted by the nuclide. The resolution or width of the photopeak; the shape of, and area under, the Compton portion of the spectrum relative to the photopeak; and the importance of backscatter and escape peaks are functions of configuration and composition of the particular sample, detector, and surroundings.

The number and energy of specific photopeaks are utilized in identifying specific nuclides. The quantity of nuclide in a particular sample is determined from the area under some portion of the spectrum (such as the photopeak portion). The counts in a selected region are summed, and a conversion or "efficiency" factor, appropriate to the nuclide, sample configuration and counting system, is applied to convert this sum or area to the amount of nuclide in the sample.

In the case of a mixture of more than one gamma-emitting nuclide in a sample, the spectrum reflects all of the contributors and is assumed to be a linear combination of the individual contributors. Because of the limits of resolution (width of the photopeaks), additional peaks at other energies, and the Compton portion of the individual spectra, some members of the mixture will contribute counts in the region of, or in some cases even obscure, the principal peaks of various other members of the mixture.

In this study, gamma spectra were first inspected visually for the presence, location (energy), and relative size of peaks and then were evaluated by a computational procedure.

Computational procedures for interpreting gamma spectra were reviewed in the manual recently issued by the National Center for Radiological Health.[193] The method used in this study is the one described as the "Simultaneous Equations Method." This method was chosen for a number of reasons. It was the method most familiar to the investigator at the time of the beginning of the study and it was being used in a large number of public health laboratories at that time. This method is less subjective than the less elaborate stepwise nuclide-by-nuclide "stripping" method, and errors in the estimation are not compounded successively in one direction as severely as when stripping is performed. In addition, it is less sensitive to instrument instability than some of the more sophisticated methods and once the initial simultaneous calibration equations have been solved, computations can be performed with a calculator or even by hand if necessary.

Briefly, in this method the interferences between some number (N) of contributors to a composite spectrum are corrected for by setting up and solving a system of N simultaneous equations involving the unknown contributions of the N nuclides in terms of counts in the photopeak regions. The original equations are developed from counting a standard of each of the nuclides separately with the system and in the configuration to be used for the unknown samples. Once the simultaneous equations have been solved to yield explicit equations for each nuclide, these same latter equations can then be used for all subsequent sample computations until such time as the system needs recalibration.

A computer program for calibration computations was written in FORTRAN II and later converted to FORTRAN IV for use with the University of Florida IBM 709 computer. This program reads the conditions of the calibration and the counting data for the various standards. It then averages any replicate counts for the same nuclide, computes the matrix corresponding to the coefficients of the simultaneous equations, inverts the matrix in order to obtain the coefficients for the explicit equations, and computes counting efficiency (counts per disintegration) in the photopeak region for each of the nuclides. Identifying information, the computer coefficients, and computer efficiency are printed out and all the necessary calibration information is also punched on cards for subsequent use in computations. For performing the subsequent sample calculations, a program was written in FORTRAN II, later converted to FORTRAN IV for the IBM 709, and also converted to FORTRAN IV for use with the University of Florida IBM System 360 computer. This latter program reads in the calibration coefficients and efficiencies and reads the background counts and sample gross counts for each sample. It then computes and writes out the concentration and the associated two-standard deviation counting error of each specified radionuclide in terms of disintegrations per minute and pCi per unit (weight or volume).

CHAPTER V

RESULTS OF BEEF SAMPLING

Individual samples were collected from 45 animals at seven different stations.* Duplicate analyses were performed on the lean portion of each of these samples with repeat duplicate analyses on one sample for a total of 92 lean beef analyses. Four of the samples were collected from two stations during the preliminary work in May, 1966, and the balance were collected between January and June, 1967. A departure from the original plan took place when samples were collected at Station 4 from several groups of animals that were obviously not fed to the same degree of finish as the majority of the animals. The data from Station 4 are given special consideration in this presentation of results.

The major gamma-emitting nuclides observed in the beef samples were ^{137}Cs, ^{40}K, and ^{226}Ra and its decay products, bismuth-214 (^{214}Bi) and lead-214 (^{214}Pb). Quantitative computations were made for ^{137}Cs, ^{40}K, and ^{226}Ra in equilibrium with its daughters. It was necessary to include the latter activity in order to correct for the interference with the determination of ^{137}Cs and potassium by the ^{214}Bi that occurred in a large number of the beef samples. No particular significance is placed on the quantitative values for radium and daughters since there was no assurance that equilibrium actually existed and, indeed, no attempt was

*The single sample collected at Station 6 consisted of lean stew meat from the farm's retail outlet and although it was counted here as one animal, it may actually have represented more.

made to prevent the loss of the gaseous radon-222 intermediate. Cesium-137 is reported as pCi/kg of wet weight and as picocuries per gram of potassium (pCi/g K). Since essentially none of the potassium is contained in the fat, the latter method of reporting compensates for variations in the completeness of trimming of the fat. On the other hand, it obscures the effect on ^{137}Cs of fat distributed through the lean.

Cesium-137 Content of Lean Meat from Grain-Fed Beef, 1967

A summary of ^{137}Cs concentration, potassium content, and ^{137}Cs/potassium ratios of lean meat from grain-fed Florida beef sampled in 1967 is shown in Table 5 and in Figure 6. There is an apparent gradual decrease in ^{137}Cs content from south to northwest. The highest station average was 296 pCi/kg (78.8 pCi/g K) at Ona and the lowest station average was 105 pCi/kg (31.8 pCi/g K) at Pensacola. The highest individual value, 539 pCi/kg (133.6 pCi/g K), was also found at Ona and the lowest individual value, 55 pCi/kg (16.3 pCi/g K) occurred at Pensacola. The statewide average of all samples was 214 pCi/kg (54.4 pCi/g K).

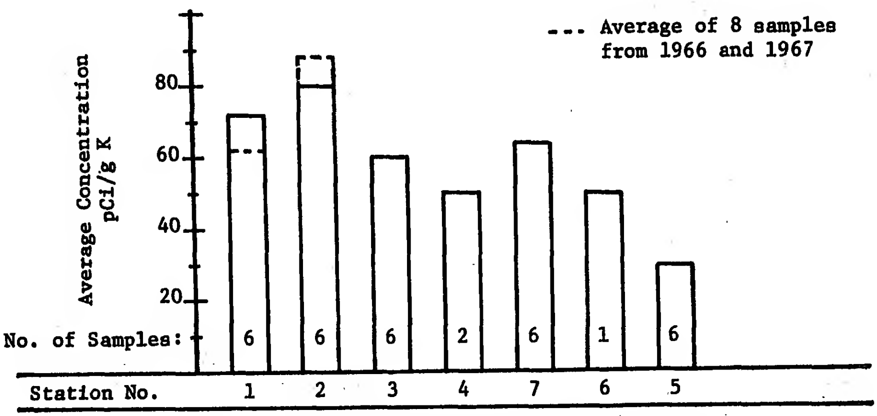


FIGURE 6. CESIUM-137 CONTENT OF LEAN MEAT FROM GRAIN-FED FLORIDA BEEF--1967

TABLE 5

SUMMARY OF CESIUM-137 CONTENT OF LEAN MEAT FROM GRAIN-FED FLORIDA BEEF, 1967

			^{137}Cs pCi/kg		K g/kg		^{137}Cs/potassium pCi/g K	
	Location	Animals	Ave.*	Range	Ave.*	Range	Ave.*	Range
1	Belle Glade	6	263 ± 10	109 - 403	3.65 ± .10	3.26 - 4.22	71.0 ± 3.4	30.4 - 95.3
2	Ona	6	296 ± 13	163 - 539	3.66 ± .12	3.30 - 4.04	78.8 ± 4.4	49.5 - 133.6
3	Brooksville	6	211 ± 11	153 - 283	3.57 ± .11	3.09 - 3.97	58.4 ± 3.7	49.5 - 72.5
4	Gainesville	2	173 ± 19	142 - 203	3.53 ± .19	3.31 - 3.75	48.4 ± 5.9	42.5 - 54.3
7	Quincy	6	214 ± 10	182 - 277	3.44 ± .11	3.27 - 3.58	62.1 ± 3.6	53.0 - 77.4
6	Jackson County (Graceville)	1**	175 ± 26	---	3.43 ± .26	---	51.1 ± 8.7	---
5	Pensacola	6	105 ± 10	55 - 174	3.37 ± .11	2.97 - 3.82	31.8 ± 3.3	16.3 - 54.0
	Statewide Average of Samples	33	214 ± 25	55 - 539	3.53 ± .05	3.09 - 4.22	54.4 ± 1.6	16.3 - 133.6
	Average of 7 Stations	-	205 ± 6	---	3.52 ± .06	---	57.4 ± 1.9	---

*Confidence intervals represent the two-standard deviation counting error only and are not based on the total variance from all sources.

**This single sample was a composite of lean beef from meat in the show case at the farm commissary and may have represented more than one animal.

The effect of geographic location on ^{137}Cs content of lean meat from grain-fed Florida beef was tested by performing an analysis of variance on the ^{137}Cs/potassium ratios reported for the samples at the five stations from which the full complement of six samples were collected (Stations 1, 2, 3, 5, and 7). This analysis, presented in Table 6, shows that, at the $\alpha = 0.01$ level, there is sufficient evidence to indicate a location effect.

TABLE 6

CESIUM-137 CONTENT OF LEAN MEAT FROM GRAIN-FED FLORIDA BEEF, 1967--EFFECT OF GEOGRAPHIC LOCATION (EXPRESSED AS CESIUM-137/POTASSIUM RATIO)

ANALYSIS OF VARIANCE

Source	Degrees of Freedom	Sum of Squares	Mean Square	F
Stations	4	7,526.77	1,881.69	4.56***
Error	25	10,312.11	412.48	--
Total	29	17,838.88	--	--

***Significant at the $\alpha = 0.01$ level.

The data were then examined to determine where significant differences occurred. The same five stations were arranged in order of ^{137}Cs/potassium ratios and the Tukey procedure for simultaneously testing all pairs of means was applied.[194] As shown in Table 7, the "eastern" stations (1, 2, 7, and 3) constitute a group for which there is insufficient evidence to indicate a significant difference between stations, but all stations in the group are significantly different from Station 5. The "northern" stations (5, 3, and 7) constitute a similar group, differing significantly from Stations 1 and 2. The inability to establish the significance of any smaller differences in station means is due to the high variation within stations. It is possible that by increasing the number of samples per station and/or by reducing the within station variance (such as blocking on some variance contributing characteristic not identified here) more of the apparent station differences could be demonstrated to be statistically significant.

TABLE 7

RANKING OF FIVE FLORIDA BEEF SAMPLING STATIONS ACCORDING TO CESIUM-137/POTASSIUM RATIO OF LEAN MEAT

BASED ON 1967 SAMPLES, SIX GRAIN-FED ANIMALS PER STATION

Station	2	1	7	3	5
Average pCi/g K	78.8	71.0	62.1	58.4	31.8

Solid lines indicate stations showing no significant difference at the $\alpha = 0.01$ level.

Simultaneously testing of all possible pairs of means according to the procedure of Tukey.[194]

Inspection of the data suggests a trend of highest ^{137}Cs levels in the southern part of the State, intermediate in the central and north-eastern part of the State, and lowest in the far western part of the State. Statistical testing of the data supports the hypothesis of a location effect and supports the hypothesis of a systematic geographical trend to the extent that the data can be grouped into two overlapping groups of adjacent stations.

Effect of Year of Collection

Two samples each were collected at Station 1 and 2 during May, 1966, and six samples each were collected at the same stations during 1967. As shown in Table 8, the two stations appear to show temporal trends in opposite directions, so that any overall year-to-year difference is insignificant, at least for the amount of data available.

TABLE 8

EFFECT OF YEAR OF COLLECTION ON CESIUM-137 CONTENT OF LEAN MEAT FROM GRAIN-FED FLORIDA BEEF

Station	No. of Samples		^{137}Cs pCi/kg			$^{137}Cs/K$ pCi/g		
	1966	1967	1966	1967	Ave.	1966	1967	Ave.
1	2	6	189.0	263.0	226.0	55.0	71.0	63.0
2	2	6	374.0	296.0	335.0	100.1	78.8	89.5
Ave.			281.5	279.5	280.5	77.5	74.9	76.2

Effect of Feeding Program and Meat Quality on Cesium-137 Levels

After cancellation of a scheduled kill of experiment station animals at Station 4, an attempt was made to obtain alternative samples from animals grown in the same general area. It was not possible in the time available to obtain samples that were of the same quality of meat as those obtained from experiment station animals and that were positively identified as locally fed. It was possible to obtain four samples from a local packing plant consisting of unidentified cuts from four locally fed animals. This meat was intended for ground beef and meat products (frankfurters, bologna, etc.) and was of a much lower quality than meat which is usually sold as retail cuts. These animals, slaughtered in April, 1967, were identified only as "two had some grain, two fed primarily grass."*

As shown in Table 9, two USDA GOOD samples collected in January, 1967, from two steers fed corn and citrus pulp on an experiment station dry lot in this same general area had ^{137}Cs levels of 204 and 142 pCi/kg (54.3 and 42.6 pCi/g K), respectively. Reference to Table 5 shows that these values fell within the general range of all the "eastern" samples. By contrast, the four packing plant samples had ^{137}Cs contents ranging from 293 to 12,500 pCi/kg (90.4 to 3,710 pCi/g K). Two of these animals had ^{137}Cs levels that were of a general magnitude about equal to (ranging from one to two times) those of the other "eastern" samples and the other two had levels 10 to 100 times as high. It was suspected at this point

*Comments by the packing plant manager on the feed of these and subsequent animals from this source were probably based on inspection of the condition of the animals and quality of the meat.

that the higher levels of ^{137}Cs in the meat were due to animal diets composed primarily of grass. Several weeks later, four additional samples were obtained from the same packing plant along with as much identifying information as obtainable in order to further investigate this source of meat. As shown in Table 9, all of these animals were cows of varying breeds, the meats were of low grade, and again two samples were identified as from grass fed animals and two from animals that had received some grain. A range of results very similar to the first four samples was obtained, with the totally grass-fed animals having the highest ^{137}Cs levels.

TABLE 9

VARIATION OF CESIUM-137 CONTENT* OF LEAN FLORIDA BEEF AS INFLUENCED BY FEEDING PROGRAM AND GRADE OF MEAT

Sample No.	Animal Description	Feed	Meat Grade	^{137}Cs pCi/kg	K g/kg	$^{137}Cs/K$ pCi/g
Samples from University of Florida Beef Research Unit (trimmed from short loin)						
			Collected 1/7/67			
7	Steer	Dry lot corn, citrus pulp	Good	204 ± 29	3.75 ± .30	54.3 ± 8.9
8	Steer		Good	142 ± 24	3.31 ± .24	42.6 ± 7.8
Samples from local packing plant buying only local animals (trimmed from unidentified portions)**						
			Collected 4/26/67			
36	4 animals identified only as "two had some grain, two fed primarily on grass"			293 ± 24	3.24 ± .24	90.4 ± 9.9
39				507 ± 29	3.51 ± .27	144.0 ± 13.7
37				1,840 ± 36	3.69 ± .27	498.0 ± 37.8
38				12,500 ± 73	3.36 ± .28	3,710.0 ± 309.0
			Collected 5/10/67			
50	Hereford cow	Some grain	Utility	567 ± 27	3.90 ± .26	145.0 ± 12.0
53	Hereford cow	Some grain	Cutter	808 ± 41	4.09 ± .38	198.0 ± 21.2
52	Jersey cross cow	Grass-fed	Not specified	5,530 ± 68	4.97 ± .40	1,130.0 ± 97.2
51	Brahama cow	Grass-fed	Canner	9,840 ± 64	3.62 ± .27	2,720.0 ± 200.0

*Confidence intervals represent the two-standard deviation counting error only and do not include other sources of variance.

**Comments concerning feed of these animals were supplied by packing plant. They are probably based on condition of animals and appearance of meat.

CHAPTER VI

RESULTS OF VEGETABLE SAMPLING

A total of 165 vegetable samples were collected, analyzed, and evaluated for gamma-emitting radionuclide content. Of these, 29 were collected during the preliminary sampling in May and June, 1966, from six different stations in four regions. The remaining 136 were collected during January through July, 1967, from 19 stations in the five designated regions. As summarized in Table 10, the 1966 samples represented 10 different crops and the 1967 samples represent the 12 designated crop classifications.*

The principal radionuclides detected in vegetable samples in addition to ^{40}K were ^{137}Cs and naturally occurring ^{226}Ra and its daughters. Following the arrival of fresh fallout, presumed to be from Chinese nuclear weapons tests, short-lived nuclides (half-lives on the order of one week) appeared briefly and moderately long-lived activities (half-lives ranging from one month to one year) appeared and gradually diminished.

Cesium-137 results are considered first in this chapter, and a consideration of other nuclide results follows.

*Crops, categories, stations, and regions are defined in Chapter III.

TABLE 10

NUMBERS OF FLORIDA VEGETABLE SAMPLES ANALYZED FOR GAMMA RADIOACTIVITY BY REGION, CATEGORY, AND SAMPLING PERIOD

Year, Category	Region SE	SW	C	NE	NW	State Total
1966						
Leaf, Stem	3	0	11	0	0	14
Fruit, Seed, Pod	1	7	3	0	2	13
Root, Tuber	0	0	2	0	0	2
All Categories	4	7	16	0	2	29
1967						
Leaf, Stem	14	13	13	9	9	58
Fruit, Seed, Pod	14	12	1	16	7	50
Root, Tuber	1	6	0	12	9	28
All Categories	29	31	14	37	25	136
1966 & 1967						
Leaf, Stem	17	13	24	9	9	72
Fruit, Seed, Pod	15	19	4	16	9	63
Root, Tuber	1	6	2	12	9	30
All Categories	33	38	30	37	27	165

Cesium-137 Content of Florida Vegetables-- Average Levels and Region Variation

The results are summarized by region for each of the two sampling periods and for the total study in Table 11 and in Figure 7. A striking feature of these results is the wide range of values reported for nearly all regions.

TABLE 11

CESIUM-137 CONTENT OF FLORIDA VEGETABLES
SUMMARY BY SAMPLING PERIOD AND REGION

	May-June, 1966			January-July, 1967			1966 & 1967		
Region	N	x̄	R	N	x̄	R	N	x̄	R
SE	4	50 ± 10	0-111	29	24 ± 6	0-176	33	27 ± 5	0-176
SW	7	71 ± 9	24-110	31	104 ± 7	0-357	38	98 ± 6	0-357
C	16	64 ± 7	0-561	14	118 ± 11	26-356	30	89 ± 6	0-561
NE	0	NS	NS	37	48 ± 6	0-214	37	48 ± 6	0-214
NW	2	24 ± 20	23- 25	25	23 ± 6	0- 67	27	23 ± 6	0- 67
Regions	4	56 ± 7	24- 71	5	63 ± 3	23-118	5	60 ± 4	23-98
Samples	29	61 ± 5	0-561	136	58 ± 3	0-357	165	59 ± 3	0-561

N = number of samples or regions
x̄ = average, pCi/kg wet weight
R = range of sample or region averages, same units as mean
NS = not sampled

Confidence intervals of means are based on the two-standard deviation counting error only and do not include other sources of variation.

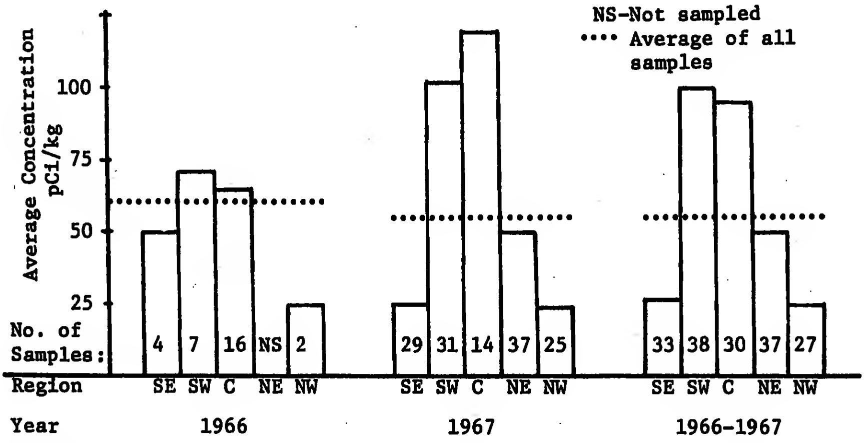


FIGURE 7. CESIUM-137 IN FLORIDA VEGETABLES ACCORDING TO SAMPLING PERIOD AND REGION WHERE GROWN

The ^{137}Cs content of the 29 samples from 1966 averaged 61 pCi/kg (wet weight) while the 136 samples from 1967 averaged 58 pCi/kg with an overall sample average for all 165 samples of 59 pCi/kg. The data suggest a geographic trend with maximum values in the regions designated Southwest and Central, the lowest values in the Southeast, and Northwest, and intermediate values in the Northeast.

The apparent regional effect was tested for the 1967 sampling period with an analysis of variance. Examination of the data showed that the variance increased in a general fashion as the mean increased and that frequency distributions of sample means were skewed to the right. Both observations suggest that in spite of other sources of variation, the analytical results were strongly influenced by the behavior of radioactive decay, which can be described by a Poisson distribution (variance equal to the mean). Accordingly, in order to stabilize the variance, the data was transformed for the purpose of the analysis of variance by taking the square root of the reported sample means. From this analysis, shown in Table 12, it can be seen that there was sufficient evidence at the $\alpha = 0.01$ level to indicate a regional effect.

The Scheffé procedure was used to test the transformed data to determine which of the regional averages were significantly different.[194] As is shown by the ranking in Table 13, the central, southwest, and northeast regions constitute a group for which there is insufficient evidence to indicate a significant difference between regions, but all of these regions are significantly different from both the northwest and southeast regions. The northeast, northwest, and southeast regions also constitute a similar group, differing significantly from the central and southwest regions.

TABLE 12

CESIUM-137 CONTENT OF FLORIDA VEGETABLES, 1967 SAMPLES
EFFECT OF GEOGRAPHIC LOCATION

ANALYSIS OF VARIANCE
(SQUARE ROOT TRANSFORMATION)

Source	Degrees of Freedom	Sum of Squares	Mean Square	F
Regions	4	618.50	154.63	9.49***
Within Regions	131	2,134.91	16.30	--
Total	135	2,753.41	--	--

***Significant at the $\alpha = 0.01$ level.

TABLE 13

RANKING OF FIVE FLORIDA VEGETABLE GROWING REGIONS
ACCORDING TO CESIUM-137 CONTENT, 1967 SAMPLES

(SQUARE ROOT TRANSFORMATION)

Region	Central	Southwest	Northeast	Northwest	Southeast
Number of Samples	14	31	37	25	29
Average $\sqrt{pCi/kg}$	9.28	8.87	5.83	4.30	3.78

Solid lines indicate regions showing no significant difference at the $\alpha = 0.01$ level.

Effect of Year of Sampling on Cesium-137 Levels in Florida Vegetables

Although there was insufficient data collected in 1966 to analyze the effect of both regions and years in a two-way classification, it still would be desirable to know if the average ^{137}Cs levels in vegetables changed over the two years. In addition, it would be desirable to combine the data from the two sampling years in order to provide more information per unit when the effect of individual stations, categories, and crops is examined.

In columns four and five of Table 14, the two years are compared within each of the regions in which sampling was conducted both years. There is not a great difference in overall averages of all samples, but when the data is examined within individual regions, there is a suggestion of region-year interaction, with a possible decrease with time in the southeast region, a possible increase with time in the southwest and central regions, and no apparent difference in the northwest region.

The difference between sampling periods was tested within each region as shown in columns six through nine of Table 14. A series of "t"-tests was performed on the transformed data using the pooled estimate of within years and regions variance. This provides a simple method of comparing years when there is insufficient data for a two-way classification. However, it must be kept in mind that in using successive "t"-tests, the chance of concluding there is a difference when none actually exists is compounded to a higher probability than indicated by the α level employed.

From Table 14, it can be seen that, with the method of testing employed, there is insufficient evidence to conclude a difference between years in the southeast, southwest, and northwest regions, even at the $\alpha = 0.20$ or greater level.

TABLE 14

COMPARISON OF CESIUM-137 LEVELS IN FLORIDA VEGETABLES
1966 AND 1967 SAMPLING PERIODS

Region	N 1966	N 1967	$\bar{x}$ 1966	$\bar{x}$ 1967	$\sqrt{\bar{x}}$ 1966	$\sqrt{\bar{x}}$ 1967	d	t
SE	4	29	50	24	5.954	3.782	-2.172	-0.978
SW	7	31	71	104	8.172	8.869	0.697	0.400
C	16	14	64	118	6.305	9.282	2.977	1.955*
NW	2	25	24	23	4.847	4.299	-0.548	-0.179
Average of Samples	29	99	61.0	62.1				
Average of Regions			52.2	67.2				

N = number of sample
$\bar{x}$ = average, pCi/kg wet weight
d = $\sqrt{\bar{x}_{1967}} - \sqrt{\bar{x}_{1966}}$
t = $d/s_p\sqrt{1/n_1 + 1/n_2}$
*Significant at the $\alpha = 0.10$ level.

The only region in which there is sufficient evidence to indicate a difference in years is the central region in which, applying the test outlined, the difference was significant at the $\alpha = 0.10$ level, with the value of "t" being very close to the critical value for the $\alpha = 0.05$ level.

In summary, with the small amount of data for 1966, there is not sufficient evidence to indicate a difference between years in the south-east, southwest, and northwest regions. There is some evidence for a difference between years in the central region, though the use of four successive "t"-tests may increase the probability of erroneously making this conclusion to a value greater than 0.05.

Cesium-137 Content of Florida Vegetables--Other Observations

The results of ^{137}Cs were also briefly examined with regard to such factors as collection station, vegetable category, and crop. Since various combinations of these factors were not sufficiently represented by the data, it was not possible to employ statistical testing to examine the results in greater detail than in the preceding sections. This does not prevent inspecting the data for apparent effects and trends. However, in doing this some of the observations will be based on relatively small numbers of samples and effects of the various factors will not always be completely separated.

Category effect

In Table 10, page 86 it was seen that not all categories were equally represented in determining regional means; this arrangement could bias the determination of regional effect if there actually were significant category differences.

Average ^{137}Cs levels are summarized by both region and category in Table 15 for each sampling period, and the corresponding ranges are summarized in Table 16. When 1966 and 1967 are considered as the single period, the statewide averages of samples within categories are almost identical for all categories and thus do not suggest a category difference. If all regions are weighted equally and category averages are computed by averaging regional averages within each category, a possible higher ^{137}Cs level is suggested for the "fruit" category. This is due primarily to the high "fruit" average for the central region and must be viewed with a certain amount of reservation since this particular average is based on only four samples.

TABLE 15

AVERAGE CESIUM-137 CONTENT OF FLORIDA[1] VEGETABLES--SUMMARY BY CATEGORY, REGION, AND YEAR OF SAMPLING
(pCi/kg)

Year, Category	Region SE	SW	C	NE	NW	Average of Regions	Average of Samples
1966							
Leaf, Stem	54 ± 11	NS	28 ± 8	NS	NS	41 ± 7	35 ± 7
Fruit, Seed, Pod	38 ± 25	71 ± 9	214 ± 16	NS	24 ± 20	87 ± 9	94 ± 7
Root, Tuber	NS	NS	39 ± 18	NS	NS	39 ± 18	39 ± 18
Average of Categories	46 ± 14	71 ± 9	94 ± 9	NS	24 ± 20	--	--
Average of Samples	50 ± 10	71 ± 9	64 ± 7	NS	24 ± 20	--	--
Average of:	All Categories	= 56 ± 7	All Regions	= 52 ± 6		All Samples	= 61 ± 5
1967							
Leaf, Stem	31 ± 8	81 ± 12	119 ± 12	55 ± 10	29 ± 9	63 ± 5	65 ± 5
Fruit, Seed, Pod	16 ± 8	132 ± 10	95 ± 31	30 ± 9	16 ± 13	58 ± 7	50 ± 5
Root, Tuber	31 ± 38	96 ± 18	NS	68 ± 11	22 ± 10	54 ± 11	58 ± 7
Average of Categories	26 ± 13	103 ± 8	107 ± 17	51 ± 6	22 ± 6	--	--
Average of Samples	24 ± 6	104 ± 7	118 ± 11	48 ± 6	23 ± 6	--	--
Average of:	All Categories	= 58 ± 3	All Regions	= 63 ± 3		All Samples	= 58 ± 3
1966 & 1967							
Leaf, Stem	35 ± 7	81 ± 12	77 ± 7	55 ± 10	29 ± 9	56 ± 4	59 ± 4
Fruit, Seed, Pod	17 ± 7	109 ± 7	184 ± 14	30 ± 9	18 ± 11	72 ± 5	59 ± 4
Root, Tuber	31 ± 38	96 ± 18	39 ± 18	68 ± 11	22 ± 10	51 ± 10	57 ± 7
Average of Categories	28 ± 13	96 ± 6	100 ± 8	51 ± 6	23 ± 6	--	--
Average of Samples	27 ± 5	98 ± 6	89 ± 6	48 ± 6	23 ± 6	--	--
Average of:	All Categories	= 58 ± 3	All Regions	= 60 ± 4		All Samples	= 59 ± 3

NS = not sampled
Confidence intervals of means are based on the two-standard deviation counting error only and do not include other sources of variation.

TABLE 16

RANGES OF CESIUM-137 CONTENT IN FLORIDA VEGETABLES SUMMARIZED BY REGION, CATEGORY, AND SURVEY PERIOD
(pCi/kg)

Year, Category	Region SE	SW	C	NE	NW	Range of Regions	Range of Samples
1966							
Leaf, Stem	0-111	NS	0- 82	NS	NS	28-54	0-111
Fruit, Seed, Pod	38(SS)	24-110	43-561	NS	22-82	24-214	23-561
Root, Tuber	NS	NS	36- 43		NS	39(SS)	36-43
Range of Categories	38-54	71(SS)	28-214	NS	24(SS)	—	—
Range of Samples	0-111	24-110	0-561	NS	23-25	—	0-561
1967							
Leaf, Stem	0-176	0-253	0-356	0-207	0-67	29-119	0-356
Fruit, Seed, Pod	0- 48	13-357	95(SS)	0-214	0-40	16-132	0-357
Root, Tuber	31(SS)	22-294	NS	11-113	4-42	22- 96	0-294
Range of Categories	16- 31	81-132	95-119	30- 68	16-29	—	—
Range of Samples	0-176	0-357	26-356	0-214	0-67	—	0-357
1966 & 1967							
Leaf, Stem	0-176	0-253	0-356	0-207	0-67	29- 81	0-356
Fruit, Seed, Pod	0- 47	13-357	39-561	0-214	0-40	18-184	0-561
Root, Tuber	31(SS)	21-294	35- 43	11-113	4-42	22- 96	0-294
Range of Categories	17- 35	81-109	39-184	30- 68	18-29	—	—
Range of Samples	0-176	0-357	0-561	0-214	0-67	—	0-561

NS = not sampled
SS = single sample, category, or region

Examination of individual category averages within regions does not suggest any consistent pattern. Even if there were a category effect which was not accounted for in the test of regional differences, the regional averages within categories, presented in Table 15 and superimposed on Figure 8, suggest that the same regional trend shown for all samples combined is also shown within each of the categories.

Station effect

Individual station averages for the combined years, 1966 and 1967, are presented in Figure 8. This figure shows the relative uniformity between station means within each of the regions and supports the rationale used to group adjacent stations into geographical regions. The portion of the figure for all samples regardless of category suggests that, with the possible exception of Station 13, no other grouping of adjacent stations into a small number of geographic regions could have produced more uniformity of station averages within regions. It may be noted that the average for Station 13 was based on only one sample which by itself does not appreciably affect the regional mean.

The uniformity of stations also holds generally, although with greater variation, within each of the categories.

Crop effect

Regional average ^{137}Cs levels for each crop are shown in Figure 9. This figure shows that, whenever a crop was sampled in a sufficient number of regions to make a comparison, the same general trend exists within crops as was shown within categories and as was shown for all samples combined. Deviations from the general pattern appear primarily to involve crop-region classes which have only small numbers of samples.

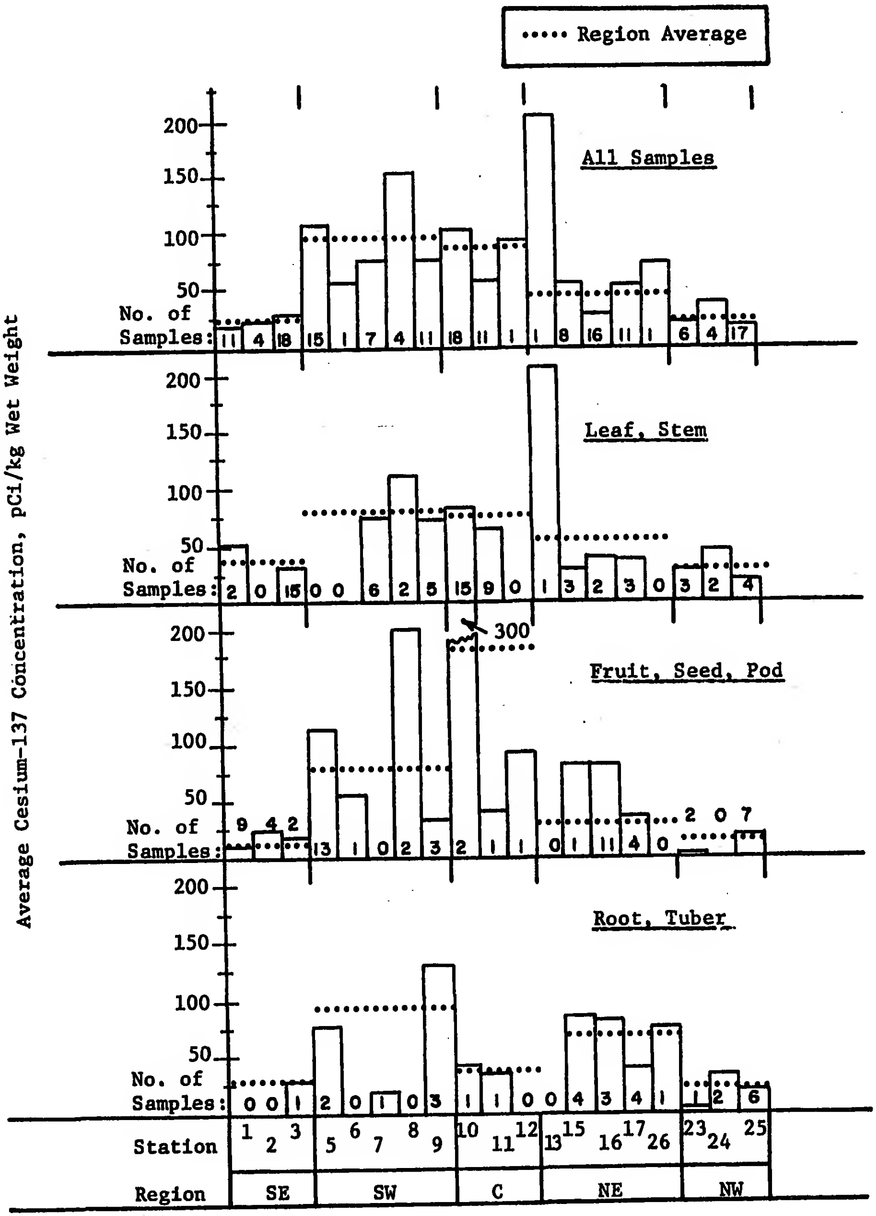


FIGURE 8. CESIUM-137 IN FLORIDA VEGETABLES, CLASSIFIED BY SAMPLING STATIONS WITHIN CATEGORIES, 1966 AND 1967

Figure 9 also supports the decision not to consider a category effect when region differences are analyzed. Although some crops appear to be consistently higher than the others, these crops fall in <u>all three</u> categories and there is considerable crop-to-crop variation within each category.

In Figure 10, the data are arranged by crop within region. This figure shows that the crops with the highest levels of activity (state-wide average greater than the overall average of 60 pCi/kg) were cabbage, greens, tomatoes, cucumbers, and potatoes. In this figure, it is also shown that these are the crops which exceed the respective regional averages for all crops in the greatest number of regions. Greens were consistently above the particular regional average in all regions. Cabbage was high in the central and southern part of the State but not in the northern part where most sample levels were frequently less than the associated counting error. The high overall tomato average was due primarily to the high averages at the two southwest stations where the tomatoes were sampled. Cucumbers were above the regional average in three of the five regions where they were sampled and the cucumber average was strongly influenced by two high samples at Station 10. Potatoes were above the regional average at each of the four regions where they were sampled.

<u>Unusual samples</u>

Individual results ranged from no detectable activity in a number of samples to several high samples exceeding 300 pCi/kg. Individual high samples are listed in Table 17.

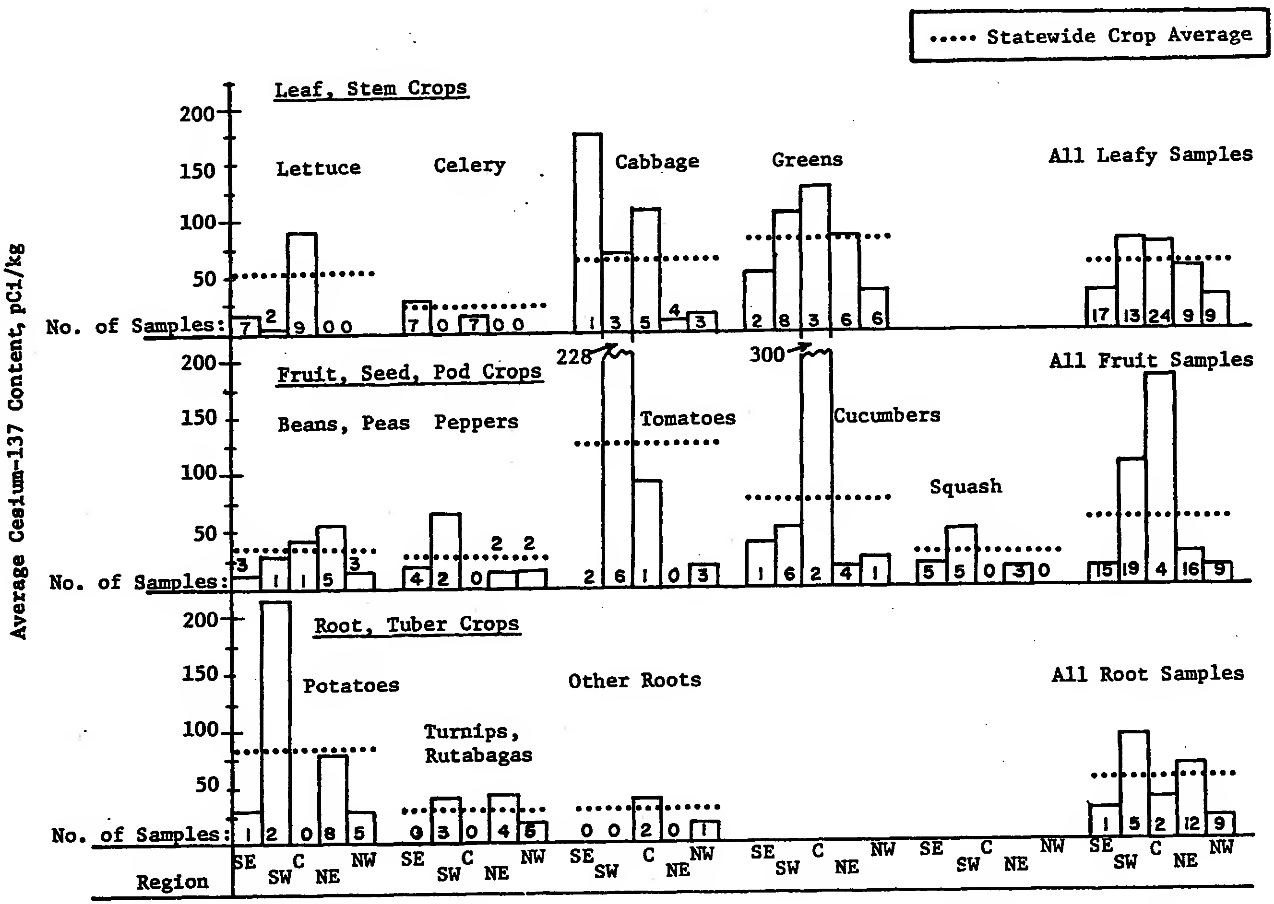


FIGURE 9. CESIUM-137 IN FLORIDA VEGETABLES, CLASSIFIED BY REGIONS WITHIN CROPS, 1966 AND 1967

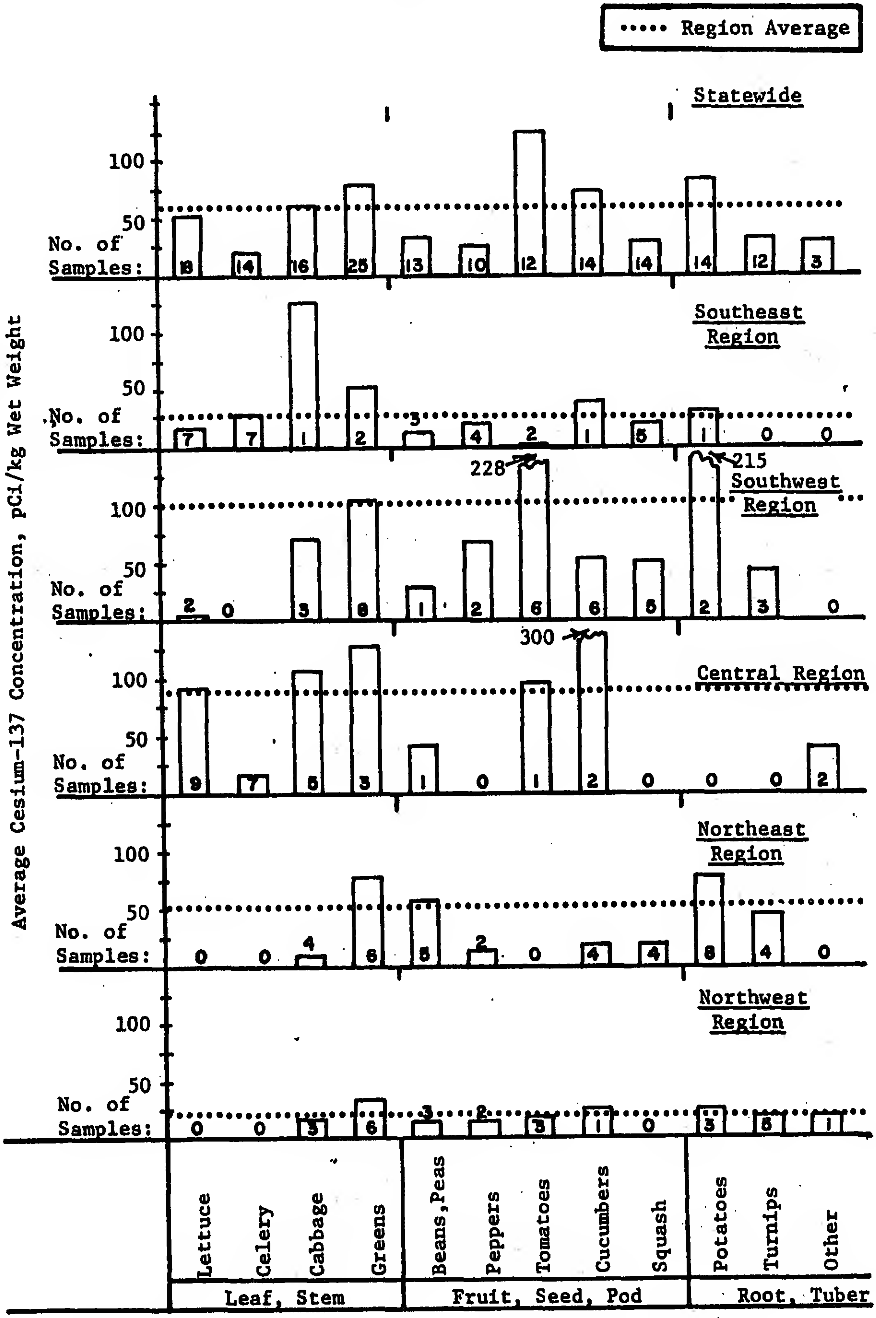


FIGURE 10. CESIUM-137 IN FLORIDA VEGETABLES, CLASSIFIED BY CROPS WITHIN REGIONS, 1966 AND 1967

TABLE 17

CESIUM-137 LEVELS IN FLORIDA VEGETABLES,
HIGH INDIVIDUAL SAMPLES

Sample No.	Collection Date	Region-Station	Category	Crop	No. of Analyses	Ave.* pCi/kg
15	5/23/66	C-10	Fruit	Cucumber	2	561 ± 40
166	5/28/67	SW-8	Fruit	Tomato	1	357 ± 36
140	3/2/67	C-10	Leaf	Lettuce	1	356 ± 47
139	4/11/67	C-10	Leaf	Lettuce	1	316 ± 43
152	4/11/67	SW-5	Fruit	Tomato	1	310 ± 34
138	3/2/67	C-10	Leaf	Cabbage	1	308 ± 40
154	4/11/67	SW-5	Fruit	Tomato	1	305 ± 36
197	6/7/67	SW-9	Tuber	Potato	1	294 ± 39

*Confidence intervals represent the two standard deviation counting error only and do not include other sources of variation.

Other Gamma-Emitting Radionuclides

Gamma-emitting radionuclides potentially appearing in Florida vegetation samples were identified from a consideration of (1) natural, fallout, and radioactive waste sources, (2) literature reports of radionuclides found in vegetation, and (3) nuclides found previously in Florida environmental samples. Following the inspection of some of the actual spectra, the following eight nuclides (or decay series combinations) were selected, in addition to ^{137}Cs, for quantitative computation: ^{144}Ce/Pr, ^{131}I, ^{106}Ru, ^{95}Zr/Nb, ^{54}Mn, ^{226}Ra and daughters, potassium (as determined from ^{40}K), and ^{140}Ba/La. Some physical characteristics and parameters used for computations are included in Appendix B for all nine components.

Radium and daughters

The radium decay products, ^{214}Bi and ^{214}Pb, appeared in detectable quantities in more than half of the vegetable samples. For the purposes of computation and correction for interferences with other nuclides, these nuclides were considered to be in equilibrium with the ^{226}Ra parent and calibration was performed with such an equilibrium mixture. These nuclides were included primarily in order to correct for interferences with other nuclides by the gamma rays of numerous energies emitted by this decay series. As in the case of the beef samples, no particular significance has been placed on quantitative values for this decay series because no attempt was made to assure equilibrium in the sample.

Manganese-54

Manganese-54 (314 day half-life) was included in the calibration mixture because it has been reported in vegetation samples in recent years and because it was found in forage and Spanish moss samples by this laboratory in 1965. However, essentially no ^{54}Mn was found in either 1966 or 1967 in vegetable samples in this study.

Fission products other than cesium-137

Iodine-131 and $^{140}Ba/La$ (8.05 and 12.8 day half-lives, respectively) were not detected in the 1966 samples. However, this does not conclusively demonstrate the absence of these nuclides since any quantities originally present could have undergone considerable decay in the 50-60 days that elapsed from collection to analysis of these particular samples. Both of these nuclides were found in the majority of the lettuce and greens samples collected during January 15-18, 1967. Barium/lanthanum-140 was also identified in some of the greens samples collected February 14-15, 1967, but no ^{131}I was detected at this time and neither was found at the next collection one week later or any time thereafter.

Ruthenium/rhodium-106 (1.0 year half-life) was not detected in 1966 samples, but activities identified as $^{144}Ce/Pr$ and $^{95}Zr/Nb$ (285 days and

65 day/35 days, respectively) were found in nearly all 1966 lettuce and greens samples which were collected May 21-24, 1966. Zirconium/niobium-95 and activities tentatively identified as $^{144}Ce/Pr$ and $^{106}Ru/Rh$ were found in nearly all the lettuce and greens samples collected January 15-18, 1967, in the majority of this type of samples collected February 14-15, 1967, and in a small number of samples collected February 22, 1967. These activities were found in essentially none of the samples collected after February, 1967.

These fission products were found in all parts of the State which were sampled during the indicated time periods. However, these activities were observed very rarely in other leaf and stem crops such as celery or cabbage or in fruit, root, or tuber crops.

Recounting after a decay period frequently showed a greater reduction in the activity designated as $^{144}Ce/Pr$ and $^{106}Ru/Rh$ than could be accounted for by radioactive decay alone. This observation suggests that a portion of the activity attributed to these nuclide pairs may have been due to the shorter-lived ^{141}Ce and ^{103}Ru (32.5 and 40 days half-lives, respectively) which are also found in fresh fission-product fallout.

CHAPTER VII

DISCUSSION OF RESULTS

Five aspects of the reported ^{137}Cs levels deserve discussion: the geographical variation, other variations in observed results, the magnitude of the levels observed, the influence of these levels on human exposure, and possible mechanisms and factors influencing these levels.

Geographic Variation of Cesium-137

Both the grain-fed lean beef and the vegetables show a geographic trend in ^{137}Cs levels. The highest values occur in the southern and central parts of the State, levels decrease in the northern parts of the State, and they are the lowest in the northwestern part of the State. This is the same general trend as shown by the raw milk levels reported by others.[2-7]

Geographic variations in these three media are compared in Figure 11. Geographical differences in milk and meat ^{137}Cs levels are not so pronounced as in vegetables. The vegetables show much lower levels in the southeastern part of the State, relative to the rest of the State, than do the other two media. Levels in the northeastern part of the State do not drop off as rapidly for milk and meat as for vegetables. The levels in meat from Quincy are higher, relative to the rest of the State, than milk and vegetable levels for the corresponding vicinity. Levels within the latter two media did not differ greatly between this vicinity and the extreme northwestern part of the State.

Since dairy and beef animals both receive other feeds in addition to locally grown forage, it can be expected that animal products would

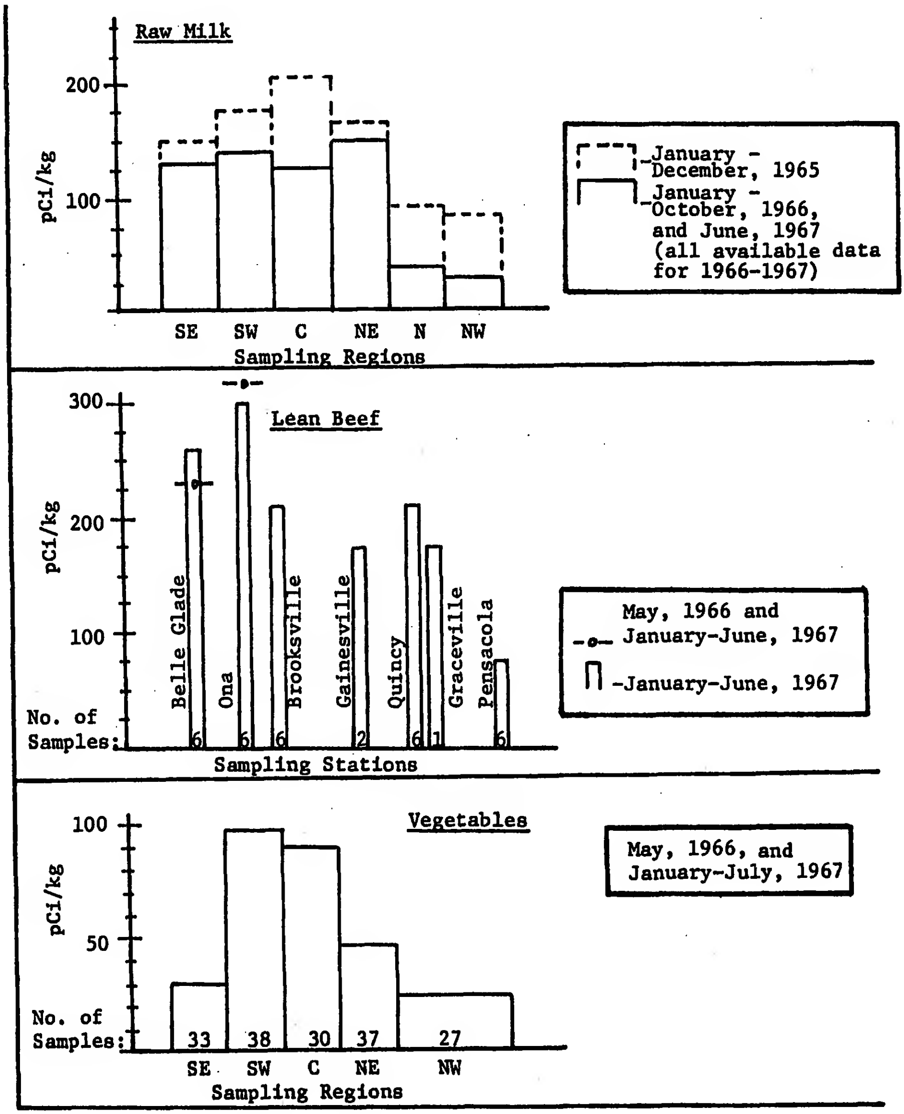


FIGURE 11. GEOGRAPHIC VARIATION OF CESIUM-137 IN FLORIDA MILK, BEEF, AND VEGETABLES

show less pronounced regional variations than vegetables. This may be part of the explanation for the striking difference in the southeastern region between relative levels in vegetables and those in the two animal products.

Where regional differences in milk levels are greater than they are in fattened beef, it is probably due to differences in ^{137}Cs intake by the animals. The intake of dairy cattle will be more characteristic of locality than the diet of feed-lot cattle if the dairy animals are consuming large amounts of local grass or hay, while the fattened beef animals are consuming considerably more grains and supplemental feeds. These latter feeds are likely to have lower levels of ^{137}Cs than grass and a much wider distribution of origins.

With the relatively short effective half-life of ^{137}Cs in cattle (on the order of 20 days), feed-lot cattle at slaughter would be essentially in equilibrium with their diet and would show very little reflection of levels of intake from pastures before they were put on the feed lot.

Other Variations in Cesium-137 Levels

There is a pronounced difference between ^{137}Cs levels in good to choice meat from feed-lot animals and those in lower quality meat from animals identified as having been fed on various amounts of grass. Although the magnitude of difference is much greater, the direction of the difference is similar to that reported by Ward and Johnson.[51] In 1963, the levels they found in meat from dairy cows and calves on pasture or hay were about three times as high as the levels they found in meat from dairy cows on hay and grain, and in meat from the feed-lot beef animals (Table 18). At the same time, although unable to completely separate the influence of

TABLE 18

CESIUM-137 LEVELS REPORTED IN BEEF AND OTHER MEAT BY VARIOUS INVESTIGATORS

Investigator	Ref. No.	Meat - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Roessler	--	Lean Beef, Good and Choice, Grain-Fed	Fla.-Statewide	Jan.-June, 1967	33	214	55-539
			South Fla.	May, 1966	4	282	162-402
			South Fla.	Jan.-June, 1967	12	280	109-539
			Northwest Fla.	Feb.-June, 1967	6	105	55-174
		Lean Beef, Cutter, Canner, Utility Grades, Grass-Fed	Northcentral Fla.	April-May, 1967	8	3,982	293-12,500
Farmer	128	Beef Muscle	Nevada Test Site	Dec., 1957	2	370	350-390
				May, 1958	3 Herds	470	NR
				Nov., 1958	3 Herds	770	NR
USDA	130	Beef Rib Meat	United States	1960	76	4.2*	0.1-32.4
USPHS	129	Beef Muscle	New Mexico	Early 1962	1	140	--
Setter et al.	133	Beef	United States	1962	52	47.5	0-290
			South U. S.	1962	12	47.8	0-290
		Pork	United States	1963	46	118	30-500
			South U. S.	1963	15	128	45-240
Ward and Johnson	51	Beef on Feed Lot	Ft. Collins, Col.	1963	44	101	--
		Dairy Cows on Hay and Grain			9	114	24-173
		Dairy Cows, Calves on Pasture			11	330	116-578
Straub et al.	105	Meats	Cincinnati, Ohio	August, 1958	NR	160	NR

TABLE 18 (continued)

Investigator	Ref. No.	Meat - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Tri-City Study	150	Meat	Average of 3 Cities	1962	3	54	42-69
				1963	3	122	71-156
				1964	3	96	24-142
				1st Qtr. 1965	3	286	213-338
				2nd Qtr. 1965	3	215	174-286
				3rd Qtr. 1965	2	126	93-159
				4th Qtr. 1965	3	210	145-276
				All of 1965	11	217	93-338
Gustafson	65	Meat, Extrapolated	Chicago	Jan.-June, 1966	NR	125	NR
				Jan.-June, 1967		100	
Hvinden	141	Meat, Cow	Norway	1959	NR	500	NR
				1960		700	
				1961		300	
				1962		1,000	
				1963		2,100	
				1964		1,100	
				1965		1,700	
		Meat, Sheep		1959		800	
				1960		2,200	
				1961		1,800	
				1962		2,000	
				1963		5,700	
				1964		2,100	
				1965		3,800	
Madshus	124	Beef	Southwest Norway	Late Fall, 1964	NR	900	300-2600
			Southeast Norway			2,500	1,600-4,400
			North Norway			2,300	800-4,100
		Lamb	Southwest Norway			6,700	1,600-17,400
			Southeast Norway			10,000	2,400-23,700
			North Norway			8,500	4,000-14,700

TABLE 18 (continued)

Investigator	Ref. No.	Meat - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Bruce	139	Meat, Home Produced	United Kingdom	1961	NR	21	NR
		Beef and Mutton		1962		62	
		Meat, Various Kinds		1963		135	
		Home Produced and Imported		1964		490	
	140	Meat, Home Produced and Imported		1965		280	
Miettienen	72	Reindeer Meat	Finnish Lapland	Winter, 1960	NR	35,300	NR
				1961	NR	--	7,400-18,000
Chandler and Snaveley	78	Caribou Meat	Alaska	Dec., 1963 - April, 1966	132	13,800	400-57,000
		Reindeer Meat			152	18,000	2,080-47,000
Anonymous	82	Caribou Meat	Alaska	Oct.-Nov., 1966	15	16,000	10,000-25,000
		Reindeer Meat			10	25,000	10,000-35,000
Madshus	142	Reindeer	Norway	1964-1965	10	33,500	10,800-51,200
Plummer	131	White-Tailed Deer in the Wild	Georgia	1966	NR	9,000	NR

N = number of samples
$\bar{x}$ = average content
R = range of reported values
NR = not reported
USDA = United States Department of Agriculture
USPHS = United States Public Health Service

*Determined by ashing and chemical separation.

age, diet composition, and type of animal, they reported transfer coefficients* from diet to meat that decreased with animal age and that were only about one-third as high for all dairy cows as for beef animals in the feed lot.

Ward and Johnson found that ^{137}Cs levels of forages exceeded those in grain by an order of magnitude.[51] Porter _et al._, in investigating the Tampa milk shed, reported levels for pangolagrass hay that were nearly an order of magnitude higher (on a dry weight basis) than other components of the dairy animals' diet and Williams and Nettles reported similar differences between Florida grasses and hays and other components of dairy feed.[8,7]

Porter shows Tampa pangolagrass ranging from 3,700 to 9,600 pCi/kg in 1963-1964.[8] Ward and Johnson report transfer coefficients of less than 0.01 for dairy animals, about 0.03 for beef animals, and 0.04 to 0.1 for six-month old calves on pasture.[51] Using an extreme ^{137}Cs concentration in grass of 10,000 pCi/kg, an extreme transfer coefficient of 0.1, and assuming an intake of 10 kg (dry weight) of grass per day, an equilibrium meat concentration of 10,000 pCi/kg can be predicted. This analysis suggests that _either the feed concentration, the transfer from feed to meat, or both, were extremely high in the case of several of the animals sampled_.

Possible sources of high intake, over and above the grass levels quoted, may have been poor pastures where the animals were forced to browse on close-growing plants in the litter, or where they had access to

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*Transfer coefficient-daily intake of radioactivity/radioactivity concentration per kilogram of meat.

the epiphyte, Spanish moss. Spanish moss (Tillandsia usneodies) was reported by Porter et al. to have very high levels of ^{137}Cs and similar results have been found in this laboratory.[8] Wykes, studying grassland in the Midwest, reported much higher concentrations of this nuclide in the root mat than in the grass.[47] As an example of the effect of differences in grazing, the presence of ^{137}Cs levels in moose 3 to 50 times lower than those in caribou from the same area has been attributed to the absence of lichen and possible plant litter in the moose diet.[70]

No attempt was made here to determine effect of sex, breed, age, or feeding program on the ^{137}Cs levels of beef from feed-lot cattle.

In this study, there did not appear to be significant differences in ^{137}Cs levels between categories of vegetables, but certain crops within various categories seemed to consistently exhibit the highest levels of this nuclide. In contrast to the results of this study, Laug reported great differences between vegetable categories in 1960-1962, with the following ranking, in decreasing order of activity: leafy vegetables, brassicae, root vegetables and fruits, and potatoes (Table 19).[138] He reported lower levels in potatoes than in root vegetables, while in this study, potatoes had consistently higher values than the other root crops. On the other hand, the data of Setter et al. shows levels in potatoes to be higher than those in lettuce and cabbage.[133]

No difference could be shown in this study between the sampling years 1966 and 1967. Because of the small number of samples collected in 1966, particularly of beef, a rather large difference must exist before it can be detected.

TABLE 19

CESIUM-137 LEVELS REPORTED IN VEGETABLES BY VARIOUS INVESTIGATORS

Investigator	Ref. No.	Vegetable - Description	Location	Date	N	pCi/kg $\bar{\bar{x}}$	pCi/kg R
Roessler	--	All Vegetables	Southeast Fla.	1966-1967	33	27	0-176
			Southwest Fla.		38	98	0-357
			Central Fla.		30	89	0-561
			Northeast Fla.		37	48	0-214
			Northwest Fla.		27	23	0-67
			Fla.-Statewide		165	59	0-361
Straub, *et al*.	105	Leafy Vegetables	Cincinnati, Ohio	August, 1958	NR	49.0	NR
		Root Vegetables				53.3	
		Legumes and Corn				57.0	
Setter *et al*.	133	Lettuce	United States	1962	105	12	0-900
			South		2	2.5	0-20
		Cabbage	United States	1963	56	2.8	0-75
			South		5	7.2	0-25
		Potatoes	United States	1962	45	14	0-55
				1963	64	36.6	0-250
			South	1962	2	35	--
				1963	9	57.0	25-110
		Apples	United States	1962	114	14.7	0-80
			South		17	16.0	0-45
		Oranges	United States	1963	25	20	0-90
			South		4	74	0-90
Food and Drug Administration	137	Snap Beans	United States	1960-1962	3	50	36-70
		Black-eyed Peas			3	250	195-309
		Cabbage			5	437	ND-1030
		Collards			1	511	--
		Mustard Greens			1	3,990	--
		Parsley			3	4,680	3,210-5,5540
		Carrots			1	51	--

TABLE 19 (continued)

Investigator	Ref. No.	Vegetable - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Food and Drug Administration	137	Potatoes	United States	1960-1962	2	104	73-135
		Turnips			1	413	--
		Pumpkin			1	90	--
		Tomato			1	38	--
Laug (Food and Drug Administration)	138	Leaf Vegetables	United States	1960-1962	20	1,470	ND-5,540
		Legumes (Beans & Peas)			34	264	ND-1,940
		Brassicae			17	124	ND-1,030
		Ave. Except Fruit and Root				570	ND-5,540
		Root Vegetables			13	79	ND-413
		Fruits*			51	79	ND-553
		White Potatoes			14	38	ND-291
Florida State Board of Health	4	Lettuce, Endive, and Parsley	Florida	Jan.-Mar., 1964	5	35	7-97
		Brassicae			16	20.5	8-41
		Greens			10	89	10-160
		Ave. All Leafy			31	41	7-160
		Rutabaga and Turnips			5	50	30-71
		Other Roots			5	29	4-35
		Ave. All Roots			10	39	4-71
		Mixed Leafy and Root			6	82	32-133
		Ave. All Vegetables			47	49	4-160
		Sugar Cane			1	91	--
		Citrus Fruit			4	32	17-52
		Citrus Skin			4	80	60-105
Tri-City Study	150	Fresh Vegetables	New York, Chicago, San Francisco	End of 1962	3**	7.6	3.2-10.3
				End of 1963		16	8-22
				End of 1964		6.1	3.6-8.6
				1965 Average	11***	49.5	9-179

TABLE 19 (continued)

Investigator	Ref. No.	Vegetable - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Tri-City Study	150	Root Vegetables	New York, Chicago, San Francisco	End of 1962	3**	29	18-38
				End of 1963		5.4	1.3-9.8
				End of 1964		8	3-12
				1965 Average		8	2.14
		Potatoes		End of 1962	3**	38	15-73
				End of 1963		26	10-46
				End of 1964		12	1-30
				1965 Average		13.5	1-30
Gustafson	65	Vegetables--Average Values from Extrapolation of graph	Chicago	Mid. 1965		46	--
				June, 1966		27	--
				Jan.-June, 1967		18	--
DeRuyter *et al.*	122	Curly Kale	The Netherlands	Nov., 1964 - March, 1965	NR	178	NR
		Brussel Sprouts				34	
Miettinen *et al.*	72	Potatoes	Finnish Lapland	Oct.-Dec., 1961	NR	10	NR
Bruce	139	Leafy Brassicas	United Kingdom	1961	NR	4	NR
				1962		19	
				1963		40	
				1964		30	
		Potatoes		1961		5	
				1962		32	
				1963		73	
				1964		30	

TABLE 19 (continued)

Investigator	Ref. No.	Vegetable - Description	Location	Date	N	pCi/kg $\bar{x}$	pCi/kg R
Madshus	142	Potatoes	Norway	1964-1965	95 farms	100	Max.=1,215
		Carrots				50	Max.= 250

N = number of samples
$\bar{x}$ = average content
R = range of reported values
NR = not reported
ND = nondetectable level

*Includes cucumbers, squash, and tomatoes, as well as berries and tree fruits.
**Composite fourth quarter samplings from three cities.
***Eleven composite samplings from three cities.

Variation with time within the 1967 sampling period was not considered. In Figure 1, it was shown that nationwide radionuclide levels in milk do show seasonal variations that reflect variations in levels in grass, which in turn reflect seasonal variations in deposition. To a certain extent, meat should reflect any fluctuations in diet, because of the short biological half-life of cesium in beef. However, there is very little information in the literature from which the degree of seasonal variation in ^{137}Cs levels in beef or vegetables might be determined. In the Tri-City program, meat showed very little evidence of seasonal variation and there was a gradual decrease during the year of 1965, though levels at San Francisco increased during the last quarter of that year.[65,150] Levels in Chicago vegetables, sampled quarterly since 1961, showed a general tendency to reflect seasonal deposition patterns through 1964. During 1965, when all three stations were sampled quarterly, levels fluctuated so greatly that no seasonal trend was apparent.

In contrast, levels of ^{137}Cs in Alaskan caribou and reindeer do show a definite seasonal pattern with maxima in the spring after a winter of grazing lichens, which accumulate the deposited material, and a minimum in the fall after grazing the summer on other plants, which accumulate the nuclide to a lower degree.[70,71] There was no apparent year-to-year trend through 1966 in the caribou and reindeer, primarily because levels in lichens were not changing appreciably.

It is entirely possible that the data in this study, all collected in the spring, reflects a seasonal effect and this must be kept in mind when evaluating the magnitude of reported levels.

Magnitude of the Observed Cesium-137 Levels

It is very difficult to compare values obtained in this study with literature values because the general levels of long-lived radionuclides in the environment have been decreasing the last several years and there is a one to three-year lag before results of radioactivity measurement appear in the literature.

Gustafson has extrapolated the radioactivity levels in food categories measured at Chicago beyond 1965 to the end of 1966.[65] These extrapolations were extended into 1967 for the purpose of comparison to the data collected in this study and 1967 values derived from these extended extrapolations are included in Tables 18 and 19. Such extrapolations can be justified on the basis of the following considerations:

1. Fallout deposition peaked in 1964 and has been decreasing since then with a half-life of about two years,

2. Plant uptake of ^{137}Cs is reported to be primarily rate-dependent and accordingly ^{137}Cs levels at harvest in both vegetables and animal feeds should be decreasing at approximately the same rate,

3. Cesium has a short biological half-life in cattle, and animal levels follow the levels of feeds; therefore animal levels should also decrease after 1964 with an approximate two-year half-life, and

4. Strontium-90 in the total diet has decreased since 1964 with a half-life of about three years and ^{137}Cs in the diet should be decreasing even more rapidly since it has less cumulative dependence.

Objections to such an extrapolation are the following:

1. Chinese weapons tests since 1964 are contributing some additional ^{137}Cs to the environment,

2. The degree of cumulative dependence of ^{137}Cs may vary from place to place,

3. Some feeds are stored and fed later, making animal levels less directly related to current deposition levels,

4. Gustafson's extrapolation was performed before the fourth quarter 1965 results were available, and does not take into account that ^{137}Cs levels in Chicago vegetables increased in the fourth quarter, and[150]

5. In considering a wider sampling basis, meat levels at San Francisco also went up during the fourth quarter, bringing the three-city average up as well.

Beef ^{137}Cs levels (pCi/kg) appeared to be about two to three times the reported milk concentrations (pCi/l) in the same region of the State. If it is assumed that on the average both beef and dairy animals were receiving comparable levels of ^{137}Cs intake, this observation is consistent with the ratio of the respective transfer coefficients as reported in the literature.[51] This is also consistent with the relative average levels in the two media as reported for 1965 in the Tri-City diet sampling program.[150]

Authors reporting levels of ^{137}Cs in meat over a number of years all show an approximate doubling from 1962 to 1963. If this doubling factor is applied to Setter's 1962 beef data to correct it to 1963, his results for throughout the United States are comparable to Ward and Johnson's 1963 results for feed-lot cattle in Colorado.[133,51] Differences between species of animals might be expected, especially since feeding habits differ greatly.[34] Because of possible species differences, some caution must be exercised in comparing findings to the Tri-City diet data or to Gustafson's data since the composition of the meat sample is not specified in either of these two.

It can be seen from the data of Bruce and of Hvinden in Table 18, that in the northern hemisphere, beef ^{137}Cs levels reached a peak some time during the period 1963-1964 and have been decreasing since then.[139,140,141]

Based on these temporal patterns, Ward and Johnson's 1963 data should represent levels just before or very near the maximum, and levels in comparable animals should have decreased to as low or lower levels by 1966 and 1967. The levels reported in this study for Pensacola are comparable to their 1963 feed-lot beef; the results for feed-lot beef from the balance of Florida were one and one-half to three times their 1963 feed-lot data and 0.7 and 0.9 times their values for pasture-fed beef.

The most recently published values for ^{137}Cs concentration in meat in the United States are the 1965 results from the Tri-City program in which levels are reported for meat as a category (exclusive of poultry, fish, shellfish, and eggs).[150] These data are from market sampling so that the first quarter 1965 maximum probably reflects the later months of the preceding year. It would seem reasonable to expect the spring 1966 levels to be similar to those at the end of 1965, and the spring 1967 levels to be somewhat lower. The overall average for Florida in this study, 214 pCi/kg, is comparable to the 1965 Tri-City average of 217 pCi/kg and the 1965 year-end Tri-City average of 210 pCi/kg. The range of station averages in this study, 105-296 pCi/kg, is comparable to the range of stations averages in the 1965 Tri-City sampling, 170-300 pCi/kg. The range of individual sample results, 55-539 pCi/kg, is wider than the range of individually reported values in the 1965 Tri-City sampling, 33-338 pCi/kg, but this would be expected if individually reported results in the latter program represent composite samples. The Tri-City results for 1965 are higher than the 1967 results for northwestern Florida but are only about two-thirds of the levels found for southern Florida.

The average values found in meat from the Pensacola sampling station are comparable to the extrapolated 1967 value derived from Chicago Tri-City program results. Average values for the other stations are all higher than this extrapolated figure, and the average for the Ona station is about three times as high.

The fact that levels in grass-fed animals were higher than in feed-lot animals is consistent with what was reported by Ward and Johnson.[51] However, the average of these eight samples, 4,000 pCi/kg, is an order of magnitude higher than the average reported by these authors for pasture animals in 1963, and the highest value found is about 36 times as high as their highest reported value. These extreme values are comparable to what has been reported for cattle in Norway and for reindeer and caribou in the artic.[72,78,124,139-142] The higher values are also comparable to the average level reported by Plummer for ^{137}Cs in Georgia deer browsing in the wild.[131]

It is even more difficult to compare vegetable results to those reported in the literature than it is for meat because ^{137}Cs levels reported in literature are so variable.

The average levels reported here for most Florida vegetables in 1966-1967 are higher than those reported by Straub for 1958 and by Setter for 1962-1963.[105,133] The regions of the State showing the lowest levels were occasional exceptions to this. For the most part, levels observed in this study were lower than those reported by Laug for the United States during 1960-1962, except for potatoes which had higher levels in the southwest and northeast regions.[137]

Statewide levels in this study were generally higher than the Florida averages reported for 1964.[4] Compared to this overall 1964 average, 1966-1967 levels were about the same or lower in the Southeast and Northwest, about the same in the Northeast, and higher in the southwestern and central parts of the State.

The Tri-City program samples showed considerable variation among cities and quarters in 1965. However, it would be expected from the trend since 1963 that 1966 and 1967 average levels would be lower than in 1965.[150] Average ^{137}Cs levels found in Florida vegetables in this study were generally higher than the average of 1965 levels reported for the Tri-City program, except for the above-ground vegetables in the southeast, northeast, and northwest regions of the State which had levels equal to or lower than the 1965 Tri-City values.

The levels of ^{137}Cs in vegetables in the Southeast and the Northwest are comparable to those derived for 1967 from Gustafson's extrapolations of Chicago data, while the levels in the rest of the State are higher than the extrapolated predictions.[65]

Since vegetables were selected for this study because high levels of ^{137}Cs have been observed in animal forages, it is appropriate to compare the observed vegetable levels to reported forage levels. The forage levels reported for Tampa forage by Porter _et al_. and Cromroy _et al_. are reported on the basis of dry weight.[8,111] In order to put results on a comparable basis, some of the vegetable results from this study, reported on a fresh weight basis, were corrected to a dry weight basis using United States Department of Agriculture values for water content of the various vegetable types.[195] The averages for each of the leafy crops collected in the southwest region (the region including Tampa) were corrected to a dry weight basis, the corrected averages weighted for number of samples, and a weighted average for leafy vegetables of 750 pCi/kg was computed.

Greens, the leafy crop showing the highest ^{137}Cs levels in that region and probably most comparable in exposed leaf surface to animal forage, have an average ^{137}Cs content on a dry matter basis of 875 pCi/kg. The individual samples having the highest levels were also corrected to a dry weight basis and these results are tabulated in Table 20.

TABLE 20

FLORIDA VEGETABLE SAMPLES WITH HIGHEST CESIUM-137 LEVELS
DRY WEIGHT BASIS

Sample	Station	Vegetable	pCi/kg (Wet Wt.)	% Water	% Dry Matter	pCi/kg (Dry Wt.)
15	C-10	Cucumber	561	94.0	6.0	9,350
166	SW-8	Tomato	357	93.5	6.5	5,492
140	C-10	Lettuce	356	94.0	6.0	5,933
139	C-10	Lettuce	316	94.0	6.0	5,267
152	SW-5	Tomato	310	93.5	6.5	4,769
154	SW-5	Tomato	305	93.5	6.5	4,692
138	C-10	Cabbage	308	92.4	7.6	4,053
197	SW-9	Potatoes	294	79.8	20.2	1,455

The average values corrected this way are about an order of magnitude lower than Porter's 1963-1964 results for pangola hay, about one-fifth of the 1966 Tampa pangola hay, and about one-half of some of the other reported levels in grass. The highest vegetable results are greater than the highest pangola hay results, but based on a larger number of samples. It appears that the average vegetable levels, expressed on a dry weight basis, are lower than the results reported in the literature for pangola hay from the Tampa area, but that maximum values in vegetables are probably comparable to maximum values in grasses and other animal forage. Some of the difference between vegetables and animal forage can be attributed to the fact that the vegetable samples were all washed before analysis while forage is analyzed on the same basis as it is fed to the animal.

Influence of Observed Cesium-137 Levels on Human Intake and Exposure

The ultimate criterion for evaluation of the significance of observed environmental radiation or radioactivity levels is the effect of these levels on human exposure.

The results of the analysis of Florida beef support the conclusion of Ward and Johnson that human intake (and thus body burdens) would be related to the type of beef consumed.[51] Since the higher levels were in meat from the lower grade carcasses which is made largely into ground beef and processed meats such as bologna and frankfurters, the results also support the suggestion of these authors that ^{137}Cs body burdens resulting from beef consumption may be related to the economic status of the family.

The total radionuclide intake per unit time from all sources, I, may be expressed in terms of individual dietary components (air and water are negligible contributors) in the following manner:

$$I = \sum_{i} w_i \; C_i$$

where C_i = concentration per unit weight of the ith dietary item,

and w_i = weight of the ith dietary item consumed per unit time.

In preparing radionuclide intake evaluation of this data, food categories and estimated average daily intakes for adults in the United States were adapted from the reports of the Tri-City diet sampling program.[150] Quantity intake values in that program are based on the

United States Department of Agriculture values from the 1955 Household Food Survey. For comparison purposes, two intake estimations have been derived from published values and are presented in Table 21. The first was obtained by averaging all the 1965 quarterly data reported for the three cities in the Tri-City program, and the second from extensions to 1966-1967 of Gustafson's extrapolations of Chicago data.[150,65]

TABLE 21

CESIUM-137 INTAKE ESTIMATED FROM PUBLISHED VALUES

Food Category	kg/day	1965 Tri-City Average pCi/kg	1965 Tri-City Average pCi/day	1965 Tri-City Average % of Total	Chicago Values Extrapolated to 1966-1967 pCi/day	Chicago Values Extrapolated to 1966-1967 % of Total
Milk	.605	62.5	37.8	32.1	15	19.5
Meat	.315	108.6	34.2	29.1	20	26.0
Grain	.266	95.9	25.5	21.7	30	39.0
Vegetables	.350	25.1	8.8	7.4	3	3.8
Fruit	.310	36.8	11.4	9.7	9	11.7
Total	1.846		117.6	100.0	77	100.0

In evaluating the Florida data from this study, the assumption was made that the food consumption values used in reporting Tri-City data were applicable to Florida consumers and that, because of production, marketing, milling, and distribution patterns, grain products contain essentially the same average levels of ^{137}Cs wherever consumed throughout the United States.

Intake estimates were prepared on the basis of levels observed in the regions designated as Central and Southwest Florida, the two regions showing the highest ^{137}Cs levels in all media. Intakes due to locally grown foods in the northwestern part of the State should be the lowest in the State, and, since ^{137}Cs levels in that part of the State are essentially the same as expected for the nation on the average, the Northwest average should be similar to national averages. Intakes in regions showing intermediate levels of radioactivity should have correspondingly intermediate values.

The amount of locally grown food consumed will vary from family-to-family and probably varies with socio-economic status and with the isolation of the place of residence. For the purpose of this evaluation several hypothetical cases are considered and presented in Table 22.

The individual in Case I has cosmopolitan eating habits; milk and dairy products are locally produced, but the other components consist of such commodities as western beef and packaged frozen fruits and vegetables, and are represented by national averages.

The "average man" in hypothetical Case II consumes local dairy products, and the higher grades of locally fed meat and receives about half of his fruit and vegetable intake from local sources.

The individual in Case III has the greatest local ties, consumes local dairy products, and meats or game with levels corresponding to the average values found for grass-fed animals and receives all his fruits and vegetables from local sources.

In Table 21, it was seen that for the nation in 1965, milk, meat, and grain products contributed approximately equally to ^{137}Cs intake, and fruits and vegetables combined made up less than 20 per cent of the total intake. The Chicago values, extrapolated to 1967, show all levels decreasing with milk and fruits and vegetables decreasing in relative

TABLE 22

CESIUM-137 INTAKE ESTIMATE FOR THREE FLORIDA CASES
1966-1967

Food Category	kg/day	Case I pCi/kg	Case I pCi/day	Case I % of Total	Case II pCi/day	Case II pCi/day	Case II % of Total	Case III pCi/kg	Case III pCi/day	Case III % of Total
Milk	.605	135	82	56.9	135	82	35.0	135	82	5.7
Meat	.315	a	20	13.9	250	80	34.2	4,000	1,260	87.8
Grain	.266	a	30	20.8	a	30	12.8	a	30	2.1
Vegetables	.350	a	3	2.1	94	33	14.1	94	339	2.3
Fruit	.310	a	9	6.3	a	9	3.8	94	301	2.1
Total	1.846	--	144	100.0		234	100.0	--	1,435	100.0

a = Florida daily intake for this category assumed to be same as Chicago value extrapolated to 1966-1967.

importance. By comparison, because of the levels in Florida milk, the major source of ^{137}Cs intake is milk for the Case I individual. His intake is about 122 per cent of the national estimation for 1965 and about 187 per cent of the Chicago value extrapolated to 1967.

For Case II, milk and meat contribute about equally and together contribute about 70 per cent of the intake while fruits and vegetables slightly exceed grain products in contributing to the balance. The total intake is about two times and three times, respectively, that estimated for 1965 and that extrapolated to 1967.

In Case III, meat contributes nearly 90 per cent of the ^{137}Cs intake and total intake of the nuclide is greater than 10 times the 1965 national estimate and nearly 20 times the projected 1967 estimate.

The estimated total intakes all fall below the value of 4,400 pCi/day recommended by the Federal Radiation Council as a guide for the population at large, although the Case II estimate approaches 5 per cent, and the Case III estimate is slightly more than 30 per cent of this value. This guide could be exceeded by groups of people consuming foods, particularly meats, with the extreme values reported.

Of the number of methods of estimating body burden and total-body dose rate, the simplest method is to assume an equilibrium condition in which intake levels have remained constant for a sufficiently long period of time so that losses from the body are exactly equal to intake. This approach produces low estimates in times of decreasing intake levels and high estimates in times of increasing levels. In this method, when ^{137}Cs losses are described by a single exponential function, body burden, Q, may be estimated:

$$Q = I\,A\,\frac{1}{\lambda} = I\,A\,\frac{T_{eff}}{0.693} = 1.44\,I\,A\,T_{eff}$$

where I = intake as previously defined,

A = fraction of intake absorbed,

λ = elimination constant, and

T_{eff} = the effective half-life, $0.693/\lambda$.

For ^{137}Cs, the physical half-life is so much longer than the biological half-life that the effective half-life may be taken as equal to the biological half-life and absorption is nearly 100 per cent, so that:

$$Q = 1.44\,I\,T_b.$$

For an assumed value of T_b = 100 days:

$$Q = 144\,I.$$

Body burdens predicted in this manner are shown in Table 23 along with values from the literature. It can be seen that the body burden for Case I is slightly more than estimated for the entire United States in 1965 and measured in Chicago residents at the beginning of the year, and it is about twice that estimated from the projected intake for 1967. The Case II value, directly reflecting a higher estimate of radionuclide intake, is about 50 per cent higher. The Case III value is greater by an amount proportional to the higher estimated intake and is comparable to those measured during April-May, 1966, in Alaskan men eating caribou or reindeer several times a week.[75] The estimated body burdens, even in Case III, are below the Federal Radiation Council's Radiation Protection Guide of 3,000 nanocuries (nCi) for whole body exposure of individuals in large population groups (1,000 nCi when estimation is made for a suitable sample of the group rather than measuring individuals).[62]

TABLE 23

REPORTED BODY BURDENS OF CESIUM-137

Location	Condition	Source	Date	nCi Total Body	Range
Central Florida	Case I	This study—estimation from food items	1966 - 1967	21	
	Case II			34	
	Case III			210	
Chicago		Whole-body counting summarized by Rivera[150]	Start 1965	14	
New York City			End 1965	15	
United States		Estimation from diet levels, Tri-City Program	1965	17	
Chicago		Estimation from extension of Gustafson extrapolation of diet levels[65]	1966 - 1967	11	
Alaska	Caribou Consumption:	Fitzpatrick et al.—whole-body counting[75]	April - May, 1966		
	Daily (66 men)			780	ND-2,400
	Less than daily (93 men)			190	ND-1,100
	Weekly (29 men)			80	ND- 270
	Monthly (35 men)			60	ND- 290
	None (195 men)			15	ND- 150
	All (418 Men)			180	ND-2,400

ND = not detected = less than 40 nCi

Dose rate may be computed for the equilibrium case:

$$R = \frac{11\ Q}{W}$$

where R = dose rate, rads/year,

Q = body burden, (μCi) as previously derived,

W = body weight, kg,

and 11 = a conversion factor, kg-rads/μCi-year,

when the average energy absorption in the body is assumed to be 0.59 MeV/disintegration.

The expression for intake may be incorporated:

$$R = 11\ \frac{I}{W} \times 1.44\ T_b$$

or, in the case of the 70 kg standard man and T_b = 100 days,

$$R = 22.6\ I.$$

For Cases I, II, and III, the dose rate from ^{137}Cs computed in this fashion would be 3.25, 5.3, and 32 millirads/year, respectively. These contributions to the total dose rate may be compared to the Radiation Protection Guide of 500 mrem in a year to the whole body of individuals in the general population or an average of 170 mrem when measuring a suitable sample of the group rather than individuals.[62]

Because of the uncertainties from region-to-region, family-to-family, and person-to-person in dietary habits, food preferences, and quantities of intake, the differences between adults and children, and the generalized nature of many of the assumptions made, the estimates of ^{137}Cs intake, body burdens, and dose rate in this chapter can only be considered as crude indications of the possible significance of some of the radioactivity levels observed.

Possible Mechanisms and Factors Influencing Cesium-137 Levels in Florida

No evidence can be found to indicate that the ^{137}Cs currently found in the Florida environment has any source other than past or current fallout deposition from nuclear testing.

According to the literature, environmental levels of ^{137}Cs are generally independent of total accumulated deposition because this nuclide is bound so tenaciously to the soil. It is recognized that uptake can be related to a certain extent to accumulated deposition over the immediately preceding one or two years because ^{137}Cs can remain available in the root mat and in organic material for a time after deposition. However, uptake is usually considered to be primarily due to direct deposition and thus the levels are considered to be primarily related to the current rate of deposition.

According to the generalized theory of latitudinal distribution of fallout deposition, Florida should lie considerably south of the northern hemisphere maximum (approximately 40°) and in a region of rapidly diminishing levels from north to south.[34,37] Strontium-90 levels in soil samples collected from sites throughout the country, including one each in North and South Florida, support this theory.

This generalized pattern is modified by local factors. Maritime exposure has been raised and then discounted as one of the possible modifying factors.[37,196]

Precipitation is generally believed to be a significant local modifier of distribution patterns, with high precipitation areas having increased deposition rates and accumulated deposition. If radioactivity levels are highly rate dependent, amount of rainfall during the particular growing season of the vegetation fed or sampled could be highly significant. In addition, Ward and Johnson report immediate response in vegetation and milk to variations in amount of ^{137}Cs deposited by precipitation.[48,197]

No direct studies have been performed yet in Florida to determine the effect of either average rainfall, growing season rainfall, or individual rains on environmental ^{137}Cs levels. Examination of the mean annual precipitation contours in Florida for the period 1931-1955 shows that the highest annual rainfall, greater than 56 inches, occurs in much of what was designated in this study as the southeast and northwest regions.[198] Most of what were designated as southwest, central, and northeast regions lie below the 56 inch contours. Exceptions to this general pattern are a small region of about 56 inches and greater north of Tampa (Brooksville area) and a sharp gradation from 56 inches to 48 inches and less south and east of Miami and Homestead. It appears that the 1966-1967 levels of ^{137}Cs in Florida vegetables show an inverse relationship to the average annual precipitation.

Only limited significance can be placed on the above observation, however, because precipitation can be highly localized and can vary considerably from year-to-year. No consideration was given to actual precipitation at the particular growing site during the whole growing period for the particular samples collected or during the period immediately preceding sampling. Examination of local rainfall records for late 1966 and early 1967, as they become available, should be particularly interesting.

Studies of ^{137}Cs in vegetables in 1960-1963 showed considerably more radioactivity in leafy vegetables than in other types, but no such difference could be demonstrated in this study. This might be due to surface contamination being more thoroughly washed off in sample preparation. On the other hand, the leaf activity found in earlier studies could have been due to insoluble contamination from foliar deposition tightly retained on the surface of the leaf and foliar deposition absorbed by the leaf, but not effectively translocated throughout the plant. If it is assumed that ^{137}Cs deposited in 1966-1967 would be no more soluble than that deposited earlier, the absence of selectively high levels in leaf crops in 1966-1967 suggests that in these later years, ^{137}Cs is being distributed throughout the plant and thus must have come from some source other than foliar deposition. Coupled with the soil uptake by grasses demonstrated by Cromroy _et al._, this is strong evidence for the role of a cumulative dependent mechanism and uptake from the soil in Florida ^{137}Cs levels.[111]

Unusual environmental factors must be involved in the ^{137}Cs levels found in Florida, although it has not been determined whether these involve unusual deposition, unusual uptake by biota from available ^{137}Cs, or unusual availability of the accumulated deposit. Rates of ^{137}Cs deposition in various parts of the State have not been evaluated, but it does not seem likely that deposition can account for the variations and levels seen. Neither does it seem likely that an unusual uptake would be an inherent characteristic of all of the wide variety of plant species in which unusual levels of this nuclide have been noted. This leaves, as the most likely explanation, some unusual environmental factor or mechanism accentuating the uptake from currently deposited ^{137}Cs or making the accumulative deposit in the soil more available for uptake. Climatic factors, physiographic features, cultivation practices, and soil practices are some of the factors which might play a role in unusual ^{137}Cs uptake.

The existence of factors resulting in unusual biospheric accumulation of ^{137}Cs has important implications in waste disposal, operation of nuclear facilities, and hazard evaluation, and this situation still deserves considerable exploration.

Other Radionuclides

The primary emphasis in this study was on ^{137}Cs and only limited attention was given to analysis and interpretation of other nuclide results.

The presence of radium daughter activity in such a large proportion of the samples analyzed suggests that radium may be a significant source of environmental radiation exposure. Except for measurements of radon daughters in air and gross alpha activity in a variety of other media, radium and its daughters have not been studied to any extent in Florida.

Nuclide activities, in addition to ^{137}Cs, ^{40}K, and radium, which occurred in a sufficient number of samples to merit consideration were primarily fresh fission products. The fresh fission-product activity in the May 21-26, 1966, samples indicated that Florida received deposition from the Chinese atmospheric nuclear detonation of May 9, 1966. This is consistent with the report of Grundy and Snavely which shows an intrusion over Florida of the fresh fission-product debris cloud about May 17-19, 1966.[166]

The fresh fission-product activity in January and February of 1967 can be attributed to the Chinese atmospheric nuclear test of December 28, 1966.[199]

The activity found in vegetables at this time appeared to be surface contamination due entirely to direct deposition on exposed leafy surfaces. The activity appeared only in the very leafy vegetables--lettuce and greens. Very little activity was found in the celery samples which consistented mainly of stalks after about one-half of the leafy top was removed in sample preparation. No activity was detected in cabbage from which the outer leaves were discarded before analysis.

The activity disappeared from the vegetables at a more rapid rate than would be indicated by the half-lives of some of the nuclides and it never appeared in other than leafy samples. It appears that very little of these nuclides was absorbed from the surface contamination and translocated. This distribution pattern is in contrast to those exhibited by ^{137}Cs and ^{226}Ra and daughters, nuclides which appeared in all vegetable types and appeared to have been widely translocated in the plant.

The statewide occurrence of these fresh fission-products is also in contrast to the occurrence of ^{137}Cs which had a definite geographic pattern.

Discussion of Sources of Error

Most computational procedures for resolving complex gamma spectra into individual components, including the simultaneous equations method used here, require an a priori assumption of the composition of the mixture. This results in two inherent weaknesses. If a nuclide is present that is not in the calibration matrix, an error can result. The magnitude of this error for the various nuclides reported depends upon the particular spectrum and the amount present of the omitted nuclide. The second weakness is related to the effect of including nuclides in the calibration mixture which are not in the sample spectrum. This results in an increase in the standard deviation of the level reported for each of the nuclides which are present.

During the time that this study was carried out, several nuclear weapons tests were conducted in the atmosphere, resulting in the transient appearance of short-lived nuclides in the environment. The very short-lived nuclides (half-lives less than seven days) were not included in the calibration matrix. However, this should not have introduced a serious error since all of the 1966 and most of the 1967 samples were counted a sufficiently long time after the arrival of fresh fallout to eliminate or at least greatly reduce the contribution of these components.

It should be noted that the cerium calibrations were made with an equilibrium mixture of ^{144}Ce/Pr (half-life of 285 days) and ruthenium calibrations with one of ^{106}Ru/Rh (1.0 years). The presence of ^{141}Ce (32.5 days) and ruthenium-103 (40.0 days), with a gamma spectra similar but not identical to those of their respective long-lived fission product isotopes, probably contributed some error to the results reported for samples contaminated with recent fallout.

Although ^{65}Zn has been reported in foodstuffs in the past, it was not included in the calibration matrix and the unrecognized presence of this nuclide may have been another source of error.

Only limited significance can be placed on the reported quantitative values for ^{226}Ra and its daughters. They were not necessarily in equilibrium, mainly because no effort was made to prevent the loss of the gaseous intermediate, radon-22 (^{222}Rn). This nuclide chain was included in the calibration matrix primarily to correct for interferences with other nuclides, notably interference with the 0.66 Mev ^{137}Cs peak by the 0.61 MeV bismuth-214 (^{214}Bi) peak and with the 0.36 Mev ^{131}I peak by the

0.35 MeV lead-214 (^{214}Pb) peak. For the estimation of radium daughters, the 1.12 MeV ^{214}Bi peak was selected as being the most abundant peak not directly interferring with another major photopeak. Nevertheless, the low abundance of this peak relative to the other two peaks just mentioned, contributes to the uncertainty in the estimation of ^{137}Cs and ^{131}I. In addition, the unrecognized presence of ^{65}Zn (1.11 MeV) which was not included in the calibration matrix, would result in overestimation of radium daughter interferences.

Although cesium-134 (^{134}Cs) (0.60 and 0.80 MeV) has been reported to be present in environmental samples to the extent of 1.0 to 1.5 per cent of the ^{137}Cs, it was not included in the calibration matrix. This may have introduced some errors, particularly in the reporting of, and correction for interference from, ^{54}Mn (0.84 MeV). However, because of the half-life of only two years, ^{134}Cs levels should have been well below 1 per cent of the ^{137}Cs levels at the time the samples in this study were analyzed.

Any positron emitters present in the sample would have produced a 0.51 MeV peak in the spectra which would have been interpreted as $^{106}Ru/Rh$, thus resulting in overestimation of $^{106}Ru/Rh$ and consequently in underestimation of ^{137}Cs because of over-correction for interference of the 0.61 MeV $^{106}Ru/Rh$ peak. Other nuclides not included in the calibration matrix were not believed to be present in sufficient quantities to seriously affect the accuracy of the results reported.

The analysis of the geographic variation of levels in lean beef might be criticized on the grounds that while the samples from the four eastern-most stations were all the same cut of meat and collected from experiment stations animals, the samples at Station 5 were unidentified cuts from animals on undocumented feeding programs. With regard to the cut of meat, other investigators have reported that variance due to the particular muscle sampled is not significant relative to the animal-to-animal variation and variation introduced by counting statistics.[51] With regard to the feeding program, it is possible that some or all of the animals from Station 5 were finished on diets that contained a higher pasture to grain ratio than contained in the experiment station diets. However, at a different station in this study, increasing the forage to grain ratio in the animals' diet markedly increased the ^{137}Cs content of the meat.

The influence of ^{137}Cs contribution to the environment by Chinese weapons testing since 1964 was not considered in extrapolating literature values to 1966-1967 for comparison to the values obtained in this study. It was assumed that this contribution was not significant in relation to the other uncertainties in such an extrapolation.

Further Investigations Suggested by the Results of This Work

The results of this study suggest further work in at least four areas, (1) assessment of human exposure to ^{137}Cs, (2) study of environmental factors contributing to ^{137}Cs levels, (3) studies of natural radioactivity in Florida, and (4) derivation of parameters for conducting monitoring and surveys.

Assessment of human exposure

Further investigation of Florida meat and meat products is indicated. Such work should be accompanied by a determination of the origin and quantities produced and consumed of various meat grades and meat products, information probably available from published literature. Further surveys are needed of levels of ^{137}Cs in products sold over the counter, particularly ground beef and processed meats such as frankfurters and bologna, to determine the extent to which the higher levels observed in the lower quality carcasses are reflected in these products. Corollary work would be the investigation, on a wider basis, of the relative levels of ^{137}Cs in feed-lot and range cattle in Florida.

In view of some of the predictions of this study, a second indicated area of human exposure assessment is the investigation of human body burdens of ^{137}Cs--both the variation within the State and a comparison with other reported values. Data could be obtained from three sources--other installations that have counted Florida persons, retrieval of ^{137}Cs content from whole-body counts already performed at the University of Florida, and a deliberate study of ^{137}Cs levels in Florida individuals.

Several commodities neglected in this study also deserve further consideration. Since fresh water fish have been shown to have high ^{137}Cs levels in other parts of the country and according to Gustafson these levels can significantly affect intake of this nuclide by individuals of groups substituting fish for a considerable portion of the meat in the standard diet, this medium deserves further study in Florida.[54,55]

Citrus was not included in this study but it deserves further consideration because it is a very economically important crop in Florida.

As pointed out by several authors, there is also a need for further work in developing methods of predicting human exposure on the basis of environmental measurements.[65,150] It appears that levels of ^{137}Cs are sufficiently high in certain segments of the Florida environment to make such studies under natural conditions more feasible here than in many other parts of the country.

Environmental mechanisms

Although various investigators have studied the Florida environments, the particular environmental factors responsible for the observed ^{137}Cs levels still have not been identified. It still is necessary to determine what portion of the observed levels can be attributed to deposition rate and what portion to accumulated deposition. If accumulated deposition makes a significant contribution, the environmental mechanisms making the Florida situation so unique should be described.

One question is the magnitude and distribution of ^{137}Cs deposition in Florida. This might be evaluated by examining past and current data of the USAEC and Florida State Board of Health sampling stations and perhaps by conducting prospective deposition sampling at one or more locations. As an alternative, distribution might be estimated from surface air concentrations since air sampling continues at the various stations of the Florida Radiation Surveillance Network and filters are available from the past several years.

Another aspect of ^{137}Cs deposition is the question of whether or not $^{137}Cs/^{90}Sr$ ratios have been constant in deposition or whether they have varied geographically as they do in milk. Information might be retrieved from past and future data from the USAEC deposition sampling program and deposition sampling of the Florida Radiation Surveillance Network augmented by additional or modified stations if necessary. Alternatively, the ratios might be estimated from surface air samples or from deposition as estimated by statewide Spanish moss sampling. Precipitation residues from 1955-1966 and air sampling filters from the Florida Radiation Surveillance Network have been turned over to the University of Florida and a number of statewide Spanish moss samples collected during 1966 are being held on hand.

The relative importance of rate dependence and cumulative dependence might be explored by fitting models to existing data, particularly the milk data that has been collected monthly for a number of years. It might also be explored by examining the behavior of the $^{137}Cs/^{90}Sr$ ratio with time and with changes in the deposition rate, thus determining whether the two nuclides have proportional or different dependence on deposition rate.

The work of Cromroy _et al_. strongly suggests that uptake from the soil may be an important factor in ^{137}Cs levels in Florida vegetation, and this is a promising area for further investigation.[111] Florida soils, particularly from areas where high vegetable radioactivity levels were observed, can be identified either by soil type or geographic location and submitted to three tests, total ^{137}Cs content (indicative of accumulative deposition), "exchangeable" or "available" ^{137}Cs content and exchange and fixing capacities. The available ^{137}Cs content and capacity might be measured by either "test tube" or "greenhouse" methods or both. The influence of modifying factors such as soil pH and soil moisture content might be examined at the same time.

Since general location of the source is available for all samples analyzed in this study and specific farm or field can be determined for many of them, further investigation also might be made of factors relating to the levels observed in specific samples. These factors might include rainfall during the entire growing season preceding sampling, rainfall in the period immediately preceding sampling, average rainfall for the area, soil type, soil pH, and fertilizing and other farming practices.

Finally, a controlled study might be performed to evaluate radionuclide balances and rates of transfer, and ultimate transfer to meat in feed lot and pasture situations.

Studies of natural radioactivity in Florida

The presence of ^{214}Bi and ^{214}Pb in so many of the gamma spectra for samples in this study and other environmental radioactivity studies currently being performed at the University of Florida suggests that higher than average levels of natural radioactivity are appearing in Florida environmental media. The vegetable and beef samples currently on hand might be analyzed for radium and other natural radioactivities and the results compared to available data to determine the importance of further natural radioactivity studies. If further study appears justified, the influence of calcium levels in Florida soils and agricultural liming practices might be important factors to explore.

Parameters for conducting monitoring and surveys

As the number of radioactivity handling or producing installations in the southeastern states grows, surveys and monitoring programs will be carried out to determine preoperational radioactivity levels and to determine influence of the operations on the environment. In addition, surveys will continue to be made to assess overall radiation exposure levels. This study has shown that wide variation is present in at least one nuclide in the Florida environment and has suggested that the frequency distribution of these levels is not normal. From the data already collected it is possible to identify important sources of variance, to derive estimates of variance for particular populations, to estimate sample sizes needed to make certain decisions and to optimize the distribution of effort between replicate samples, replicate analyses per sample and minutes of counting time per analysis. Hawthorne has reported high variability in the levels of radionuclides in various components of an agricultural system and he pointed out that in order to develop a model to describe such a system, these variations must be taken into account and the model must be constructed from actual observations upon the system over some period of time rather than from assumptions.[200]

CHAPTER VIII

SUMMARY AND CONCLUSIONS

This research involved a study of the kinds, levels, and distribution of gamma-emitting radionuclides in the Florida environment and the evaluation of these levels in terms of human exposure. In sampling and analysis, emphasis was placed on ^{137}Cs in beef and vegetables. Preliminary sampling was carried out in May and June, 1966, and full scale sampling was conducted during January through July, 1967. Analyses were performed by gamma spectroscopy on triturated samples without ashing and without chemical preparation or separation. Complex gamma spectra were interpreted in terms of the individual contributing components by use of the simultaneous equations method.

Levels of ^{137}Cs in both beef and vegetables showed geographical patterns similar to those reported earlier by others in Florida milk. Maximum levels were found in the central and southern parts of the State, with diminishing levels occurring in the northeastern part of the State and the very lowest levels occurring in the northwestern part of the State. The animal products did not show variations as great from region-to-region as did the vegetables, probably due to the averaging effect of movement of feed from region-to-region and into the State from outside. Vegetable levels differed from beef and milk, in that the levels found in the southeastern part of the State were as low as in the northwestern part while average beef and milk levels in this vicinity were only slightly less than seen in the central and southwestern parts. Meat levels did not decrease as rapidly from east to west in the northern part of the State as did milk and vegetable levels.

In contrast to the results of earlier work by others who sampled throughout the United States, there was no apparent difference in ^{137}Cs concentrations between leafy, fruit, and root categories of vegetables. Certain crops in each category did exhibit consistently higher levels than other crops in the same category.

It was not possible, with the number of samples collected, to demonstrate any clear-cut difference between the 1966 and 1967 sampling years for either beef or vegetables. Possible seasonal effects during the sampling period were not taken into consideration.

It was difficult to find radioactivity concentration values in the literature against which to compare values found in this study. There is a two- to three-year lag time in publishing data and though radioactivity levels in most environmental media have been decreasing since 1963 or 1964, they have varied so much with time and location that extrapolation is difficult.

The statewide average of ^{137}Cs levels in fattened beef was 214 picocuries per kilogram (pCi/kg), and the range of individual samples was 55-539 pCi/kg. This average is comparable to average national levels reported for beef in 1965. Levels in samples from the western part of the State averaged 105 pCi/kg and ranged from 55 to 174 pCi/kg. This average is comparable to 1964-1965 Chicago meat values extrapolated to 1966-1967. The levels in samples from the stations in the balance of the State were two to three times this extrapolated value.

Levels of ^{137}Cs in a group of samples representing lower quality meat from animals which presumably had fed on grass averaged about

4,000 pCi/kg and ranged from about 300 to over 12,000 pCi/kg. These values are much higher than those in high quality meat from feed-lot animals from the same general vicinity and are higher than any other reported beef levels in the country. The highest values approach levels reported for reindeer and caribou in the arctic.

Cesium-137 levels in vegetables from southeastern and northwestern parts of the State averaged about 25 pCi/kg and ranged from below detectable limits to 176 pCi/kg. This average level is comparable to national values extrapolated to 1967, but averages from several regions were considerably higher. The average levels in the northeastern part of the State were about 50 pCi/kg with a range from below detectable limits to about 200 pCi/kg, and those in the central and southwestern parts were on the order of 90-100 pCi/kg with a range from below detectable limits to 560 pCi/kg.

Average vegetable levels, expressed on a dry weight basis, were lower than values reported in recent years by others for pangola hay in Florida, but maximum vegetable values were higher than the highest values reported for hay.

Cesium-137 intakes were estimated for three hypothetical cases of assumed diet composition and source of food. It was estimated that radioactivity intakes from locally produced food in Northwestern Florida would be similar to the national average. On the other hand, in the remainder of the State, Florida milk alone would double the intake of ^{137}Cs and thus the body burden of this nuclide under long-term exposure conditions. It was estimated that if locally grown fattened beef was substituted for other meats in the diet in the central and southwestern parts of the State, radioactivity intake and body burdens would rise to three times the estimated national average. When it was assumed, for the same part of the State, that meat in the diet was at the average level found for grass-fed beef and that all fruits and vegetables consumed were at the average levels found for vegetables, ^{137}Cs intake and equilibrium body burdens were estimated to be about 20 times the projected national average. The intakes, body burdens, and resulting whole-body radiation doses estimated for each of these cases were less than the respective radiation protection guides, but the guides could be approached by individuals consistently consuming food at the extreme levels found.

The mechanisms responsible for the unusual patterns of ^{137}Cs levels in Florida were not identified but there is increasing evidence that, contrary to what is usually stated for ^{137}Cs, uptake from the soil plays a significant role.

A number of further studies appear to be indicated, particularly of the levels in, and possible radionuclide intakes due to, processed meat products made from the lower grades of meat, of the factors involved in ^{137}Cs levels in meat from grass-fed beef in Florida, of ^{137}Cs body burdens of persons living in Florida and the relationships of these burdens to eating habits and food sources, and of the role of soil as a source of biospheric ^{137}Cs in Florida.

Conclusions

1. The relatively high levels of ^{137}Cs reported for Florida milk are also reflected in other environmental media.

2. The geographical patterns of variation of ^{137}Cs levels seen within the State for milk are also seen (with certain modifications) for beef and vegetables, with highest levels in the central and southwestern parts of Florida.

3. Levels of ^{137}Cs in beef from grass-fed animals in Florida can be much higher than those in grain-fed animals.

4. Although varying considerably with dietary habits, intake of ^{137}Cs by humans and corresponding body burdens in a considerable portion of Florida should be significantly higher, on the average, than those for the nation as a whole.

5. Average body burdens of ^{137}Cs for groups of central Florida residents can be expected to range between 20 and 200 nCi, depending on type and source of food.

6. None of the average intakes, body burdens, or resulting total-body radiation doses estimated from ^{137}Cs levels observed in this study exceed the applicable radiation protection guides.

7. The elevated levels of ^{137}Cs in the central and southern parts of Florida appear to be due to unusual environmental factors or mechanisms influencing the uptake by vegetation from current deposition of or accumulative deposits of this nuclide. These factors and mechanisms can have important implications with regard to waste disposal, site and hazard evaluation, and operation of nuclear facilities.

8. There is increasing evidence for the importance of uptake from the soil as one of the factors in the ^{137}Cs levels observed in Florida.

9. A number of further studies are indicated, particularly of ^{137}Cs in Florida meat products, of human body burdens of ^{137}Cs in Florida, and of the soil and other factors involved in the observed levels and patterns.

APPENDICES

APPENDIX A

EQUIPMENT AND INSTRUMENTATION

TABLE 24

EQUIPMENT AND INSTRUMENTATION

Item	Purpose	Model	Manufacturer	Description or Comments
		Sample Preparation Equipment		
Food cutter	Prechopping vegetable samples	84141	Hobart	Used on one occasion only
Food slicer	Prechopping vegetable samples	4612 (with slicer attachment)	Hobart	Used on several occasions only
Blender	Sample homogenization	8000	Waring Products Division, Dynamics Corporation of America, New York	1 gallon size
Meat grinder	Meat and vegetable grinding		Enterprise	
		Radioactivity Analysis		
Marinelli beaker	Sample counting container	F-26862 F-26860	Bel-Art Plastics, Pequonic, New Jersey	Configuration 1 - 3.5 liter 2 - 1.0 liter
Multichannel analyzer system detector	Radiation detection	Matched window line	Harshaw Chemical, Cleveland, Ohio	4-inch x 4-inch right cylindrical NaI(Tl) crystal phototube
Shield	Reduce background		University of Florida	20 x 20 x 24 inches high inside dimensions; 2-inch thick lead sides, floor and cover; graded cadmium and copper lining

TABLE 24 (continued)

Item	Purpose	Model	Manufacturer	Description or Comments
Emitter follower	Drive signal cable		University of Florida	
Pulse height analyzer	Signal analysis	116	Packard Instruments, Inc., Downers Grove, Ill.	400 channel, self-contained linear amplifier and detector high voltage supply
High speed parallel printer	Parallel digital output	Monroe data/log	Litton Industries, San Francisco, California	
Parallel serial converter	Provide serial digital output	70	Packard Instruments, Inc.	
Output writer (typewriter)	Serial digital output	IBM 11C (Packard 91)	International Business Machines, Inc., New York	
Spectrum reducer	Temporary external storage	50	Packard Instruments, Inc.	Storage and read back from magnetic tape cartridge
X-Y plotter	Analog output	7590A (Packard 93)	F. L. Moseley Company Pasadena, California	X-Y plotter with pen and null detector and character printer

APPENDIX B

GAMMA-EMITTING RADIONUCLIDES INCLUDED IN THE CALIBRATION MATRIX

TABLE 25

GAMMA-EMITTING RADIONUCLIDES INCLUDED IN THE CALIBRATION MATRIX

Nuclide	Half-Life	Principal Gamma Energy MeV	Spectral Region Used for Computation MeV	Spectral Region Used for Computation Efficiency Counts/Disintegration
Cerium/Praseodynium-144	285 days	.08	.065-.165	.0254
		.134		
		.69		
Iodine-131	8.05 days	.364	.315-.405	.0616
		.64		
Ruthenium/Rhodium-106	1.0 years*	.513	.445-.545	.0121
		.612		
		.624		
Cesium-137	30.4 years	.662	.545-.715	.0397
Zirconium/Niobium-95	65 days/35 days	.76	.675-.825	.0344
Manganese-54	314 days	.835	.775-.895	.0333
Radium-226 and daughters	1,620 years*	.187		
		.242		
		.295		
		.352		
		.609		
		.769		
		1.12	1.045-1,205	.0070
		1.76		
		2.20		
Potassium-40	10^9 years	1.46	1.345-1.555	4.6283**
Barium/Lanthanum-140	12.8 days*	.16		
		.32		
		.506		
		.815		
		1.60	1.495-1.705	.0195

*Daughter half-life is sufficiently short so that equilibrium can become established in a sample held for a short period of time.

**Counts/gram.

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BIOGRAPHICAL SKETCH

Charles Ervin Roessler was born May 1, 1934, in Elysian, Minnesota, where he attended elementary schools. He attended high school at Waterville, Minnesota, and received his diploma in 1951. He received his Bachelor of Arts degree from Mankato State College, Mankato, Minnesota, in 1955 with majors in biology and chemistry and a minor in mathematics.

He attended the University of Rochester on a United States Atomic Energy Commission Fellowship and received a Master of Science degree in radiation biology there in June, 1956. He spent the summer months of 1956 at Brookhaven Laboratory, Long Island, New York, on an Atomic Energy Commission training program. From September, 1956, to September, 1958, he worked as a health physicist with the Nuclear Power Division of Curtiss-Wright Corporation at Quehanna, Pennsylvania.

He returned to graduate work with an Atomic Energy Commission Fellowship at the University of Pittsburgh in September, 1958, and received a Master of Public Health there in June, 1959. He was employed as a radiological physicist at the Florida State Board of Health from July, 1959, to January, 1965, with supervisory responsibilities for radiological health in the Division of Radiological and Occupational Health. He began his graduate study at the University of Florida under a United States Public Health Service Fellowship in January, 1965.

He is married to the former Genevieve Schleret of Owatonna, Minnesota, and they are the parents of six children.

He is a charter member of the Health Physics Society and a member of the executive council of the Florida Chapter, Health Physics Society. He is also a member of the Conference on Radiological Health, the American Industrial Hygiene Association, the Florida Section of the American Industrial Hygiene Association, the American Conference of Governmental Industrial Hygienists, and the Air Pollution Control Association.

He was certified in Health Physics by the American Board of Health Physics in 1961, was elected to associate membership in the Society of the Sigma Xi in 1959, and was elected to Who's Who Among Students from American Colleges and Universities in 1954-1955.

This dissertation was prepared under the direction of the chairman of the candidate's supervisory committee and has been approved by all members of that committee. It was submitted to the Dean of the College of Engineering and to the Graduate Council and was approved as partial fulfillment of the requirements for the degree of Doctor of Philosophy.

December 19, 1967

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