[redacted]

[redacted]
No. 15 [redacted]

405674

US DOE ARCHIVES
326 U.S. ATOMIC ENERGY COMMISSION

RG DOE HISTORIAN (DBM)
Collection 1132
Box 3363
Folder #26

R

REPORT

on

PROJECT GABRIEL

DOE 5650.2, III-12
CLASSIFICATION CANCELLED
BY AUTHORITY OF DOE/OC
JOSE DIAZ 4-20-81
REVIEWED BY DATE
Wilbur A. STRAUSER 4-20-81
REVIEWED BY DATE
By: Dick Koogle 6-12-87

CONFIRMED TO BE UNCLASSIFIED
DOE/OFFICE OF DECLASSIFICATION
M. L. KOLBAY, A.D.C. DATE: 11/7/84

U. S. ATOMIC ENERGY COMMISSION
Division of Biology and Medicine
Washington, D. C.

July 1954

[redacted]

This document contains restricted data as defined [redacted] Act of 1946. Its [redacted] the disclosure of its [redacted] in any [redacted] an unauthorized person is prohibited.

[redacted]

DOE ARCHIVES

PROJECT GABRIEL

I. OBJECTIVE

The objective of Project GABRIEL is to evaluate the radiological hazard from the fallout of debris from nuclear weapons detonated in warfare. Depending upon the conditions under which such weapons are used, the major interest may lie either in local fallout or in the superimposed long range fallout from many weapons.

II. CONSIDERATIONS INVOLVED

Estimates of the radiological hazard to man resulting from radioactive fallout involve considerations which may be classified as follows:

A. Dependence of fallout on conditions of debris formation:
   1. Height of burst 2. Yield
B. Dependence of fallout on meteorological conditions:
   1. Debris transport 2. Influence of rain
C. Observed distribution of debris:
   1. Near point of detonation 2. Remote from detonation
D. Behavior of fission products in physical environment:
   1. Air 2. Water 3. Soil
E. Uptake and metabolism of fission products by plants.
F. Uptake and metabolism of fission products by man and animals.
G. Effects of exposure of humans to radiation:
   1. Radiation from sources external to the body.
   2. Radiation from radioisotopes within the body.

These considerations are discussed in some detail in Section V, and calculations and some tentative conclusions are given in Section VI

III. ORGANIZATION

Within the Atomic Energy Commission, the Division of Biology and Medicine is responsible for effort specifically directed towards GABRIEL. Such effort includes both the support of experimental and field studies and the correlation of relevant data from a wide range of extra-divisional activities.

A. *Theoretical studies:*

A theoretical analysis of the long range aspects of GABRIEL was made in 1949 by Dr. Nicholas M. Smith, Jr.,[1, 2] Oak Ridge National Laboratory, at the request of the Atomic Energy Commission. Smith concluded that Sr–90 is by far the most hazardous isotope resulting from nuclear detonations, and that the distribution of this isotope over large areas of the earth's surface constitutes the limiting factor in estimating the long-range hazard from the use of a large number of atomic bombs.

In 1952 RAND Corporation was given a contract to make an independent study of GABRIEL, with some emphasis on the short-range aspects of fallout. Study of this phase, later called AUREOLE, has been carried as far as present information appears to permit, and a report has been prepared.[3]

Footnote references on pp. 42–43.

DOE ARCHIVES

B. *Monitoring of fallout from nuclear weapons tests:*

During the past 3 years increasing responsibility for the monitoring of fallout has been assumed by the Health and Safety Division of the AEC New York Operations Office, with aid from the weapons test organization and from a number of federal agencies. A notable feature of the current program is a network of approximately one hundred domestic and half as many foreign stations at which fallout is collected by permitting it to fall on a horizontal gummed paper or plastic sheet. The sheet is held in a frame supported by a light metal stand, two feet in height, and has an exposed surface of one square foot. At appropriate intervals the sheet is removed, folded in half with the gummed surface inside, and sent to NYOO for measurement of activity. Some of these stations are operated continuously between series of tests.

At the Nevada test site, off-site monitoring within distances of one or two hundred miles has frequently been carried out by mobile teams spotted downwind. Conventional aircraft and helicopters have been used to a very limited extent. On-site monitoring has been performed by local radiation control groups. Fallout in these locations is generally estimated from radiation levels. Since the personnel involved in these monitoring operations have been primarily interested in problems of personnel protection rather than in applications to GABRIEL, data on short-range fallout have been very incomplete.

At the Pacific Proving Grounds, monitoring of local fallout—in this case, within distances of a few hundred miles—has been generally inadequate because of operational difficulties involved.

C. *Distribution of Sr–90 from nuclear detonations:*

In the summer of 1953 RAND held a short conference of selected consultants to make an over-all review of GABRIEL. The conference recommended[4] that studies then current be supplemented by a world-wide assay of the distribution of Sr–90 from the nuclear detonations which have occurred. This assay has been designated Project SUNSHINE.

Samples for assay have included soil, alfalfa, animals, dairy products, human bones, rain and other water, etc. Samples of one or more of these materials have been obtained from each of some 20 foreign countries. Many of the samples have been obtained through the Department of Agriculture and directly by the participating laboratories; others have been obtained through miscellaneous contacts. Three laboratories have been engaged in analyses of samples for Sr–90: University of Chicago (W. F. Libby); Columbia University (J. L. Kulp); and New York Operations Office, AEC (Merril Eisenbud and John Harley). A decision to keep the existence of a worldwide assay SECRET has limited the freedom with which suitable combinations of samples might be obtained from foreign countries.

D. *Experimental studies:*

Some estimates of the radiological hazards involved in the use of nuclear weapons have been made on the basis of the considerations listed in Section II above. Our knowledge of some of the factors involved is very meager, leading to large uncertainties. While direct observation of the distribution of Sr–90 from nuclear detonations reduces some of these uncertainties, application to prediction of the distributions which may result under other sets of conditions requires knowledge of the effects of those factors which change from one set of conditions to another. Of fundamental interest are soil-plant-animal-human relationships, utilization of land, relative geographical locations, climate, etc. In addition to these, there are questions of radiological effects of exposure to radiation—questions on which Project SUNSHINE sheds no light.

Some evaluation of the factors involved in extrapolation of observations on human exposure to Sr–90 from past nuclear detonations to exposures to be anticipated from the use of nuclear weapons in warfare is built into Project SUNSHINE. For example, samples of soil, plants and

Footnote references on pp. 42–43.

DOE ARCHIVES

animals or, in some cases, milk are taken from the same pasture where possible. An effort is made to relate the Sr–90 content of babies from a large city to the Sr–90 content of the local milk supply. Some of the questions raised by field observations, as well as problems of radiological effects, are incorporated into the rather large experimental program supported by the Division of Biology and Medicine.

E. *Information from weapons tests:*

A great amount of information pertinent to GABRIEL is, of course, potentially available in connection with weapons tests. Some of this is obtained for other purposes in the normal course of events; other items become available only through projects specifically designed to supply information for GABRIEL or for scientific interests which happen to be related.

In addition to studies of weapons effects made in support of the weapons development program, it is feasible to include in the tests a number of programs supported for their general scientific interest. While final approval of such programs is subject to the judgment of the Test Director that their inclusion is feasible, programs of interest to the Division of Biology and Medicine are, to some extent, coordinated or instigated by screening committees sponsored by the Division.

IV. SPECIAL METHODS

Certain methods of measurement, particularly in connection with Project SUNSHINE, are of interest because of the high degree of specialization involved. Because of the amount of detail involved in their description, they are presented as Appendix A.

V. RECENT DATA PERTAINING TO PROJECT GABRIEL

In this section are discussed recent data which pertain to the steps of Project GABRIEL as listed in Section II. Specific references are given only for material taken from formal reports.

A. *Dependence of fallout on conditions of debris formation:*

The ultimate time scale and spatial distribution of fallout from an atomic weapon detonation are greatly influenced by the conditions of debris formation: height of the burst above the ground, the yield of the burst, and presumably also the soil conditions. The conclusions listed below have been deduced [3] from detailed studies of a large number of American weapons tests (almost exclusively Nevada series).

Airburst is defined to mean a shot in which the fireball does not touch the ground. Tower shots (100–300 feet elevation) include those detonations in which a fraction less than one-half of the fireball contacts the ground. Surface shots are those in which one-half or more of the fireball contacts the ground. The percentages of fallout cited below are based on survey meter readings inside a circle of 200 miles radius and gummed paper collections outside this circle. While the gummed paper collections are here assumed to have an efficiency of 100%, the true value is certainly lower, as discussed later.

1. *Initial distribution of radioactive material in the vertical:*

a. *Airburst.*—About 99% of all the activity remains in the mushroom or toroidal cloud in a finely divided form, and some 1% is left in the wake, extending from the burst point up to the mushroom. The fallout curtain of slowly descending particles of intermediate size develops after the cloud stabilizes, but this appears to constitute even less than the wake and is usually barely detectable in the case of an airburst.

b. *Tower shot.*—Most of the material is taken up with the mushroom, but the settling of the entrained dust and sand apparently brings some of the debris back down into the stem and results in some radioactivity at all altitudes. The fraction of the total radioactivity removed from the mushroom in this way is in the order of 20% to 50%.

Footnote references on pp. 42–43.

DOE ARCHIVES

c. *Surface shot.*—The picture is about the same as in the tower shot, except that more surface dust and sand is entrained; therefore the fraction falling is probably greater.

2. *Dry fallout in the first few hours:*

a. *Airburst.*—Practically no fallout is observed, even in the case of low air bursts where the dust column catches and joins with the mushroom in the course of its rise. The fallout curtain constitutes less than 1%, and does not begin to reach the ground until several hours after the burst.

b. *Tower shot.*—The height of tower and yield have an effect on the fraction which falls in the first few hours, the trend being towards larger fractions being scavenged for the larger yields and the lower burst altitudes, as one would expect. The observed early fallouts in Nevada range from 5% to 30%, with the most probable fraction about 15%. Most of these shots have been over sandy desert. A few detonations have been over significantly finer grained soil, and in these cases close-in fallout has been considerably lower.

c. *Surface shot.*—The only data available for early fallout from surface shots are from a low kiloton device in Nevada and a megaton-range device on a Pacific coral atoll. Information on neither of these was as accurate as could be wished, but estimates for close-in fallout (within a few hundred miles for the Pacific shot) were in the range of 50% to 90%.

3. *Deposition after the first few hours on the continent from rain and dry fallout:*

a. *Airburst (low yield).*—The yield of a bomb determines the height to which the debris is carried. When the yield is in the order of 5 KT or less a large fraction remains down in the rain-bearing level, where it can be scavenged by rain. In such cases about 15% of the total yield falls on the North American continent from tests in Nevada.

b. *Airburst (high yield).*—Yields of 10 KT or more result in the mushroom cloud and 99% of the material being carried above the top of the rain-bearing level (about 20,000 ft. in summer). Thus, the rainout and fallout from such a burst can come only from the wake and fallout curtain (amounting to about 1%) or from an occasional thunderstorm or towering cumulus cloud which results in downward mixing of the material. The continental fallout from such bursts in Nevada has generally accounted for only about 1% of the total activity.

c. *Tower shot.*—Here, as stated above, the radioactive debris is spread over all altitudes to some extent, and the continental fallout from Nevada tests has usually amounted to about 5% of the total activity. These have all been from "high yield" tests, which in this context means greater than about 10 KT, so that most of the material is carried above the rain-bearing level.

4. *World-wide fallout:*

There are essentially no data available as yet which permit a correlation of very distant fallout with conditions of debris formation. At large time and distance, fallout has not been unambiguously identifiable with any single shot in a series of unlike detonations.

5. *Physical and chemical composition of debris:*

Data available on physical and chemical composition of bomb debris do not give a comprehensive picture. The type of particle collected for study depends so strongly on the nature of the collecting apparatus that representative sampling has not been achieved. Therefore, only qualitative descriptions of the particles are possible.

The height of burst has a marked influence on the method of formation of the particles, and therefore on their chemical, radiochemical, and physical properties. The following methods of formation and properties have been observed:

a. The gaseous mixture of fission products, earth, and bomb casing condensed into droplets and solidified before coming into contact with particles of liquid or solid earth. These particles from fission products were black, glassy, ferro-magnetic, and highly radioactive. They contained considerable iron, probably as magnetite.

DOE ARCHIVES

b. The gaseous mixture of fission products, etc., condensed on liquid droplets of earth which never became hot enough to vaporize. Particles formed in this manner were partially black or green, glassy, and with the activity rather uniformly distributed throughout the particle.

c. The gaseous mixture of fission products, etc., condensed on solid earth particles which were passing through the incandescent cloud. Only the surfaces of such particles were active.

d. Various agglomerations of the foregoing particle types.

Measurements have been performed on collected debris to determine if fractionation takes place in the relative abundance of the various fission product isotopes. The following partial picture can be constructed from present evidence:

a. Nearly all airborne debris samples collected within a few days after a detonation have shown comparatively little fractionation. For example, Sr–89 and Ba–140 abundance has nearly always lain between 0.4 and 1.5 relative to Mo–99 despite the noble gas precursors of these isotopes which might reduce condensation. However, examples of fractionation have been observed after both kiloton and megaton range surface shots, in which airborne debris is enriched and close in fallout depleted in Sr–89, 90 by a factor of 10–100. The picture is not yet entirely consistent.

b. Fallout within the U.S. from Nevada tower and air bursts has not been seriously fractionated in Sr–89 and Sr–90. Within a factor of a few, the abundance of these isotopes relative to each other and to the gross fission activity appears to have been as expected assuming no fractionation.

c. Residual high altitude air activity after a few months appears from limited data to be enriched many fold in Sr–89, 90 relative to gross fission product activity.

B. *Dependence of fallout on meteorological conditions:*

Fallout depends on meteorological conditions in 2 respects: Location of fallout and quantity of fallout. Considerable information obtained both theoretically and experimentally is available on these points.[3]

1. *Close-in fallout:*

The typical close-in fallout pattern for tower shots in Nevada has been an elongated plume downwind from the detonation site. Often the point of maximum dose rate, except at the site itself, has been found many miles downwind, as a result of debris having fallen from tropopause altitude in about 3 hours. Attempts have been made to correlate fallout pattern with local meteorological and debris cloud height conditions. One method has been to trace the path of fall of typical particles (e.g., 125 micron diameter at one test site) from cloud to ground assuming Stoke's law. However, vertical convection distorts the predictions of this analysis. Another method has been to establish purely empirical correlations between wind conditions and fallout pattern observed in past shots. The results of one such analysis indicated maximum surface dose rate was proportional to wind velocity shear with altitude. However, it is very likely that this overall correlation depends upon other interactions of meteorological conditions which may have only local validity.

2. *Statistical estimate of long range cloud transport:*

In order to predict the position to which a particular point in a debris cloud will be transported by the wind, statistical calculations have been carried out for Nevada shots over a 2-day period at the 300 milli-bar air pressure level (c.30,000 ft.) in the spring season. The mean

Footnote references on pp. 42–43

DOE ARCHIVES

trajectory endpoint under these assumptions was found to be over eastern Kansas at 24 hours, and over northeast North Carolina at 48 hours. Around each of these mean endpoints a circle can be drawn inside of which half the actual endpoints in a large number of tests would be expected to fall. The radii of the 24-hour and 48-hour circles are approximately 800 and 1,300 miles respectively. Insofar as cloud location data from airplane tracking are available, they are consistent with this meteorological prediction. From these results the probability of fallout from Nevada tests appears to be substantially the same at all points in the eastern United States.

The debris in a single mushroom is progressively diluted, chiefly through the effects of wind velocity shear with altitude. Statistical calculations have been performed on the basis of meteorological records to gain a quantitive understanding of this mode of dispersal as pertinent to intervals of 12 and 24 hours after Nevada shots in the spring season. Among other more detailed conclusions, the results show that after 12 hours the 200 mb (c.40,000 ft.) endpoint is found, on the average, 120 miles east of the 300 mb endpoint; after 24 hours, 175 miles southeast. If the debris cloud is initially a sphere tangent at top and bottom to the 200 mb and 300 mb planes, respectively, it will be smeared out by wind velocity shear into a 120-mile ribbon at 12 hours, and a 175-mile ribbon at 24 hours. Additional dispersal of the debris is caused by gravity, diffusion, and turbulence.

3. *Effect of rain on fallout:*

Calculations and experimental observations have been performed to determine the influence of rain on fallout. Calculations, however, depend on so many parameters which are inaccurately known that they can probably be relied upon only for qualitative conclusions.

Some of the conclusions enumerated in a study by the RAND Corporation on the basis of both theory and experiments, are these: (1) The most important mode of action of rain is mechanical scavenging of debris by falling raindrops; in comparison, mechanical scavenging by cloud droplets, rain induced down drafts, and electrostatic scavenging by raindrops or cloud droplets are small effects. (2) Fraction of activity removed from air by rain falling through it is dependent mainly on total rainfall, not intensity of rainfall. (3) To the extent that quantitative predictions are reliable, half the activity in air through which rain falls would be removed by about 0.04 inches of rain. (4) Mushrooms from bursts of greater than 6 to 8 KT would be altogether above rain bearing layer (maximum altitude about 20,000 feet) except in the case of thunderstorms (sometimes extend to 50,000 feet).

Experimental comparisons between rainout and dry fallout may be rather inaccurate because of differences in collector efficiency for the two conditions. In analyzing a set of the New York laboratory's gummed paper fallout collections, the activity of papers exposed during rainy days was compared with those exposed on dry days. Papers exposed during rain were on the average 2.5 times more active than the dry collections. The efficiency of the gummed paper for rainfall collections has been estimated in simulated rainfall experiments to be 80%, 50%, and 20% for particles of 2, 1, and 0.4 micron diameter, respectively, but average efficiency for field conditions is unknown. Since rain occurs during only a small fraction of any specified period at any one place, it may be that cumulative dry fallout is greater than cumulative rainout.

No experimental observations on rainout close in to a shot are available since tests are conducted only when local weather is dry.

Rain has been analyzed for its Sr–90 content by both Libby [5] and Kulp.[6] Libby's series included 20 samples from the Chicago area collected between November 1952 and October 1953, and Kulp's included 14 samples from N. Y. between January 11 and April 1, 1954. Average activity except during test series was about 4 dpm Sr–90/gal, but rose both in June 1953 and April 1954 to peaks of approximately 100 dpm Sr–90/gal.

Footnote references on pp. 42–43.

DOE ARCHIVES

4. *Atmospheric storage of debris:*

The length of storage of debris in the atmosphere influences both the uniformity of geographical distribution of fallout and the rate at which it becomes available for oral uptake. The significance of height of burst and yield have already been mentioned. Storage is greater for airbursts than for tower shots than for surface shots. Also, it is greater for those shots whose cloud penetrates into the stratosphere than for those whose cloud remains in the troposphere than for those whose cloud remains in the rain-bearing layer. The mixture of the various types of shots in any one weapon test series has made it difficult to interpret observed storage in terms of these parameters. However, certain limits may be set from the experimental data now available.

Reasoning from present observed geographical distribution of fallout to storage time, one obtains a certain amount of information on this time. For tower or air shots in Nevada (spring, 1953), it is shown below that fallout per unit area in the eastern half of the United States was only a few times as great as that in western Europe. When allowance is made for spread in latitude of cloud trajectories at increased distance from detonation site, it appears that half life of storage under these conditions is greater than one week.

It is also shown below that an estimate of at least 15% of the debris from this same test series had fallen out by June 14, 1953. Allowing for gummed paper inefficiency and insufficient time for fallout from the final large shot, it seems probable that the half time for fallout was less than 2 months.

Direct measurements have been performed on air activity collected at various altitudes. These suggest an upper limit for the fraction of weapon debris, from past tests, which has been stored in the atmosphere over long periods. The New York laboratory has analyzed air filter samples from surface level in New York, air filter samples collected by jet planes at 40,000 feet in New Mexico, and balloon borne electrostatic precipitator collections at 80,000 feet over New Mexico. The 40,000 feet and 80,000 feet samples proved to be 50–100% Sr–89 and Sr–90. At the beginning of 1954, Sr–90 alone represented 5–15% of the total; during the spring tests it was a lower percentage. The results are shown in Table 1.

While these data are too few and variable to permit extensive interpretation, probably at least one important conclusion can be drawn. By comparison with soil Sr–90 measurements reported below, it seems likely that less than half of the total Sr–90 produced prior to the test series of the spring, 1954, was still in the air at the start of 1954.

TABLE 1

Date	Surface	40,000 feet	80,000 feet
Dec., '53 through March, '54.	Av. 0.005 dpm/ft³ . . .	Av. 0.017 dpm/ft³ . . . . . (range: 0.001–0.05)	Av. 4.3 x 10^{-4} dpm/ft³
April, '54 through June, '54.	Av. 0.01 dpm/ft³ . . . . (range: 0.001–0.02)	No measurements made.	Av. 5 x 10^{-3} dpm/ft³ (range: 0.4–30 x 10^{-3})

C. *Present distribution of weapon debris:*

The present geographical distribution of weapon debris can be estimated from three types of measurement: Survey meter data on close-in fallout taken immediately after detonation; gummed paper measurements at United States and, in recent tests, foreign stations; and analysis of soil for Sr–90 in Project SUNSHINE.

1. *Close-in fallout:*

The nature of the fallout patterns in the neighborhood of the test site has been discussed earlier. Within a radius of 200 miles of the Nevada test site approximately 10% of all debris

DOE ARCHIVES

produced (about 20% of tower shot debris) is believed to have been deposited, generally in narrow plumes. Maximum downwind "hot spot" dose rate from tower shots has been in the order of 1 r/hr at 3 hours, corresponding roughly to the deposition per square mile of 0.01 KT of fission debris.

Data on close-in fallout from multi-megaton detonations are available with reasonable reliability only for the coral atoll surface shot of March 1, 1954. This was estimated to be very roughly 50% in a plume extending several hundred miles east. Exclusive of the downwind islands in Bikini atoll itself, the nearest island in the fallout path (at a distance of 100 miles) had a dose rate of roughly 250 r/hr at 6 hours after detonation, corresponding approximately to 2 KT of fission debris per square mile.

2. *World-wide gummed paper collections:*

Systematic gummed paper collections at a large number of points throughout the United States have been carried out for all five American test series since October, 1951.[7-11] Foreign collections have been made for all three tests since November, 1953.[9-11] Data from the Pacific tests of 1954 are as yet available in only preliminary form.

The distribution in the United States of debris from both the Nevada and Pacific tests has been observed at approximately 100 weather stations. Since fallout in each series has been dominated by 2 or 3 shots, the statistical variation in trajectory and therefore the area of fallout has given greater geographical nonuniformities than would be the case for a large number of shots. The February 1954 contamination in the United States was dominated by the test series of the spring, 1953, with somewhat less contribution from spring, 1952, much less from the Pacific tests of 1952, and only a small component from all other tests. Cumulative activity in the United States in February 1954, was about 1,000 dpm/ft^2 over most of the country, assuming 100% gummed paper collection efficiency. The West Coast was lower by a factor of perhaps 10, Nevada and states east to Colorado were higher by a factor of about 2 or 3, and Florida was lower by a rough factor of 2 or 3. There were apparently some isolated small areas of activity greater by as much as a factor of 5 or 10 than their surroundings. Nonuniformities were presumably greatest near the test site, but high local rainouts have been observed as far east as Albany, N.Y.

The distribution in foreign countries of debris from Nevada tests has been observed essentially in only one series, spring, 1953, at 12 stations outside North America. Interpolating through weather data between the widely separated stations, the following picture of world-wide fallout per unit area relative to that in the eastern U. S. can be inferred:

Southern hemisphere	<1%
North Atlantic, north temperate zone in Europe and Asia	15–40%
Arctic	<15%
North Pacific	5%
North tropics (Atlantic)	5–15%
North tropics (rest of world)	2–10%

The world-wide distribution of debris from the Pacific tests of November 1952, is a complicated pattern. North America and South America to 20°S. latitude showed fallout varying unsystematically over a 10-fold range, with an average of about 700 dpm/ft^2 as of 1 January, 1953. Western Europe, Africa, and Asia as far east as India, averaged about 150 dpm/ft^2, again with considerable spread. The Asian Pacific seaboard and Pacific Islands showed wide variations ranging from a high of 230,000 dpm/ft^2 on Iwo Jima to levels of a few thousand dpm/ft^2. These data are difficult to interpret for the following reasons: (1) Origin of debris, whether from large yield surface shot or smaller air burst, is ambiguous. (2) Amount of close-in fallout is

Footnote references on pp. 42–43.

DOE ARCHIVES

completely unknown. (3) Complicated wind structure blew debris below 10,000 feet and above tropopause to the west, intervening debris to the east.

Comparisons have been made between total debris production and total fallout as estimated from test site survey data and world-wide gummed paper collections. The gummed paper is here assumed to have an efficiency of 100%, although probably it is less than 50%. For the Nevada series of the spring, 1953, where approximately half of the debris was produced in tower shots and half in air drops, the following debris inventory through June 14, 1953, has been estimated: [11]

Within 200 miles of test site	9% of debris produced	
U.S. (excluding 200 mile circle)	3	Do.
Northern Hemisphere (excluding U.S.)	12	Do.
Southern Hemisphere	1	Do.
World total fallout	25%	Do.

For the Pacific tests of November 1952, observed world-wide fallout [9] was only a few percent of debris produced. However, data from the spring, 1954, tests make it apparent that a large fraction must have fallen out unobserved within a few hundred miles of the test site in the case of the high yield surface shot.

3. *Soil analysis for Sr–90:*

The analysis of Sr–90 concentrations in soil is being carried out in Project SUNSHINE. Libby [5] has measured approximately 500 dpm/ft^2 of Sr–90 for 7 alfalfa fields in the Chicago area, as of September 1953. A somewhat higher value may be obtained when further measurements on fused rather than HCL–leached soil samples are completed. A single soil sample from Ankara, Turkey, showed about 250 dpm/ft^2 Sr–90. Harley has found activities of about 0.5 μc Sr–90/ft^2 on the most highly contaminated Pacific islands following the test of March 1, 1954. Additional analyses of foreign soil samples will soon be available.

Libby's Chicago data were approximately 5 times higher than predicted from the gummed paper data for the region (assuming no fractionation of the isotopes in the gummed paper collections). This may indicate that the gummed paper efficiency is only 20%. However, Harley's [12] analyses of Sr–89 and Sr–90 activity in 3 pastures in New York and New Jersey, one in Georgia, and one in Utah, gave ratios of measured to predicted values of only one or two.

D. *Behavior of fission products in physical environment:*

1. *Air:*

The behavior of weapon debris in the air has been covered in Section B above, dealing with meteorology, rainout, and atmospheric storage.

2. *Water:*

H. A. Thomas of Harvard University has carried out extensive measurements on contamination of rivers and reservoirs by fallout in the Boston area. Approximately 0.8% of the activity which falls out in that region has been found within weeks in the streams. Since 2% of the watershed is covered by water, Thomas has concluded that only the activity which falls on a water surface finds its way into the water system. His measurements do not exclude the possibility of slow leaching over a long period of time.

W. F. Libby [5] has analyzed during 1953 the Sr–90 content of the following rivers: Mississippi, Mosel, Seine, and Donav (near Ulm, Germany). The average activity was 0.4 dpm Sr–90 per gallon (range 0–1.1 dpm/gal), or less than that of rainfall by a very rough factor of 10.

Thomas has studied the effect of standard water purification procedures on the gross fallout content of reservoir water. Alum flocculation plus rapid sand filtration reduced the activity

Footnote references on pp. 42–43.

DOE ARCHIVES

by a factor of 1.5–5. This was a considerably lower factor than obtained in experiments on water contaminated artificially by pile produced isotopes.

G. V. Alexander [13] at UCLA has measured the Ca and stable Sr content of the water supplies of 50 American cities, both before and after purification. The Ca and Sr were in general found to be removed in equal percentage. For chemical treatment (alum or $FeSO_4$, sometimes plus lime, lime-soda ash, or lime-phosphates) the Sr was removed in the range of 10–70%. Water softeners only (phosphates-zeolite) removed about 70% in the 2 cities examined. The observed Sr/Ca ratio (by weight) had a mean of 6×10^{-3} (range $0.9—30 \times 10^{-3}$).

3. *Soil:*

Measurements on the distribution of fallout with depth of soil indicate that it is held fairly firmly in the top inches of soil. Thomas found the following relative gross activities at various depths in Boston soil in 1953: ¼″–5, 1″–3, 4″–1. Libby [5] has obtained data on the Sr–90 content of the layers 0–1″ and 1–6″ for 7 Chicago milkshed alfalfa fields in October 1953. The total content of the 1–6″ layer averaged 1.7 times as great as that of the 0–1″ layer (range 0.6–3.5). (Activity per gram soil in the 1–6″ layer therefore averaged ⅓ that in the 0–1″ layer.) Approximately half of this Sr–90 had fallen on the soil at 1½ years before measurement, and approximately half at ½ year before measurement.

Data available at present indicate that the major portion of the long range fallout is in a state readily available to plants. M. M. Weiss [14] at Brookhaven observed that about half the activity in rain was filterable through papers which passed particles of less than ½ micron diameter. Libby [5] analyzed successive extracts of the same sample of soil, the first extraction with ammonium acetate containing the available Sr–90, the second with dilute HCl containing a part of the unavailable Sr–90. (A third extract, namely complete solution of the soil, will be analyzed in the future.) The ratio of available to total observed Sr–90 (ammonium acetate fraction to ammonium acetate + HCl fractions) was 0.8, 0.7, and 0.4, respectively, on the 3 different soil samples measured.

Harley [12] compared extraction of bomb debris Sr–89, 90 from soil by the 3 methods of complete solution of the soil, 10 minutes leaching with 6N HCl, and 10 minutes leaching with ammonium acetate. Limited data indicate that in 2 of the 3 soils, leaching removed more than half the Sr–89, 90, but in one case only 10%.

A program has been undertaken to determine the natural (stable) Sr content of soil. These measurements will be correlated with similar data for plants, animals, and humans as an analogy to the Sr–90 uptake problem under equilibrium conditions. Preliminary data obtained by Menzel and Heald of the U. S. Department of Agriculture show a mean Sr/Ca ratio (by weight) of about $2–4 \times 10^{-3}$ for ammonium acetate leachates from a set of Wisconsin and Illinois alfalfa fields. Further measurements on these and other sites will be forthcoming. G. V. Alexander's raw water data, which presumably are indicative of the Sr/Ca ratio of the associated watershed, are consistent with these soil analyses.

E. *Uptake and metabolism of fission products by plants:*

The uptake of bomb debris by plants has been considered significant only as a step toward the contamination of humans through the food chain. Sr, which has been generally considered to be the most hazardous, has been studied most extensively.

1. *Strontium:*

Plant uptake of Sr has been examined by two different approaches: (1) Controlled laboratory or field experiments, and (2) observations on the actual atomic test debris now disseminated throughout the world.

Footnote references on pp. 42–43.

DOE ARCHIVES

a. *Experiments on uptake through roots.*—It has been fairly clearly established that the uptake of Sr by plant roots is controlled predominantly by the quantity of available Ca in the soil: Sr and Ca behave nearly like isotopes of the same element.

Menzel, of the U. S. Department of Agriculture, grew cowpeas on 42 American soils to which equal amounts of bomb debris had been added. Available Ca in these soils ranged from 0.7 to 48 milliequivalents Ca/100 gm soil. The Sr–90/Ca ratio of the plants was approximately inversely proportional to the available Ca in the soil over the full range of calcium availability.

In another set of experiments on a particular type of soil (Evesboro) to which known amounts of Sr–89 had been added at two carrier levels, the results listed in Table 2 were found. The distribution factor k_{Sr}, defined as $(Sr/Ca)_{plant}/(Sr/Ca)_{soil}$, indicates the discrimination which the plant makes between Sr and Ca uptake.

TABLE 2

Crop	(Sr/Ca) soil	k_{Sr}	k_{Ba}
Barley	0.017 (by milliequivalents)	0.45	0.020
	0.0017	0.39	0.022
Buckwheat	0.017	0.49	0.023
	0.0017	0.43	0.028
Cowpeas	0.017	0.53	0.057
	0.0017	0.37	0.053

By combining these two sets of experiments, Menzel concluded that average uptake of Sr from the 42 American soils was best fit by a distribution factor of $k_{Sr}=0.36$.

Comparisons of Sr and Ca competition have been made for root uptake from nutrient solutions. Under these conditions, Menzel found a distribution factor of 1, in agreement with the published data of Collander.[15] Further experiments on the stable Sr content of plants and soil under natural conditions are in progress to clarify the picture of comparative Sr and Ca uptake.

When Sr is not distributed uniformly throughout depth of soil, uptake depends on root depth. Menzel has compared the feeding of year old alfalfa and Kentucky bluegrass on Ca–45 layered at 3 different depths in experimental plots. The results, shown in Table 3, illustrate this dependence, although depth of feeding varies widely with soil conditions.

TABLE 3

Depth of Ca^{45}	Alfalfa uptake	Bluegrass uptake
0″	2.5 units	9.3
6″	5.7	4.7
12″	2.8	0.8

b. *Experiments on uptake through the leaves.*—Experiments on leaf uptake are underway at Michigan State College and the University of Arizona. Preliminary experiments at Arizona have shown that a considerable fraction of Sr placed on leaves as a dilute nitrate solution is retained. Biddulph has shown that Ca–45 administered to leaves in solution form can be fairly readily metabolized but is not translocated out of the absorbing leaf.

c. *Observations on atomic test debris (Project Sunshine).*—Correlated sets of alfalfa and soil samples from the Chicago milkshed, September, 1953, have been analyzed for Sr–90 content

Footnote references on pp. 42–43.

DOE ARCHIVES

by Libby.[5] His results have been reported in Sunshine Units, where 1 S.U.$=10^{-12}$ curies Sr–90/gm Ca, or approximately 1/1000 of the present maximum permissible body burden for man (1 μc/1000 gm Ca in adult skeleton). The average content of 11 alfalfa samples was 9 S.U. (range 4.3–21 S.U.). The top inch of the associated soils averaged 12 S.U., and the 1″–6″ depth averaged 4 S.U. A similar alfalfa-soil pair from near Ankara, Turkey, on October 6, 1953, yielded values of 2.1 S.U. for the alfalfa and 1.2 S.U. for the top inch of soil.

Detailed comparison of individual alfalfa-soil pairs shows little correlation between Sr uptake and quantity of available Ca. This lack of correlation between Sr and Ca uptake, and the excessively high activity of the alfalfa compared with soil, both indicate that the contamination of the alfalfa was by direct fallout on the leaves rather than by root uptake. (American weapon test series ended 4 months, and the Russian series two or three weeks, before sample collection.)

Kulp [6] at the Lamont Geological Observatory (Columbia U.) New York, made preliminary tests which also indicate the dominance of leaf over root uptake in the first months after weapon detonations. Corn (maize) leaves from Wisconsin (Sept. 1953) were first leached 5 minutes in 50% HCl, then ashed. From the leachate was recovered 1.2 gm Ca of 0.5 S.U., and from the ash 5.6 gm Ca of <0.05 S.U. The inference is that most of the activity was adhering to the outside of the leaves.

2. *Barium:*

Ba–140, a 12-day fission product of high fission yield, might be expected to behave similarly to Sr. However, its uptake by roots appears to be lower, as demonstrated by the low k_{Ba} in Menzel's experiments listed in Table 1. The distribution factor k_{Ba} is defined as $(Ba/Ca)_{plant}/Ba/Ca)_{soil}$. The question of root uptake may be academic because the short life of Ba–140 would not allow serious concentration by this path. No experiments on leaf uptake and retention are yet available.

3. *Ruthenium:*

Several experiments have been performed on a laboratory scale which permit comparison of root uptake of Ru with that of Sr.

Nutrient solution experiments by Menzel at the U.S. Department of Agriculture have shown that under some conditions Ru uptake is controlled by iron content according to the relationship $(Ru/Fe)_{plant}=0.1\ (Ru/Fe)_{solution}$. The Fe and presumably Ru was complexed with versene. If this relationship held under field conditions (which it may not, because of the complex chemistry of Ru) root uptake of Ru would be far lower than uptake of Sr: soil usually contains more Fe than Ca (which controls Sr uptake), while plants contain about 1% as much Fe as Ca.

Kermit Larson's [16] group at UCLA carried out greenhouse experiments on the comparative root uptake of Ru and Sr by barley and several vegetables. Carrier free isotopes (chemical form of Ru ambiguous) were mixed into soils. The ratio of Ru to Sr taken into the plant tops varied from 0.02 to 0.002 of the ratio in the soil.

Menzel [17] grew 9 different crops (forage crops and vegetables) on soil contaminated with close-in fallout from a weapon test. The soil was comparatively rich in Ca, with resultant suppression of Sr uptake. The ratio $(Ru/Sr)_{plant}/(Ru/Sr)_{soil}=0.02$.

Experiments on leaf uptake of Ru have not been performed.

4. *Yttrium and rare earths:*

Several experiments on root uptake of Y and the rare earths have been performed for soil contaminated with separated isotopes and with actual weapon debris.

Kermit Larson's [16] group at UCLA compared the uptake of Sr–90, Y–91, and Ce–144 by barley, carrot, lettuce, bean, and radish plants from 6 artificially contaminated soils. Uptake to plant tops of both Y and Ce was less than that of Sr by a factor greater than 100.

Footnote references on pp. 42–43.

DOE ARCHIVES

Rediske and Selders [18] at Hanford grew barley in soil artificially contaminated with Y and Sr. Y uptake was lower than Sr by 2 to 4 orders of magnitude.

The same group grew tomatoes in soil contaminated with close-in fallout from a weapon test.[19] Considerable fractionation had reduced the Sr content of the debris relative to the rare earth content, and the date of the experiment (1½ years after detonation) was such that Sr activity relative to rare earth was at its minimum. Under these conditions rare earth activity in leaves was 4 times greater than Sr activity. However, relative to corresponding activities in the soil, rare earth uptake was ⅙ that of Sr.

Menzel examined uptake of rare earths as well as of Ru and Sr by forage crops and vegetables from soil contaminated with weapon debris similar to that in the Hanford experiment. Despite reduction of Sr in the soil by fractionation, rare earth activity in the plant leaves averaged ⅐ the Sr activity at 1½ years after detonation. While the experiments of Menzel and the Hanford group are not entirely consistent, both indicate that root uptake of rare earths relative to Sr is low in the case of actual close-in fallout.

F. *Uptake and metabolism of fission products by man and animals:*

1. *Strontium:*

As with plants, the uptake of Sr by man and animals has been studied both by laboratory experiments and by observations on actual atomic test debris as now present in nature. A few measurements are available also on stable Sr content of human bones.

a. *Laboratory experiments.*—The retention of Sr by animals has been examined in numerous experiments. In Appendix B is a fairly complete collection of data on those experiments which permit an understanding of Sr metabolism in relation to Ca metabolism. About half of this information is taken from the published literature.

From these data it may be concluded that when Sr and Ca are administered under equivalent conditions, the Sr/Ca ratio retained falls between 0.25 and 1. This was true for:

(1) Wide range of species (mouse, rat, steer).
(2) Wide range of age.
(3) Wide range of carrier Sr.
(4) Conditions of normal or disturbed Ca metabolism.
(5) Oral or injected route of administration.

While most of the experiments have determined comparative Sr and Ca retention over only a short period of time, the 2 rat experiments, plus the fact that Sr and Ca excretion is low after the first few days of retention, indicate a similar ratio would hold over longer periods.

These data show that under conditions of widespread environmental contamination, Sr uptake and retention by animals and presumably by man would be determined by the dietary Ca status.

The extent of Sr incorporation into milk is an important problem, since milk is the most important source of Ca in the American diet. Comar has performed experiments on cows in which the Sr/Ca ratio in feed, blood, and milk was measured under equilibrium conditions. Typical relative values were 1.0, 0.37, and 0.13 respectively. In the Sunshine observations referred to in the next section, Libby found the Sr/Ca ratio in milk from 10 farms to be 15% of that of the alfalfa on which the cows were feeding (range 9–36%, with the spread probably attributable to sampling errors of various sorts).

b. *Observations on weapon test debris in environment.*—Human, animal and animal product samples from the U.S. and foreign countries have been analyzed [5, 6, 12] for Sr–89 or Sr–90.

These data are compiled in Appendix C. The outstanding conclusions are the following:

(1) Dependence of activity on longitude is not large.

Footnote references on pp. 42–43.

DOE ARCHIVES

(2) Animal bones lie in the range of a few Sunshine units.
(3) Milk and cheese have an activity of 1–2 S.U.
(4) Chicago human stillborns at 0.14 S.U. are lower in activity than the local milk by a factor of 10, a part of which is undoubtedly caused by a placental barrier to Sr transfer. (Comar found the Sr/Ca ratio in rat fetuses to be about ½ that of the mother at equilibrium.)

No direct measurements are yet available on the current animal intake of Sr–90 produced in the 1954 test series. However, for purposes of comparison of the body burden of this Sr–90 with that of the 1954 I–131, discussed later, a rough estimate can be made. Cumulative Sr–90 fallout in the U.S. from the 1954 series is thought to be very roughly 50 dpm/ft^2. By analogy with previous tests, it is expected that grazing animals would acquire a body burden of the order of 1 Sunshine unit from this quantity of Sr–90 fallout.

The interpretation of all these data for long term hazard evaluation is made difficult by uncertainties in the current route of entry of Sr–90 into the food chain. If the present picture is dominated by external leaf contamination with current fallout, present data would give an exaggerated estimate of the long term hazard.

c. *Stable Sr measurements.*—Measurements on the stable Sr content of human bones have been made as part of the picture of the soil-plant-animal Sr chain under equilibrium conditions. Tipton [20] studied bones from 28 individuals by activation analysis, finding an average Sr/Ca ratio of 2×10^{-4} by weight (range $0.2–7 \times 10^{-4}$). Hodges, et al,[21] made spectrographic analyses on the bones of 26 individuals ranging in age from fetus to 75 years, and of 12 cadavers preserved since 1914. The fetuses had an average Sr/Ca ratio of 4.5×10^{-4} by weight (range $4.2–5.3 \times 10^{-4}$), the others 7×10^{-4} (range 4.5–15). No significant differences were observed between the Sr/Ca ratios of different bones in the same individual, nor between the 1949 and 1914 series of specimens.

It appears that the stable Sr/Ca ratio of bone is very roughly 10% that of soil. This correlates well with expectations of cumulative discrimination between the 2 elements in the food chain. Estimates of discrimination against Sr are: soil-plant, factor of 2, cow feed-milk, factor of 6; food-bone, factor of 2. In the U.S., 75% of food Ca is in milk, 25% in plants. Weighted discrimination is then $[(25\%)(\frac{1}{2}) + (75\%)(\frac{1}{2})(\frac{1}{6})]\ [\frac{1}{2}] = 10\%$.

2. *Fission products other than strontium:*

Observations, both by the United Kingdom and by the United States on quantities of radioiodine in human urine, animal thyroids, etc., and of radioruthenium in bomb debris have directed attention to the relative importance of these materials to GABRIEL.

Accidental exposure of personnel to short-range fallout from one of the Pacific tests, 1954, provided some data on this subject. Urine samples taken from natives exposed on one island and Americans on another were brought to Los Alamos Scientific Laboratory for analyses. From the results LASL suggested the relative initial average body contents (μc basis) of the principal radioisotopes involved to be as shown in table 4.

I–131 activity has also been observed in American animal and human thyroids as a result of the spring, 1954, Pacific tests. Thyroids from some 50 cattle and 25 sheep raised in the Great Plains, Florida, Kentucky, and Massachusetts have been analyzed. Slaughter took place during the last 3 weeks of June. As of June 12, the values were in the range of $0.4–4 \times 10^{-3}$ μc I–131 per gram thyroid. A few hogs and dogs were found to contain considerably less I–131. Thyroids from 4 human autopsies in Chicago were collected and analyzed during the first 2 weeks of July. They showed measurable levels of activity, but the possibility of laboratory contamination has not been definitely ruled out. Two N. Y. human thyroids showed less than ½ dpm I–131/thyroid at the end of July.

Footnote references on pp. 42–43.

DOE ARCHIVES

TABLE 4.—ESTIMATED RELATIVE INITIAL AVERAGE BODY CONTENT
(Compared to Body Content of I–131)

Radioisotope	Monitored group of natives	Americans
I–131*	1 unit of activity	1 unit of activity.
Sr–89	0.04	0.023
Ba–140	0.006	0.015
Ca–45	0.0034	0.0023
Ru–103, 106	0.0005	0.0009

[*From the data on the relative yields of short-lived radioisotopes of iodine, it was estimated by LASL that the initial body dose rate, computed on an energy basis, from such isotopes would be greater than that from I–131 by a factor of 100, and that the integrated dose from such short-lived isotopes of iodine would be greater than that from I–131 by a factor of 2.]

It is reasonable to expect human intake of I–131 to be largely through milk. Data obtained by Brody at the U. of Missouri indicate 10% iodine transmission into cows' milk. Experiments at Hanford on sheep suggest that approximately 20% of the iodine ingested by these animals was transmitted to their milk.

G. *Radiological effects of various isotopes inside body:*

1. *Sr–89 and Sr–90:*

The currently available experimental data on the toxicity of Sr–89 and Sr–90 are tabulated in Appendix D. Skeletal dose rate has been calculated under the arbitrary assumptions of skeletal weight equals $1/10$ body weight and retained dose equals 50% of injected dose (unless further data on retention available). Considerable information is available on the acute effects of these isotopes, but only a very little on the chronic effects.

The best present estimate for chronic Sr–90 toxicity in humans is based on human Ra data converted to Sr–90 by comparisons of Ra and Sr–89 or Sr–90 in animals. Brues has estimated, both from the large experiment on mice in Appendix D and from other small scale experiments on rats, rabbits, and dogs, that Sr–90 would be $1/10$ as hazardous to humans as Ra (microcurie basis).

Human data on chronic Ra toxicity are now available [22] for 50 individuals who were given Ra salts either orally or intravenously for medical purposes, and 28 luminous dial workers who ingested Ra salts and salts of MsTh and RaTh. The groups include individuals with current Ra burdens of 0.01 to 22 μc. None of the 13 cases of 0.4 μc or less (including 7 between 0.2 and 0.4 μc) shows any symptoms after 18–30 years. Skeletal changes have been shown radiographically in several cases of body content between 0.5 and 1 μc, and in almost all cases of greater body burden. Eight of the 28 luminous dial workers and 5 of the 50 medically "treated" individuals developed malignancies, the lowest associated burden being 0.5 μc, and the next two lowest, 0.8 μc. Several patients have carried a body burden of greater than 10 μc for over 20 years and are still alive.

Since 15 individuals in each group, dial painters and medically "treated" patients, were discovered as a result of symptoms, conclusions deduced from these series are perhaps somewhat biased in the direction of excessive hazard.

Using the Ra to Sr–90 conversion factor of 10, one would estimate from these data that 1 μc Sr–90 adult body burden at 20 years after exposure would be safe, 5 μc would begin to produce radiographically demonstrable skeletal changes, and 50–200 μc might correspond to the LD 50 in 20 years.

Footnote references on pp. 42–43.

DOE ARCHIVES

It is hoped that the chronic toxicity experiments now in progress at the University of Utah can be extended so as to permit comparison of Sr-90 with Ra, MsTh, and Pu in dogs. Such a comparison would give a much more reliable conversion factor between Ra and Sr than is now available.

2. *I-131:*

In view of the probable importance of I-131 in GABRIEL, it would be desirable to compile a critical review of the dependence of thyroid damage on I-131 dose. This will be done in the near future. The sources of information are almost entirely in the open literature.

VI. CALCULATIONS AND CONCLUSIONS

From the data previously presented, it is possible to construct certain conclusions about the nature and magnitude of the radiological hazard to man from nuclear weapon debris. It would be superfluous to emphasize that such conclusions are over-simplifications based on insufficient data whose pertinence to chaotic wartime or postwar conditions is limited. A crudely estimated error is associated with each numerical value. When it is stated that a parameter has a value of N with a probable error (PE) of a factor of 2, it is meant that the most probable value is considered to be N, with an estimated 50% chance that the true value falls between $\frac{N}{2}$ and 2N (i.e., a normal error distribution is assumed on a log scale).

The hazard may be expressed in a semi-quantitative manner by the equation:

$$H = F(q,t)\ D(o,t)\ S(o,d,r)$$

where

H = hazard

F(q,t) = fallout in terms of *quantity* (KT of fission products/mi^2) and *time* (days) after detonation

D(o,t) = average dose in rads to the *organ* concerned as a function of *time* after detonation, for a fallout of 1 KT/mi^2.

S(o,d,r) = sensitivity of the *organ* concerned in terms of damage as a function of *dose* and dose *rate*. (See section V., G)

A. *Fallout:*

The following figures will be used in the calculations of hazard.

1. *Airburst:*

All debris falls out exponentially with time.

For shots whose mushrooms stay below the tropopause (approximately 100KT or less), time for 50% of debris to be deposited on the earth's surface is 20 days (PE=factor of 1.5).

For larger shots, whose clouds penetrate well into the stratosphere, there is essentially no information pertinent to rate of fallout. It is expected, however, that the higher the yield the less rapid the fallout. As a result of slow mixing of stratosphere and troposphere, half time for fallout could be several times as long as for troposphere debris.

In any large scale war essentially all detonations would take place in the North Temperate Zone. From troposphere clouds 75% (PE=factor of 1.2) of the debris would be deposited in the N. Temperate Zone. From the stratosphere clouds, one might guess 60% deposition (PE=factor of 1.3) in the N. Temperate Zone. Assuming a half time for fallout of 20 days and a mean time for debris to circle the world (easterly movement) of 15 days, fallout per unit of longitude would vary around the world by less than a factor of 2 for an average shot. If detonations took place over a considerable spread in latitude within the N. Temperate Zone, few large populated regions would receive fallout (KT/mi^2) differing from the mean by a factor

DOE ARCHIVES

greater than 4. (If most of the debris were produced in a brief period, e.g., a few weeks, short period irregularities in precipitation might increase irregularity in fallout distribution.) Since the area of North Temperate Zone is $5.3 \times 10^7 mi^2$, average deposition per KT detonation would be about $\frac{0.7}{(5.3)(10^7)} = 1.2 \times 10^{-8}$ KT/mi².

2. *Surface burst:*

Much of the debris from surface shots falls out within 12–24 hours. A value of 70% may be estimated (PE=factor of 1.5). The fraction of debris which does not fall out in 24 hours is considered, in the absence of better information, to behave like airburst debris.

B. *Dose:*

1. *External gamma radiation:*

Assume:

(1) 1 KT/mi² at 1 day gives external gamma dose rate of 625 rads/day at 3 feet above an infinite plane.

(2) Humans, through shielding of houses, etc., receive 30% of infinite plane dose (PE=factor of 2).

(3) Unshielded infinity dose subsequent to fallout at t days after detonation= $(2.5)(625\ t^{-0.4})$ (PE=factor of 2). An arbitrary change has been made from prediction of $t^{-1.2}$ decay law to allow for weathering; i.e., a $t^{-1.4}$ decay curve is assumed.

The infinity dose (whole body) received subsequent to a fallout of 1 KT fission debris/mi² at various days after detonation is shown in Table 5.

2. *Debris ingested through soil-plant chain:*

a. *Sr–90 (calculations for infant or child):*

Assume:

(1) Half of Sr–90 fallout remains in soil in available form until decays or is taken up by plant (PE=factor of 1.5).

(2) Available Ca/ft² to depth of 6″ (plow depth)=8me/100 gms (PE=factor of 2).

(3) 70% of plant Ca and Sr uptake is from top 6″ of soil (PE=factor of 1.4).

(4) $\frac{(Sr/Ca)\ plant}{(Sr/Ca)\ soil} = 0.5$ (PE=factor of 1.4)

(5) $\frac{(Sr/Ca)\ skeleton}{(Sr/Ca)\ food} = 0.5$ (PE=factor of 1.5)

(6) 70% of Ca intake is in milk (PE=factor of 1.4). Depends on age, dietary habits, etc. Less outside U. S. Remainder in plants.

(7) $\frac{(Sr/Ca)\ milk}{(Sr/Ca)\ cow\ feed} = 0.15$ (PE=factor of 1.3)

(8) Skeletal Ca=15% of skeletal weight (PE=factor of 1.2).

(9) Average Sr–90 dose in skeleton is only 0.6 of value it would have if full skeleton of young adult were constituted within 1 year after fallout (PE=factor of 1.3). (This reduction in dose is brought about through dilution of current deposition in the previously formed skeleton of a rapidly growing adolescent, or through radiological and environmental attrition of activity before it is deposited in an individual who was an infant at the time of fallout. For adults, dose would be less.)

The infinity dose received for a fallout of 1 KT/mi² is shown in Table 5.

DOE ARCHIVES

b. *Sr–89 (calculations for infant):*

Assume:

(1) Assumptions (1)–(8) for Sr–90.

(2) Period between fallout and growth into plant for human uptake is 2 half-lives (PE=factor of 2).

(3) Total skeletal weight of infant is average of at least 5 times increment of skeletal weight during period of dose (PE=factor of 1.5). For a 6 month fetus or young infant, there would be less dilution. However, for these conditions an extra discrimination against Sr is present in the placenta (for rats, a factor of 2) or in the predominance of milk in the diet. A much greater dilution would take place for adults.

The infinity dose received by an infant for a fallout of 1 KT/mi^2 is shown in Table 5.

c. *Other fission products.*—The plant uptake of all other fission products is so low, or their half-lives are so short, that they represent a much smaller hazard than the Sr isotopes.

3. *Debris ingested after direct fallout on leaves:*

One important parameter required in this calculation is an estimate of the fraction of fallout retained on the leaves. This may be expressed in terms of the square feet of fallout retained per pound of vegetation, or ft^2/lb. There are several pieces of experimental data which bear on this parameter, although they do not support a reliable calculation. In the following computations, a basic assumption is made that the activity is retained on or in the vegetation for 20 days (PE=factor of 3).

The alfalfa analyzed by Libby showed an average Sr–90 activity of 20 dpm Sr–90/gm Ca, or about 180 dpm/lb dry vegetation. This represents accumulation approximately through the latter ⅔ of September, during which very roughly 20 dpm/ft^2 Sr–90 is thought to have fallen out. This gives 180/20=*9 ft^2/lb.*

Grass measured under similar conditions by Harley gave a value of *3 ft^2/lb.*

The sheep and calves analyzed for Sr–90 (Appendix C) integrated the fallout over the summer of 1953. Making several rough assumptions about fallout, sources of Ca in diet, etc., an average value is obtained of *5 ft^2/lb.*

The cattle and sheep analyzed for I–131 in June, 1954, led to very rough values of *1 ft^2/lb.*

From these figures a value of 4 ft^2/lb (PE=factor of 3) is selected as the basis of further computations. This value corresponds to 10% retention by the vegetation on a typical pasture with a stand of 1000 lbs/acre at time of fallout.

a. *Sr–90 (calculations for infant or child):*

Assume:

(1) $\frac{\text{(Sr–90/Ca) food}}{\text{(Sr–90/Ca) vegetation}}$ =0.4 (PE=factor of 2) as a result of selection against Sr in milk component of diet.

(2) Ca content of vegetation is 0.5% on dry weight basis (PE=factor of 1.5).

(3) Intake extends through period of 20 days (PE=factor of 3).

(4) Biological half-life long compared with radiological half-life. (Most of excretion which takes place occurs within a few weeks or months of ingestion. Assumptions (5) and (6) on retention apply to long term retention.)

(5) $\frac{\text{(Sr/Ca) skeleton}}{\text{(Sr/Ca) food}}$=0.5 (PE=factor of 1.5).

(6) Skeletal Ca=15% of skeletal weight. (PE=factor of 1.2).

DOE ARCHIVES

(7) Activity incorporated in skeleton during 20 days of intake is diluted in total skeletal mass at least 100 times greater than 20 day incremental mass (PE=factor of 2). (In rapidly growing adolescent this dilution takes place at time of ingestion. In infant, dilution takes place with subsequent growth before a large fraction of the activity decays.) Few localized "hot spots" exceed average skeletal dose rate by factor greater than 10.

The infinity dose resulting under these assumed conditions from a fallout of 1 KT/mi² is shown in Table 5.

b. *Sr–89 (calculations for infant):*

Assume:

(1) $\frac{\text{(Sr–89/Ca) food}}{\text{(Sr–89/Ca) vegetation}}=0.2$ (PE=factor of 2) as a result of selection against Sr in milk. When infant on mother's milk, double selection would occur. For older child on solid diet, selection against Sr would be by smaller factor, but dilution in (3) would be greater.

(2) Assumptions (2)–(6) for Sr–90.

(3) Activity incorporated in infant's skeleton during 20 days' intake is diluted in total skeletal mass at least 10 times greater than 20 days incremental mass (PE=factor of 1.5).

The infinity dose computed, under these conditions, for a fallout of 1 KT/mi² is shown in Table 5.

c. *Ba–140 (calculations for infant):*

Assume same as for Sr–89, except that:

$$\frac{\text{(Ba/Ca) food}}{\text{(Ba/Ca) vegetation}}=0.05 \text{ (PE=factor of 3).}$$

and

$$\frac{\text{(Ba/Ca) skeleton}}{\text{(Ba/Ca) food}}=0.25 \text{ (PE=factor of 2).}$$

These assumptions lead to the infinity dose as shown in Table 5.

d. *I–131 (calculations for infant):*

Assume:

(1) 35 lbs/day cow feed intake, dry weight basis (PE=factor of 1.2).

(2) 10% of cow I–131 intake transmitted to milk (PE=factor of 2).

(3) Infant drinks 5% of milk production of 1 cow (PE=factor of 1.5).

(4) Infant thyroid mass=1.5 gm (PE=factor of 2).

(5) Infant thyroid retention=30% of ingested I–131 (PE=factor of 2).

These assumptions lead to the infinity doses as shown in Table 5.

4. *Fission product ingestion in drinking water:*

Fission products will also be ingested in the drinking water. The following general assumptions are made for all isotopes:

(1) An individual, per 70 kg of body weight, consumes an amount of water equal to the rainfall on 13 ft² (PE=factor of 2). (Two liters/day drunk, 25 inches/yr rainfall.)

(2) 2% of activity remains in solution in water, the remainder being filtered out or absorbed in watershed soil, etc. (PE=factor of 4).

(3) Delay between fallout and consumption gives exponential function for intake vs. time, half of total intake occurring in 40 days (PE=factor of 3).

DOE ARCHIVES

For particular isotopes these further assumptions are made about retention of ingested elements:

(1) Sr–89, 90—10% retention in skeleton (PE=factor of 1.5).
(2) Ba–140—5% retention in skeleton (PE=factor of 2).
(3) I–131—30% retention in thyroid (PE=factor of 1.5).

The infinity dose to the pertinent organ, as computed from these assumptions, is listed in Table 5 for a fallout of 1 KT/mi^2.

5. *Fission product inhalation:*

To estimate the intake of fission products through inhalation, these assumptions are made:

(1) Breathing rate, per kg of body weight, is 0.15 liters/min. (PE=factor of 1.5, depending on body weight).

(2) Activity per unit mass of air breathed is equal to the average activity/unit mass throughout the troposphere (PE=factor of 3).

(3) Fallout is exponential in time, the instantaneous rate of fallout being that associated with a half-life of 20 days for troposphere debris (PE=factor of 1.5).

These assumptions are made about the retention of the various elements:

(1) Sr–89, 90—10% retention in skeleton (PE=factor of 3).
(2) Ba–140—5% retention in skeleton (PE=factor of 3).
(3) I–131—20% retention in thyroid (PE=factor of 2).

The infinity doses from these isotopes to the organs in question are listed in Table 5.

C. *Final Word:*

It is impractical to evaluate further these deductions about fallout and hazard from resultant radiation dose. It is clear that more accurate experimental and observational data are needed at numerous points in the chain of reasoning. However, it is also obvious that there are several qualitatively different hazards whose absolute and relative significance depends upon many factors which are not included in this study. Dominant among these factors are the conditions of debris formation (surface vs. air bursts, stratosphere vs. troposphere debris clouds, season of year), the extent of economic and sociological chaos which is contemporaneous with fallout, and philosophical judgments as to the comparative importance of radiological and other hazards of war to various segments of the world's population.

DOE ARCHIVES

TABLE 5.—INFINITY DOSE (RADS) SUBSEQUENT TO FALLOUT AT VARIOUS TIMES AFTER DETONATION
(Assumed quantity of fallout=1 KT of fission debris per mi²)

Period between detonation and fallout	External whole body gamma radiation (All individuals)	Sr-90 dose to skeleton (Infant or child)				Sr-89 dose to skeleton (Infant)				Ba-140 dose to skeleton (Infant)			I-131 dose to thyroid (Infant)		
		Soil	Leaf	Water	Air	Soil	Leaf	Water	Air	Leaf	Water	Air	Leaf	Water	Air
1 day	470	250	1.5×10^3	10	15	10	2×10^3	3.	9	1,000	2	15	7×10^5	300	3200
10 days	187	250	1.5×10^3	10	15	8.8	1.7×10^3	2.6	7.8	580	1.2	8.7	2.9×10^5	130	1360
20 days	142	250	1.5×10^3	10	15	7.7	1.5×10^3	2.3	6.9	340	0.7	5.1	1.2×10^5	53	540
40 days	107	250	1.5×10^3	10	15	5.9	1.2×10^3	1.8	5.4	120	0.24	1.8	2.2×10^4	9.2	100
70 days	85	250	1.5×10^3	10	15	4.0	0.8×10^3	1.2	3.6	22	0.044	0.33	1.6×10^3	0.7	7.2
100 days	74	250	1.5×10^3	10	15	2.7	0.5×10^3	0.8	2.4	4.5	0.009	0.06	1.2×10^2	0.05	0.4
Probable error expressed as a factor	*3*	*3*	*7*	*7*	*6*	*4*	*6*	*7*	*6*	*8*	*7*	*6*	*5*	*6*	*5*

DOE ARCHIVES

[redacted]

APPENDIX A

RADIOCHEMICAL DETERMINATIONS

Most of the determinations of Sr–90 at low concentrations in the various materials of interest have been carried out in three laboratories: Institute for Nuclear Studies, University of Chicago (W. F. Libby); Lamont Geophysical Observatory, Columbia University (J. L. Kulp); and Division of Health and Safety, New York Operations Office, U.S. Atomic Energy Commission (J. H. Harley). While the methods used by these laboratories were evolved with some degree of collaboration, differences in detail make it desirable to include here descriptions of all three. For this purpose, minor changes have been made in descriptions submitted by the respective investigators for other purposes.

I. UNIVERSITY OF CHICAGO

A. *Preliminary treatment of samples:*

Before any chemical preparation was done, most of the solid samples were dried or burned in an oven or incinerator and then reduced to ash in a muffle furnace at 900° C. Samples treated in this way included the human, alfalfa, cheese and milk specimens. The Bureau of Plant Industry at Beltsville, Md., processed the soil samples and sent them in the form of calcium oxalate.

The water samples, including the rain, snow, lake, river and ocean samples, were filtered to remove solid material.

B. *Chemical preparation of samples:*

The ashed samples were dissolved in concentrated HCl, and concentrated HNO_3 was also added if the sample did not go into solution satisfactorily. The residue, if any, was then filtered off, and, with the exception of the human group, strontium carrier was added to the solution, which was then milked with yttrium.

The alfalfa samples were still highly contaminated after the first milking. Repeated ferric phosphate precipitations were made to remove the rare earth contaminants. The strontium carrier which had been added was isolated using Harley's method (see p. 33 of this appendix), and then purified.

The soil samples in the form of calcium oxalate were dried, ignited to oxides, dissolved in concentrated HCl, diluted with water, and milked. The milking technique adopted by the Chicago group for the determination of the Sr–90 content of various types of sample essentially consists of obtaining an HCl solution of the sample, adding carrier for the yttrium, separating the Y–90 daughter of the Sr–90 present in the sample by a phosphate precipitation of the carrier, and determining the amount of Y–90 and therefore also of Sr–90 present by absolute counting.

The technique outlined above was tested and shown to yield satisfactory results using both inactive yttrium and neodymium as carriers for the Y–90 and in both the presence and the absence of calcium in the solution. The following example of a test experiment performed with neodymium carrier and in the absence of calcium serves to illustrate the details of the

[redacted]

DOE ARCHIVES

chemical procedure, the counting technique, and the calculations involved in making a Sr–90 assay by the milking technique.

2.005 g $Sr(NO_3)_2$ were dissolved in 50 ml H_2O, and to it was added approximately 22,000 dpm of an acid solution of Sr–90, Y–90 tracer in equilibrium. A few ml of 85% H_3PO_4 were next added, followed by 0.304 gm of Nd^{III} carrier in HCl solution. The solution was stirred and heated. To the hot solution 2N NH_4OH was added gradually with vigorous stirring until the precipitation of $NdPO_4.2H_2O$ was believed to be complete. ($NdPO_4.2H_2O$ and $YPO_4.2H_2O$ precipitate out completely well below pH 1, while the phosphates of calcium and strontium do not begin to precipitate until pH 3 or 4.) The precipitate was digested with heating for 15 minutes, filtered onto a 7-cm filter paper, and washed with water. To the filtrate were added a few ml of 2 N NH_4OH. No precipitation occurred, showing that the precipitation of neodymium had been complete. Additional NH_4OH was now added to the same solution until $SrHPO_4$ was precipitated completely. The $SrHPO_4$ was also filtered onto 7-cm paper. The precipitates were dried and mounted on lucite semicylinders of 1⅞″ I.D.

The counting procedure is described in Section C below. The mounted samples were counted on a gold foil covered Q-gas flow counter of 1½″ diameter and 10″ active length. The geometry factor for the position that was maintained between the counter and the sample was previously determined with use of a similarly mounted sample of KCl and was found to be 2.50.

The activities in the two samples were followed for over 10 days. The activity in the neodymium sample decayed from an initial rate of 4500 cpm with a constant half life of 61±1 hours into a very long lived tail of 102 cpm. An aluminum absorption curve for the same sample determined within a few hours after milking was found to be absolutely straight to 360 mg/cm² and gave a half thickness value of 133 mg/cm².

When the Y–90 activity was subtracted out, the aluminum absorption curve for the strontium sample, which was also determined soon after milking, also was a straight line as far as 40 mg/cm². The half thickness value was 12.0 mg/cm².

The intensities and identity of the activities in the strontium and neodymium samples corrected to milking time were as follows:

	Y–90 activity, cpm	*Sr–90 activity, cpm*
Strontium sample	263	1068
Neodymium sample	4540	102

Applying the equation,

$$D=\frac{IG\,s/s'}{1-e^{s/s'}}\,e^{S/s'}$$

where

D=The disintegration rate of the sample in dpm
I=The counting rate of the bare sample
G=The geometry factor=2.50
S=Thickness of air between sample and counter plus thickness of counter wall (sum=2.70mg/cm²)
s=Thickness of the sample in mg/cm²
$s'=\frac{s\,½}{0.693}$, where s½ is the half thickness value of the activity in mg/cm².

The following results were obtained:

	s	*s'*	*D(Y–90)*	*D(Sr–90)*
Strontium sample	60.9	17.3	658	11,320
Neodymium sample	15.1	192	10,970	447
			11,628	11,767

A very satisfactory agreement between the disintegration rates of Y–90 and Sr–90 was obtained. The recovery of the Y–90 activity from the solution is seen to be 94% and the percentage of Sr–90 carried over in the neodymium precipitate as contamination is seen to be 3.8%.

DOE ARCHIV

C. *Counting technique:*

The absolute radioactivity measurements in the Chicago laboratories have been made with cylindrical thin-wall flow counters. These are shielded with several inches of iron and with anti-coincidence counters.

The large sample counters have been built after a design by T. Sugihara, R. Wolfgang and W. F. Libby.[23] These counters have lucite end pieces connected by three thin brass rods at their periphery. Thin plastic film 2.69 mg/cm^2, with a conducting gold-coated surface, is used for the counter wall. Miniature flow counters have been constructed with field adjusting rings at the ends and with external end supports (to avoid internal rods and their field distortion). These counters have been covered with DuPont Mylar film coated on the inner side with 0.975 mg/cm^2 aluminum. The Mylar, a poly ester, is very durable, being resistant to moderate physical stress and common organic solvents. The background for the large counters (1½″ x 6″ to 10″) runs about 6 to 10 counts per minute within the shielding mentioned above. The miniature counters (1.4 x 3.0 cm) have a background of 0.4 counts per minute. (The counters with aluminum-Mylar walls are quite photosensitive, but are ordinarily operated in the dark.)

Samples are mounted on plastic half cylinders, two of which completely surround the sample counter. The geometry is as good as is permitted by the necessity for placing absorbers between the sample surface and the counter wall. For the large counters the geometry is 38% and for the small ones 33%. The corresponding factors used in conversion of cpm to dpm are 2.7 and 3.0 respectively. These geometry determinations were made using potassium–40 as a secondary standard and using the figure 28 dps/gm K. Most powered samples are mounted with the aid of dilute agar in alcohol and water, which provides a nearly weightless binder after drying.

The samples have usually been counted to a standard error of 10 to 15% where activity is sufficient. If such precision would require an extraordinarily long counting period greater error is allowed. Backscattering corrections are not included in calculations of rates of disintegration. The plastic sample mounts "reflect" only about 8% of the radiation striking them and this backscattered radiation has only about half the energy of the original radiation. Since the samples are of finite thickness, ca. 60 to 80 mg/cm^2, most of the backscattered radiation is absorbed. The backscattering contributes about 1% of the observed activity. This is at least partially corrected for by the geometry determinations where backscattering corrections were not made.

Except for water, activities are routinely reported in "Sunshine units," based on the ratio of Sr–90 to elemental calcium in the sample. One "Sunshine unit" is defined for this purpose as 1×10^{-12} curies of Sr–90 per gram of calcium.

Freedom from contamination was demonstrated by analyses of samples of soil and powdered milk obtained by the Department of Agriculture in the years 1937 and 1943, respectively. These samples gave the following results:

Soil, sample No. C–2917, leached with NH_4Ac	0 ± 0.05 S.U.
sample No. C–2916, leached with HCl after NH_4Ac	0 ± 0.05 S.U.
Powdered milk, 73.8 gms ash	0 ± 0.08 S.U.

Results on recent samples of adult leg bones, while not serving as controls, were as low as 0.01 ± 0.006 S.U. These indicate a low upper limit to possible contamination in processing this type of material.

Footnote references on pp. 42–43.

DOE ARCHIVES

[redacted]

II. COLUMBIA UNIVERSITY

A. *Chemical treatment:*

The program undertaken at Lamont Observatory required analysis for the determination of Sr–90 in a variety of sample types. The counting equipment, designed specifically for this program, dictated to some degree the kinds of samples necessary for maximum efficiency of operation. Thus chemical procedures were developed which (*a*) would be adaptable to all of the sample types anticipated, (*b*) would be rapid and quantitative, and, (*c*) would result in a precipitate suitable for the mounting technique described in the next section.

Because of its characteristic granular texture and relatively non-absorptive properties, it was decided to count the samples as yttrium oxalate $Y_2(C_2O_4)_3 \cdot 9H_2O$. Two further advantages of this compound are its high molecular weight, resulting in relatively large samples, and its precipitation at low pH (less than 1), where the oxalates of iron and other interfering cations are soluble.

The samples fall into two categories; those naturally occurring as calcium phosphate, and all other. In all cases a pure calcium salt was first prepared. Cheese, bone, and plant samples were burned to ash in stainless steel chambers, the ash dissolved in concentrated HCl, and solids filtered off. Calcium phosphate along with strontium phosphate was then precipitated by addition of H_3PO_4 and NH_4OH. Generally these were purified by several re-precipitations and filtrations after which they were dried and weighed to determine calcium content.

Soil samples were leached for five minutes in 50% HCl solution with stirring. The liquid was filtered off, evaporated to dryness and ignited to remove organic debris. The ash was then dissolved in concentrated HCl, and filtered, and $CaCO_3$ precipitated by addition of concentrated Na_2CO_3. Again several re-crystallizations were carried out for purification purposes, after which the solid was dried and weighed to determine calcium content.

Shell samples were dissolved in HCl, filtered, evaporated to dryness and ignited to remove organic matter. The salt was redissolved and $CaCO_3$ precipitated and determined as outlined for soils.

Plant samples were treated in two ways. Leach samples were prepared from plants in exactly the same manner as described for soils. After leaching, the solid material was then burned to ash and the ash similarly treated. In most cases there was insufficient calcium present for good $CaCO_3$ precipitation, so weighed $SrCl_2 \cdot 6\ H_2O$ was added as carrier and the resulting $SrCO_3$ precipitate was stored.

Water samples were treated by addition of $SrCl_2 \cdot 6\ H_2O$ as carrier and precipitation of $SrCO_3$. The water samples were not filtered before the addition of the strontium carrier for the purpose of base exchanging any adsorbed Sr–90 on the solid material carried down by the rain. A portion of each sample was used to prepare special concentrated Na_2CO_3 solution for precipitation; thus with the exception of the negligible amount of water in the $SrCl_2 \cdot 6\ H_2O$ carrier, no other water was added to the samples.

By these methods, all of the samples were purified to either phosphates or carbonates, and were then put into solution in small volumes of HCl.

B. *Milking:*

The milking procedure was done by two methods for carbonate and phosphate samples, respectively.

The carbonate samples were milked by addition of yttrium carrier in acid solution, and precipitation of yttrium hydroxide at pH of 9. The filtrate was then set aside for accumulation of Y–90 and later milking. The $Y(OH)_3$ was dissolved off the filter paper by 50% HCl, and then NH_4OH added dropwise to just recrystallize the hydroxide. One or two drops of concentrated HCl was then added to just dissolve it again, and concentrated oxalic acid then added to precipi-

[redacted]

DOE ARCHIVES

tate the oxalate of yttrium. About 15 minutes of digestion of this precipitate with heating results in a well coagulated sample, easily filterable for the mounting technique to be described.

Milking of the phosphate samples was generally analogous to that described for carbonates, except that the phosphate of yttrium was utilized as the first precipitate rather than the hydroxide. This step was somewhat more difficult since pH control was necessary to achieve separation of the yttrium phosphate from the phosphate of calcium and strontium. By slow addition of 2 N NH_4OH to the acid solution of calcium phosphate to which yttrium carrier had been previously added, with heating and vigorous stirring, complete precipitation of yttrium phosphate occurred below pH of 1.5, while negligible amounts of calcium or strontium phosphate appeared until about pH of 2.5. Thus care in the first precipitation was necessary to obtain good separation. The precipitate was filtered and the filtrate stored for later milking. The precipitate was then treated like the yttrium hydroxide, dissolved off the filter paper, recrystallized, barely redissolved, and by addition of oxalic acid, yttrium oxalate precipitated.

C. *Sample mounting:*

The technique used of mounting samples for counting was essentially that employed by the Health and Safety Laboratory, p. 32. The oxalate precipitate was filtered by using a Fisher Filtrator with the Tracerlab stainless steel sample funnel and Whatman #42 1⅛″ diameter filter paper. The precipitate was washed with distilled water and dried by the vacuum. It was found that about 5 minutes of standing in the funnel, with the vacuum pump on, sufficiently dried the samples. The mounted samples were then dropped onto brass disks (Tracerlab) covered with Pliofilm and sealed with brass rings. The resulting mount was found to be compact and sturdy and greatly simplified the counting technique.

D. *Counting method:*

Counting is performed with a 1 inch Anton flat halogen-filled Geiger counter. Shielding consists of a complete ring of cosmic ray counters in anticoincidence with the sample counter and an outer shield of lead brick. A special housing was constructed for this work, made entirely of lucite to cut background to a minimum. The sample slides under the counter window and in operation is about 2 millimeters from the window. To illustrate the efficiency of the shielding, the G.M. tube had a background at the center of its plateau of about 10 cpm when exposed on the bench. Placed in the lead shield this was cut to about 6 cpm. Activation of the anticoincidence ring has cut the background to 2 cpm at which it has remained constant over a period of about 2 months.

The counter has been standardized with mounts of yttrium oxalate prepared from calibrated standards of Sr–90 obtained from the National Bureau of Standards. Samples were prepared by both the phosphate and carbonate method and yielded an efficiency of 31.6% ± 2.0%. Eight such standard samples have been counted over periods of several half-lives and have yielded an average half-life of 64 ± 1 hours.

Later runs on these samples have shown that less than 1% of the total activity was due to Sr–90 carried through the sample preparation. For example, one sample which initially counted about 1100 cpm decayed with a half-life of 63 ± 1 hours. Again checked about 3 weeks after preparation, this sample counted 9 cpm.

E. *Outline for analytical procedures:*

a. *Procedure for the determination of Sr–90 in water samples.*

1. Add carrier strontium. ($SrCl_2 \cdot 6H_2O$ about 20 gms. per gal. water). Stir thoroughly until completely in solution.

DOE ARCHIVES

[redacted]

2. Add saturated solution of Na_2CO_3 (with mechanical stirring) until precipitation is complete. After a short time has been allowed for the precipitate to settle, test supernatant liquid for completeness of precipitation with a few drops of the carbonate.

3. Gently digest for fifteen minutes on hot plate.

4. Suction filter. Wash with distilled water.

5. Check filtrate for completeness of precipitation, with a few drops of saturated Na_2CO_3 solution. If clear, filtrate may be stored in can for other analyses.

6. Redissolve precipitate in 50% HCl solution keeping the volume to a minimum.

7. Filter to remove solid material.

8. Re-precipitate the $SrCO_3$, following steps 3 through 6, except that the filtrate may be discarded when precipitation is complete.

9. Dry and weigh precipitate.

10. Calculate yield.

11. Repeat steps 6 through 10 if yield indicates appreciable impurity.

12. Store sample in clean bottle until ready for milking as described on p. 29.

b. *Procedure for the determination of Sr-90 in soil:*

1. Sample in 2 pound batches is leached with twice the volume of 50% HCl with stirring for five minutes.

2. Filter off liquid, and save solid portion for further testing.

3. Filtrate is evaporated to dryness.

4. Ignite dry filtrate to remove organic material.

5. Dissolve ash in concentrated HCl, and filter to remove insolubles. Discard solid.

6. Precipitate, $CaCO_3$ by addition of concentrated Na_2CO_3 solution with mechanical stirring.

7. Filter and check filtrate for completeness of precipitation by addition of Na_2CO_3 solution.

8. Dry and weigh precipitate.

9. Redissolve precipitate in 50% HCl solution, filter and re-precipitate the carbonate as in step 6.

10. Filter off, dry, and weigh precipitate. This procedure of re-crystallization should be repeated until a constant weight of $CaCO_3$ is achieved, to insure maximum purity.

11. Store samples as the solid $CaCO_3$ ready for milking in the procedure described on p. 29.

c. *Procedure for the determination of Sr-90 in bone and cheese:*

1. Ignite sample in oven at about 900°C until completely ashed.

2. Dissolve ash in concentrated HCl, and filter to remove insolubles. Discard solid.

3. Precipitate $Ca_3(PO_4)_2$ by addition of a few ml. of H_3PO_3 and raising pH with NH_4OH, with mechanical stirring.

4. Filter and check filtrate for completeness of precipitation by addition of NH_4OH.

5. Dry and weigh precipitate.

6. Dissolve the weighed solid in hot concentrated HCl and repeat steps 3 through 5 until a constant weight of precipitate is obtained.

7. Store sample as solid $Ca_3(PO_4)_2$, ready for phosphate milking procedure described on p. 29.

d. *Procedure for Sr-90 determination of shell samples:*

1. Dissolve sample in HCl.

2. Filter to remove insolubles.

3. Evaporate filtrate to dryness and ignite to remove organic debris.

4. Dissolve ash in concentrated HCl.

5. Filter off insolubles.

6. Precipitate $CaCO_3$ from the filtrate by addition of concentrated Na_2CO_3 with mechanical stirring.

[redacted]

DOE ARCHIVES

7. Filter and check filtrate for completeness of precipitation by addition of Na_2CO_3 solution.
8. Dry and weigh precipitate.
9. Dissolve precipitate in 50% HCl solution and repeat steps 6 through 8 until a constant weight is obtained.
10. Store as solid $CaCO_3$, ready for carbonate milking procedure.

e. *Procedure for determination of Sr-90 in plants:*

1. Leach sample with twice its volume of 50% HCl solution with stirring for five minutes.
2. Filter.
3. Filtrate is then treated exactly as the soil leach, steps 6 through 11 on page 28. Where insufficient calcium is present for good $CaCO_3$ precipitation, a weighed amount of $SrCl_2 \cdot 6\ H_2O$ may be added as carrier in step 6.
4. Solid plant material in the filter is burned to ash and also treated exactly as the soil.
5. Samples should be stored separately as carbonates, ready for milking.

f. *Milking procedure for carbonate samples:*

1. Dissolve sample in 50% HCl, keeping volume as small as possible.
2. Add dead yttrium carrier.
3. Add concentrated NH_4OH to precipitate yttrium hydroxide completely. Add excess NH_4OH.
4. Digest with heating for about ten minutes.
5. Filter in Fisher Filtrator, using Whatman #41 or 42 filter paper. Record time of filtration.
6. Wash solid with dilute NH_4OH solution. Use small amount, to keep volume of filtrate down.
7. Remove and store filtrate for later milkings.
8. With a clean beaker in the filtrator, and the vac. off, fill funnel containing the precipitate with dilute (about 10%) HCl solution. Allow to stand for a minute or two to dissolve the precipitate and then turn on the vac. collecting the solution in the clean beaker.
9. Wash paper with small amount of dilute HCl solution. Discard paper.
10. Add concentrated NH_4OH dropwise to filtrate until first signs of precipitate appear.
11. Add concentrated HCl dropwise to beaker with stirring to just dissolve precipitate. One or two drops should suffice.
12. Add concentrated oxalic acid to solution in excess. White precipitate should quickly appear.
13. Digest with heating for about ten minutes.
14. Filter in Fisher Filtrator using steel Tracerlab precipitation funnel and Whatman #42 1⅛″ diameter paper. If filtrate is clear it may be discarded.
15. Dry precipitate by allowing vacuum to pull on it for about 5 minutes.
16. Carefully remove paper with sample and place on Tracerlab brass disk.
17. Cover with pliofilm and secure with brass ring.
18. Note sample number on bottom of brass disk.

g. *Milking procedure for phosphate samples:*

1. Dissolve weighed amount of stored sample in as small volume of concentrated HCl as possible. Heating facilitates solution.
2. Add dead yttrium carrier.
3. Precipitate phosphate of yttrium by addition of 2 N NH_4OH. Heating and vigorous stirring materially help obtain a clean precipitate. pH should not exceed 1.5 at any point in this procedure.

DOE ARCHIVES

4. Digest with heating for about ten minutes. Check pH at the end of this time. It should be between 1.0 and 1.5 for best results.

5. Filter in Fisher Filtrator using Whatman #41 or 42 filter paper. Record time of filtration.

6. Wash precipitate with distilled water.

7. Remove and store filtrate for later milking.

8. With a clean beaker in the filtrator and the vacuum off, fill the funnel containing the precipitate with hot 50% HCl. Allow to stand for about 1 minute to dissolve the precipitate and then pull through by turning on the vacuum. Repeat this procedure to thoroughly dissolve precipitate from paper.

9. Wash paper with distilled water.

10. To the filtrate, carefully add concentrated NH_4OH with vigorous stirring until precipitate just appears.

11. With continued stirring add concentrated HCl dropwise until precipitate just dissolves.

12. Add concentrated oxalic acid in excess. The white oxalate of yttrium should appear.

13. Digest with heating for about ten minutes.

14. Filter in Fisher Filtrator using Tracerlab precipitation funnel and Whatman #42, 1⅛" diameter paper.

15. Dry by allowing vacuum to pull on paper for about five minutes.

16. Carefully remove paper and fit on Tracerlab brass disk.

17. Cover with pliofilm and secure with brass ring.

18. Note sample number on bottom of brass disk.

F. *Details of counting equipment:*

The new counting system designed for project SUNSHINE is pictured in Figure 1, with the outer housing and lead shielding removed. In use, the lucite counter housing seen in front of the main unit is within the anticoincidence ring and moves out for sample changing by sliding along the tracks seen at the open end of the ring.

The actual counter utilized is an Anton flat Geiger counter model No. 1007 with a 1 inch diameter mica window. The counter is seated within the lucite housing and power is supplied through the single cable which is wired at its other end to the front amphenol connector on the base. Careful machining of the lucite has resulted in the mica window of the counter being at a constant distance of about 2 millimeters from the top of the sample.

The samples which are mounted on brass disks fit into the receptacle in the lucite slide of the housing. A stop on the slide seats the sample accurately under the counter window when the slide is pushed in as far as possible.

Cosmic ray shielding is accomplished by means of the ring of cosmic ray counters in anti-coincidence with the sample counter. The tubes employed are Radiation Counter Laboratories Model 52–12 cosmic ray counters. The overall lengths of these tubes are 15⅛ inches, the active anode length is 12 inches. The pig tails on the far end of these tubes are connected in parallel by a copper ring which is in turn connected to the rear amphenol connector seen in the figure.

In counting position, the lucite housing is seated in the center of the A.C. ring, the long handle projecting about 2 inches from the front of the unit. By merely pulling the handle, the sample slide moves forward and the lucite housing emerges from the ring. Sample changing is thus a rapid and simple procedure, neither cables to be disconnected, nor voltages turned down.

The plateau for this counter has been found to be about 100 volts long with a slope of about .1% per volt. Optimum operating voltages are 625 volts on transmit and 900 volts on reject. The background of 2 counts per minute has been constant for about 2 months since the counter has been built.

DOE ARCHIVES

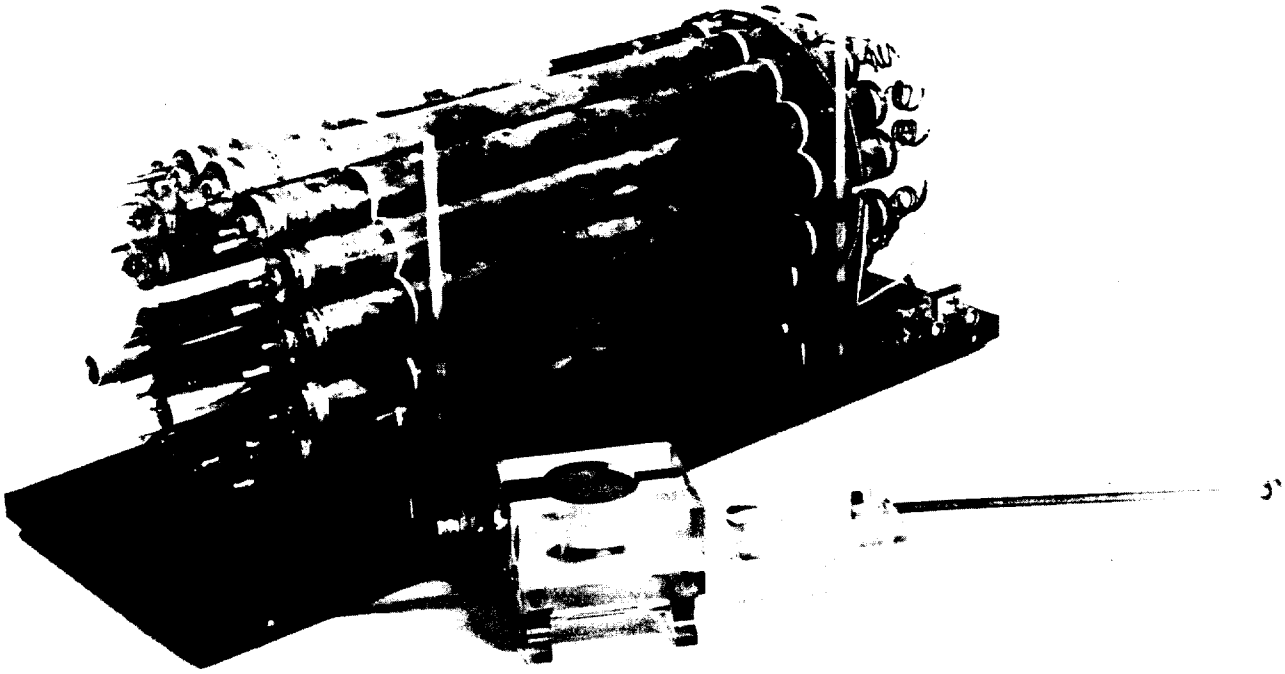

FIGURE 1

III. NEW YORK OPERATIONS OFFICE, AEC

Analytical methods for determining total strontium, total radiostrontium, and Sr–90 are discussed. The majority of the samples encountered were bone, soil, vegetation, or foods. Where different procedures are required for handling these different types of samples, the methods are given separately.

A. *Separation of total radiostrontium from five grams of bone ash:*

1. Ash the bone sample in nickel crucible at 900°C.
2. Grind in a mortar to a fine powder.
3. Weigh out 5 grams into a 250 ml centrifuge bottle.
4. Add 44 ml of water and then slowly add 154 ml of 90% nitric acid to bring concentration to 75% HNO_3.
5. Add 20 mg of Sr carrier (as $Sr(NO_3)_2$) in 2 ml of solution.
6. Stir rapidly for thirty minutes. (Mechanically)
7. Centrifuge for 10 minutes at about 2,000 rpm.
8. Decant, dissolve the precipitate in 23 ml of water and transfer to a 250 ml beaker, add slowly 77 ml of 90% HNO_3 and stir mechanically for thirty minutes.

DOE ARCHIVES

9. Filter through a fluorothene funnel on to a glass fiber filter. Transfer the funnel to a clean filter flask and wash the acid from the precipitate with anhydrous ethyl ether.

NOTE.—The fluorothene funnels are a modification of the Tracerlab stainless steel funnel, designed for preparing precipitates for counting on filter paper. The glass fiber filters are available from H. Reeve Angel and Co., Inc., 52 Duane St., New York 7, N. Y., as catalog number X-934-AH. They are more retentive than paper filters and are unaffected by 75% HNO_3.

10. Place the filter paper on a brass disk and ring assembly (Tracerlab), cover with 0.001 inch Pliofilm and beta count.

One of the standard methods for separating strontium from calcium and many other metals is the precipitation of strontium in strong nitric acid. By the addition of carrier strontium, this method has been adapted to the determination of radiostrontium. Strontium, barium, and lead are relatively more insoluble as the concentration is increased. Since other nitrates also become more insoluble with increased concentration, the selection of the strength to be used represents a compromise. Based on experimental studies, 75% HNO_3 was selected as the optimum concentration.

In the analyses 20 mg of carrier strontium is added to each sample before starting the procedure. This may be an excessive quantity, but this amount of carrier can be tolerated in beta counting and a large excess is helpful when dealing with moderately soluble precipitates such as the strontium salts used.

An experiment was performed to determine the effect of calcium on the recovery of Sr-90. Solubility measurements indicated that 200 ml of 75% HNO_3 would cause a loss of 2.6 mg of Sr or a 13% loss when using 20 mg of Sr carrier. Calcium was found to have a salting effect so that Sr recovery is improved by operating close to calcium saturation. The solubility of calcium in 75% HNO_3 is equivalent to 23.5 grams of CaO per liter. If no calcium is present, a minimum of HNO_3 should be used.

Experience showed that the presence of other fission products does not interfere in the method of analysis used for bone samples.

B. *Separation of total radiostrontium from one hundred grams of soil:*

The method described below is designed to insure recovery of the radiostrontium regardless of its state or of the nature of the soil. However, tests of leaching methods, using 6 N HCl or 1 N ammonium acetate, have shown good recovery and are to be preferred for handling large samples of soil. Details of this method are included here partly because the method is being used for certain cases and partly because of the applicability of a portion of the procedure to separation of radiostrontium from samples of vegetation and foods.

1. Dry the entire sample in an oven at 110°C.
2. Sample, using standard sampling procedure (quartering process) until approximately 125 grams of soil is obtained as a representative sample.
3. Ignite in a nickel crucible at 900°C.
4. Weigh out 100 grams of the ignited material.
5. Add 400 grams of Na_2CO_3 and mix thoroughly.
6. Fuse in a muffle at 900°C (minimum temperature) for 45 minutes (minimum time), until the melt is clear and homogeneous.
7. Cool by immersing in a beaker of cold water, withdrawing quickly at first. Continue immersing and withdrawing until the crucible is cool enough to place on a transite board. The Na_2CO_3 should not be wet at any time.
8. When the crucible is cool, turn it upside down, tap the bottom with a pestle and allow the solid fused material to fall into a mortar.
9. Break up the fused material in a hand grinder, transfer to an automatic grinder and grind to a fine powder.

DOE ARCHIVES

10. Transfer the ground fused material to a 3-liter beaker containing 500 ml of hot distilled water, stirring constantly until the particles are dispersed.

11. Add slowly with continued stirring, 1000 ml of 60% $HClO_4$. This is sufficient to neutralize the fused material and allow for an excess of acid.

12. Evaporate with mechanical stirring until dense white $HClO_4$ fumes are detected. The silica is now dehydrated.

13. Dilute to 2.5 liters with distilled water and allow to settle.

14. Filter the silica on No. 5 Whatman paper with suction. Wash with 250–500 ml of hot 5% HCl, then with 250 ml of distilled water. Transfer the filtrate and washings to the original beaker. Wash the silica with 1% H_2SO_4, and discard the washings.

15. Remove the silica from the filter paper and dry in a 110°C oven overnight. Break up the aggregated material with a mortar and pestle, then transfer to a 250 ml platinum dish.

16. Moisten the silica with 10% H_2SO_4, then add about 100 ml of HF.

17. Evaporate on a sand bath to SO_3 fumes to volatilize the silica. Cool and dilute with distilled water, transferring the resulting solution to the original soil filtrate, and add 20 mg Sr^{++} carrier.

18. Neutralize the filtrate with NaOH pellets until the pH is 4–4.5. At this point add slowly, with stirring, 50 grams of Na_2CO_3 and allow the precipitate to settle. Filter through No. 5 Whatman paper with suction and discard the filtrate.

19. When the precipitate is dry, remove from the filter paper and transfer to the original beaker. Dissolve in 200 ml of concentrated HCl and evaporate slowly to dryness. Do not bake.

20. Now add 460 ml of distilled water and stir mechanically until the residue is completely dispersed.

21. Add slowly, 1540 ml of 90% HNO_3 with continuous mechanical stirring and allow to remain stirring for ½ hour.

22. Allow to settle until the supernatant is clear, then decant as much 75% HNO_3 as possible.

23. Transfer the precipitate and remaining acid to a 250 ml centrifuge bottle, centrifuge and decant, discarding the supernatant.

24. Carry out another 75% HNO_3 separation using a volume of 200 ml.

25. At this point the bulk of the calcium should be removed leaving strontium nitrate and insoluble material. Add 100 ml of hot distilled water to the precipitate, centrifuge and decant into a 250 ml beaker.

26. Wash again with 50 ml of hot distilled water adding the supernatant liquid to the beaker after centrifugation. The residue can now be discarded.

27. Evaporate the filtrate slowly to dryness.

28. Now carry out a final 75% HNO_3 separation in the 250 ml beaker. (100 ml volume)

29. Filter through a fluorothene funnel on a glass fiber filter. Transfer the funnel to a clean filter flask and wash out the acid with anhydrous ethyl ether.

30. Place the filter on a brass disk and ring assembly (Tracerlab), cover with 0.001 inch Pliofilm and beta count. Recovery values for the above procedure are about 75%.

C. *Separation of total radiostrontium from vegetation and foods:*

1. Ash the sample in a nickel crucible at 900°C.
2. Estimate the amount of ash, add four times this weight of Na_2CO_3 and fuse.
3. Dissolve, remove silica, and collect the strontium by carbonate precipitation as for soil.
4. Run a double nitrate precipitation as for bone using acid volumes of 200 ml for the first precipitation and 50 ml for the second.

DOE ARCHIVES

D. *Determination of elemental strontium in bone.*

The simplest method of analysis is the direct solution of bone ash in HNO_3, dilution to suitable volume, and determination of strontium content by flame photometry. The direct analysis did not have sufficient sensitivity, so the separation of strontium from calcium was made by a double nitrate precipitation. At the low concentrations of strontium found in bone it was necessary to add barium as a carrier. This brought the recovery of strontium up to over 90%.

The analytical procedure is as follows:

1. Ash the bone sample in a nickel crucible at 900°C.
2. Grind in a mortar to a fine powder.
3. Weigh out 5 grams into a 250 ml centrifuge bottle.
4. Add 44 ml of water and then slowly add 154 ml of 90% nitric acid to bring concentration to 75% HNO_3.
5. Add 20 mg of Ba^{++} carrier (as $BaCl_2$) in the 2 ml of solution.
6. Stir rapidly for thirty minutes (mechanically).
7. Centrifuge for ten minutes at about 2000 r.p.m.
8. Decant and repeat steps 4 thru 7 at ½ the original volume.
9. Decant as much of the liquid as possible and transfer to a 100 ml beaker with water.
10. Evaporate to dryness and pick up with 10 ml of concentrated nitric acid.
11. Add 5 ml of 1000 ppm Li solution, dilute to 100 ml and run flame photometry.

E. *Determination of Sr-90 by separation and counting of Y-90:*

1. After the sample on the brass holder has been counted for total strontium and has reached equilibrium, it may be used for Y-90 separation.
2. Count the sample and record data.
3. Remove the sample from the brass holder. Wash the holder with about 25 ml of water. Allow washings to fall into beaker with the bulk of the paper.
4. Bring to a boil and filter thru No. 41 paper into a 50 ml platinum dish. Wash the beaker and paper with approximately 15 ml more of water.
5. Evaporate to dryness on a sand bath. Add approximately 20 ml of HF and evaporate to dryness again.
6. Pick up the residue with approximately 5 drops of HCl. Transfer to a beaker with water. Bring volume to about 50 ml. Add 10 mg of lanthanum carrier. (This carrier must have negligible beta activity.)
7. Bring to a boil and adjust pH to 6–7. Allow to stand for about 15 minutes and filter thru a 1⅛″ glass fiber paper. Place on brass holder. Cover with Pliofilm and count.
8. Record time that filtration was completed. Count and record counting date and counting time.
9. Observe decay by counting at approximately 3-day intervals for a period of about 12 days.

The procedure was tested by measuring the recovery of Y-91 added to solutions containing 20 mg of Sr as the nitrate. The recoveries found with either paper or glass fiber filters were 94–99%.

F. *Measurement of activity:*

Instrumentation for counting is similar to that used by Kulp (p. 30), except that it has no anticoincidence circuit. The background counting rate is about 6 c.p.m.

Work on the development of a scintillation anticoincidence counter is in progress. This is designed around a solution phosphor containing the active material in a volume of 5 to 10 ml.

DOE ARCHIVES

The overall problem involves the development of a suitable method of obtaining the active material in solution, the development of the optimum phosphor, and the development of appropriate instrumentation.

IV. DEPARTMENT OF AGRICULTURE

A. *Procedure for the extraction of exchangeable calcium in soils:*

1. *General Statement:*

The soil sample is leached with ammonium acetate and the filtrate evaporated to dryness. The salts are dehydrated and the residue ashed at 600°C. The ashed residue is taken up in HCl, and iron and aluminum are removed by precipitation as the hydroxides. The filtrate is acidified and calcium precipitated as the oxalate at pH 6.2.

2. *Reagents:*

Normal ammonium acetate, pH 7.0.
Hydrogen peroxide, 30–35 percent.
Hydrochloric acid, concentrated.
Ammonium hydroxide, 1:1.
Ammonium chloride, 2 percent aqueous solution.
Methyl red, 0.1 percent aqueous solution.
Oxalic acid, C.P., crystal.

3. *Procedure:*

Crush the air-dry sample with a wooden rolling pin and pass the sample through a 2 mm sieve. Place 7–10 pounds of the prepared sample in a 4-liter beaker and add sufficient ammonium acetate to cover. Stir the suspension well and let stand overnight. Fit an 18-liter carboy with a 10-inch Buchner funnel and vacuum tube. Place two thicknesses of coarse filter paper in the funnel, moisten, and apply suction. Transfer the contents of the beaker to the funnel and leach the soil with ammonium acetate until a total of 1 liter of leachate for each pound of soil is obtained. Transfer the leachate to large beakers (3 or 4 liter capacity) and evaporate almost to dryness at 90–95°C. Combine the residues in a 2-liter beaker and take to complete dryness.

Loosen as much of the dry residue as possible with a large spatula and transfer to an evaporating dish (or dishes). To the small amount of residue remaining in the beaker add 200 ml of water and 10 ml of H_2O_2. Cover and boil for 15–20 minutes. Place the evaporating dish (containing the residue) in a muffle at about 150°, heat to 600°C, and hold this temperature for about 1 hour. Remove the dish from the muffle and cool. Combine the ashed residue and the peroxide treated residue.

Add small portions of MCl with stirring until all of the carbonates are decomposed; add 10–15 ml of HCl in excess.

Heat the solution to about 90°C and add 1:1 NH_4OH with stirring until a faint odor of ammonia persists. Boil for 2–3 minutes to coagulate the precipitate and filter through coarse filter paper. Wash the filter well with 200 ml of hot NH_4Cl. Add 20 drops of methyl red and acidify with HCl. Add 5–10 ml excess HCl. Heat to 90°C and add 50 grams (25 gms. if soil is low in calcium) of oxalic acid with stirring until the solution turns a light yellow. Digest at 90–95°C for 1–2 hours. Collect the precipitate on a Buchner funnel, wash 5 times with cold water and dry.

DOE ARCHIVES

APPENDIX B

Sr and Ca Metabolism

Reference	Animal	Age at admin.	Period of retention	Method of admin.	Sr intake	Ca intake	% Sr retention	% Ca retention	Sr/Ca ratio retained	Comments
1	Mice	.....	24 hrs	I.V (lactate)	1.6 mg	.....	33%	.....	....	IV uptake of Sr chloride, lactate, gluconate essentially identical.
					0.017	.....	45%	.....	0.8	
						0.8 mg	.....	58%		
			48 hrs	Oral (lactate)	.....	.....	11%	.....	0.5	Oral uptake of Sr chloride, lactate, gluconate essentially identical.
						1.6 mg	.....	23%		
2	Mice	Adult	9–15 days	Injected	.....	.....	13–31%	53–65%	0.4	These mice were pregnant females which gave birth during period of retention.
3	Rats	Approx. 5 weeks.	3 days	Oral (lactate)	0.5–2 mg	1.5–4 mg	10%	15%	0.7	Rats reared 15–20 days (shortly after weaning) on Ca deficient diet then treated.
				Oral (lactate with vitamin D).	...*do*	...*do*	15%	31%	0.5	
				I.P (lactate)	...*do*	...*do*	11%	28%	0.4	
				I.P (lactate with vitamin D).	.....	.....	26%	45%	0.6	
4	Rats	Growing	4 days	Oral	Carrier free.	(Low Ca diet).	73%	.....	....	Effect of dietary Ca on Sr retention is apparent, though data not quantative.
					...*do*	(High Ca diet).	17%	.....	....	
		Adult	...*do*	.....	...*do*	(Low Ca diet).	16%	.....	....	
					...*do*	(High Ca diet).	2.8%	.....	....	
		Growing and adult.	...*do*	I.P	...*do*	Similar results, ranging from 70% Sr retention in young (low Ca) to 14% in old (high Ca).				
5	Rats	50 days	4 days	Intramuscular.	.....	.....	70%	81%	0.9	Sr and Ca in different rats.
		50 days	4 days	....*do*	Carrier free.	.....	65%	75%	0.9	Sr and Ca in same rats (double tracer).
6	Rats	.....	1 day	I.V	.....	.....	81%	86%	0.9	Ra studies carried out in same series; retention at successive periods 73%, 58%, 55%, 53%, respectively.
			3 days	....*do*	.....	.....	81%	88%	0.9	
			10 days	....*do*	.....	.....	67%	87%	0.8	
			30 days	....*do*	.....	.....	55%	79%	0.7	
7	Rats	8 weeks	8 days	I.P	Carrier free.	.....	72%	.....	1	
				I.V	.....	.....	.....	73%		

DOE ARCHIVES

7 (*Con.*)	Rats	6–12 mos.	8 days	I.P.	Carrier free.	.....	29%	.....	0.9	
						1.2 mg	.....	32%		
		8 weeks (low P rickets).	8 days	I.P.	Carrier free.	1.2 mg	20%	13%	1.5	Rats in this group on low P diet, therefore rachitic.
8	Rats	Daily diet from birth.	Months	Oral	Normal Sr/Ca ratio ($\sim 10^{-3}$).		.....	.....	0.35	Rats kept on standard diet of known Sr/Ca ratio throughout life.
9	Rats	Growing	Weeks	Oral	Sr/Ca intake $= 2.3 \times 10^{-3}$.		Sr/Ca retained $= 5.5 \times 10^{-4}$.		0.25	Rats kept on standard diet of known Sr/Ca ratio throughout period of growth.
10	Rats	Adult	4 days	Oral	0.45 mg/day.	.....	4–5%	.....	....	Dose fed for 7 consecutive days, animals sacrificed on 8th day.
					0.9	.....	4–5%	.....	....	
					4.5	.....	4–5%	.....	....	
					22.5	.....	4–5%	.....	....	
					45	.....	4–5%	.....	....	
11	Rats	Adult	1 day	I.P.	0.1–75 mg	.....	See comment.	.....	....	Sr retention decreased by factor of only 5 over 750 fold range of Sr injection.
12	Rabbit	6 weeks	9 days	I.V.	Carrier free to 0.02 gm Sr per rabbit.	0.02 gm Ca/day in diet.	55%	.....	....	Amount of Sr carrier had no influence on Sr retention within experimental error of 10%. Effect of dietary Ca on Sr retention is apparent, though data not quantitative since uptake of Ca from gut is unknown.
						0.2 gm Ca per day in diet.	46%			
						1.3 gm Ca per day in diet.	31%			
		6 months	9 days	I.V.	*do*	0.04 gm Ca/day in diet.	57%	.....	....	
						0.4 gm Ca/day in diet.	17%	.....	....	
						2 gm Ca/day in diet.	12%	.....	....	
		18 months	9 days	I.V.	*do*	0.4 gm Ca/day in diet.	8%	.....	....	
13	Steer	1 year	7 days	Oral	Carrier free.	.....	30%	30%	1	Retention computed by subtracting excretion from administration.

DOE ARCHIVES

APPENDIX C

Sr–89, 90 activity of animals and animal products (1 Sunshine Unit=10^{-12} curies/gm Ca)

Sample	Date of Collection	Place of Collection	No. of Items	Sr-89, 90 content (Sunshine Units)	Sr-90 content (Sunshine Units)	Analyst	Comment
Milk	Oct. 1953	Chicago milk-shed.	From 10 farms.	...	Av.=1.5 (range 0.97–2.25).	Libby	Cows feeding on alfalfa.
Milk	1953	U.S.	2	Av. 1.8 S.U. as of 1 Jan. '54.	...	Harley	Sr–90 not measured; data on other samples suggest Sr–90 30–50% of total.
Milk	Sept.–Dec. '53	N. Y.	3	Av. 2.4 S.U. as of 1 Jan. '54.	...	Harley	
Cheese	July, '53	Wisconsin	2	...	1.2, 2.1	Libby	
Cheese	July, '53	Wisconsin	1	...	0.5	Kulp	
Cheese	Early '53 (probably)	Switzerland	2	...	1.2, 2.7	Libby	
Cheese	Early '53 (probably)	Denmark	2	...	1.0, 1.1	Libby	
Cheese	1953	Japan	2	...	0.11, 0.14	Libby	
Cheese	Early '53	Azores	1	...	0.7	Kulp	
Cheese	Early '53	Norway	2	...	1, 2	Kulp	
Calf	Sept. '53	Albany, N. Y.	1	45 (as of 1 Jan. '54).	3.0	Harley	Most of fallout in this area in Apr. '53.
Lamb	Oct. '53	Logan, Utah	1	7 (1 Jan. '54)	0.8	Harley	
Calf	Sept. '53	Tifton, Georgia	1	25 (1 Jan. '54)	14	Harley	
Lamb	Sept. '53	Ithaca, N. Y.	1	8 (1 Jan. '54)	1.2	Harley	
Lamb	Sept. '53	Rutgers, N. J.	1	8 (1 Jan. '54)	1.1	Harley	
Calf	Nov. '53	Easton, N. Y.	1	...	2	Harley	Same 8 months old calf analyzed by all 3 laboratories. Further intercomparisons underway.
Calf	Nov. '53	Easton, N. Y.	1	...	6.2	Libby	
Calf	Nov. '53	Easton, N. Y.	1	...	0.45	Kulp	
Calf	Oct. '53	Wisconsin	1	...	0.26	Kulp	
Calf	Aug. '53	Montana	1	...	0.30	Kulp	Calf 6 months old.
Human stillborns	July '53–Jan. '54	Chicago	55	...	0.14 (range 0.04–0.32).	Libby	
Human stillborns	Feb. '54	Utah	1	...	0.19	Libby	
Human stillborns	Jan. '54	Vellore, S. India	3	...	0.05, 0.04, 0.04	Libby	
Human legs (adult)	Nov., Dec. 1953	Massachusetts	3	...	0.01, 0.01, 0.02	Libby	

DOE ARCHIVES

APPENDIX D

Experimental Data on Radioactive Strontium Toxicity

Reference	Animal	No. of animals	Isotope	μc/gm administered	Estimated rad/day in skeleton	Biological effects	Comments
37	Human	6	Sr^{89}	0.02–0.4	3–60	All died soon after treatment, presumably from disease for which treated (breast cancer, polycythemia vera, osteogenic and other sarcomas). Effects of radiation indistinguishable from disease.	Patients treated at U. of Calif. hospital in early '40's. In many of the cases, the dose was administered in several subdivisions over a period of a few months. Other patients treated on which data at present unavailable. Surviving patients should be followed, although with little expectation of significant findings.
	Human	1	Sr^{89}	0.06	10	Mycosis fungicides diagnosed. Effects indistinguishable from symptoms of disease. Survives without follow-up.	
	Human	1	Sr^{89}	0.1	16	Fibromyxosarcoma diagnosed. Effects indistinguishable from symptoms of disease. Survives apparently without ill effects.	
38	Dogs	24	Sr^{89} plus small Sr^{90} contamination.	0.1–3.1	16–500	30 day LD50 about 1μc/gm retained. Biological response was acute radiation syndrome.	Dogs included adults, young adults, pups. Somewhat better correlation of effect with injected dose than with retained dose.
39, 40	Dogs (adult)	9	Sr^{89} plus small Sr^{90} contamination.	0.25–1.0	40–160	4 died within 135 days, all suffered early hemorrhages, survivors had numerous bone fractures. 1 with maximum dose died at 8 yrs. with skeletal changes, no tumor. 1 with 0.5μc/gm has bone tumor at 8 yrs.	Some survive at 8½ yrs.
	Dogs (adult)	4	Sr^{89} plus small Sr^{90} contamination.	0.1	16	1 suffered bone fracture at 3 yrs., slight localized changes in mandibles.	Some survive at 8½ yrs.
	Dogs (puppy)	2 or more	Sr^{89} plus small Sr^{90} contamination.	0.5	80	Fractures, loss of teeth, underdevelopment of mandibles. Tumor in 1 dog.	Alive at 8 yrs.
	Dogs (adult)	1	Sr^{90}	0.25	70	Died 18 days	
		1	Sr^{90}	0.15	50	Acute radiation syndrome, survived more than 3 months.	Retention not measured.
		3	Sr^{90}	0.01	3	1 suffered fracture, 2 no lesions at 6 years.	Retention not measured.

DOE ARCHIVES

APPENDIX D (Continued)

Reference	Animal	No. of animals	Isotope	μc/gm administered	Estimated rad/day in skeleton	Biological effects	Comments
41	Dogs (beagles).		Sr^{90}	.025	7.5		Experiment started spring 1954.
42	Lambs	4	Sr^{90}	(0.25 retained).	75		Experiment started spring of 1954
		4		(0.05 retained).	15		
		4		(0.005 retained).	1.5		
		4		(10^{-4} retained).	0.03		
43	Monkeys	2	Sr^{90}	2.0	1,000	Died at 13–56 days, acute radiation syndrome.	Chronic toxicity experiments promised.
		4		1.0			
		2		0.5			
44	Goats	14	Sr^{89}	(1.–1.5)	150–250	30 day LD50 about 1–1.5/μc/gm. injected, 50% retained.	
45	Steer	1	Sr^{90}	2×10^{-3}	0.5	Suggestion of hemorrhagic condition under epiphyseal plate several months after active region left behind plate as bone grew. Similar condition observed in some inactive controls.	
46	Mice		Sr^{89}	(3.5 retained)	1,200	30 day LD50	
				(2 retained)	660	150 day LD50	
	Rats			(2 retained)	660	30 day LD50	
				(1 retained)	330	150 day LD50	
	Rabbits			(0.9 retained)	330	30 day LD50	
				(0.85 retained)	280	150 day LD50	
47	Mice		Sr^{89}	0.07	12	Reduction in heterophils apparent	
	Rats			0.25	40	Reduction in heterophils apparent	
	Rabbits			1.	160	Reduction in heterophils apparent	
48, 40	Rats	19	Sr^{89}	0.7	120	Tumors in 10 of 19 rats	
				0.03	5	50% with bone damage.	
	Rabbits			2	330	Tumors within 1 yr	

DOE ARCHIVES

48, 49.	Mice	2,100 altogether.	Sr^{89}	5	820	Mean life duration about ⅓ control.	The most extensive experiment on chronic Sr toxicity. Tumor latent period appears to vary as inverse square root of dose. Dependence of incidence on dose suggests that each quantity of absorbed radiation confers on tissue absorbing it a probability of tumor formation which is without limit in time.
				2.5	410		
				1	160	Significant reduction in mean life duration.	
				0.5	80	Negligible reduction in mean life duration.	
				0.25	40		
				0.1	16		
				0.05	8	Significantly higher tumor incidence than controls at this and all higher dose levels.	
50.	Humans: x and gamma irradiation of bone produce malignant tumors. Incidence apparently low. 11 cases reviewed, mostly 200 KV. x-ray treatment. In most cases, estimated bone dose about 8,000–10,000 rads. One case about 5,000 rads. Latent period 6–22 years.						

DOE ARCHIVES

[redacted]

REFERENCES

[1] Smith, N.M., First revised report of the Project Gabriel (12 Nov., 1949) (S)

[2] Smith, N.M., Gabriel Project, Reopened (8 Nov., 1951) (S)

[3] Greenfield, N.M., Kellogg, W.W., Krieger, F.J., and Rapp, R.R., R-265-AEC, the Rand Corporation (July 1, 1954). "Transport and early deposition of radioactive debris from atomic explosions, Project Aureole". (S)

[4] Kramish, et al, R-251-AEC, the Rand Corporation (Aug. 6, 1953). "World-wide effects of atomic weapons, Project Sunshine". (S)

[5] Libby, W.F., Project Sunshine Bulletin #7 (April 1, 1954) (OUO)

[6] Kulp, J.L., Project Sunshine, Annual Progress Report (April 15, 1954). (S)

[7] Eisenbud, M., NYO-1576 (Jan. 28, 1952). "Radioactive debris from operations Buster and Jangle: observations beyond 200 miles from the test site". (S)

[8] List, R.J., NYO-4512 (Feb. 25, 1953). "Radioactive debris from operations Tumbler and Snapper, part II". (S)

[9] Eisenbud, M., NYO-4522 (April 28, 1953). "Radioactive debris from operation Ivy". (S)

[10] Hubert, Machta, and List, NYO-4555. "Meteorological analysis of the transport of debris from operation Ivy". (S)

[11] List, R.J., NYO-4602, In Press. "Transport of atomic debris from operation Upshot-Knothole". (S)

[12] Eisenbud, M. and Harley, J.H., NYO-4571 (Jan. 8, 1954). "Project Sunshine—Progress from Sept. 1953–Jan. 4, 1954." (S)

[13] Alexander, G.V., Nusbaum, R.E., and MacDonald, N.S., UCLA-293 (May 20, 1954). "Sr and Ca in municipal water supplies, their occurrence and removal". (U)

[14] Weiss, M.M., BNL-252 (T-39). "A study of the airborne radioactivity at Brookhaven National Laboratory from the Nevada tests, Mar.–June, 1953". (OUO)

[15] Collander, R., Plant Physiology *16* 691 (1941). "Selective absorption of cations by higher plants". (U)

[16] Neel, Olafson, Steen, Gillooly, Nishita, and Larson, UCLA-247. "Soil-plant interrelationships with respect to the uptake of fission products: I. The uptake of Sr-90, Cs-137, Ru-106, Ce-144, and Y-91". (U)

[17] Menzel, R.G. and Brown, I.C., Bimonthly report from U.S. Department of Agriculture (March–April, 1953). "Leaching of fallout and plant uptake of fallout". (U)

[18] Rediske, J.H. and Selders, A.A., HW-25021, Biology Research—annual report 1951, p. 118 (1952). "The absorption and translocation of several fission products by plants". (U)

[19] Selders, Rediske, and Palmer, HW-28636 (July 7, 1953). "The absorption and translocation of radioactive elements from Jangle soil". (S)

[20] Tipton, Foland, Bobb, and McCorkle, Unpublished report (Mar. 11, 1953). "Spectrographic determination of trace elements in human tissue". (U)

[21] Hodges, et al, J. Biol. Chem. *185* 519 (1950). "The Sr content of human bones". (U)

[22] Looney, W.B., Unpublished report (June, 1954). "Late effects (25–40 years) of the early medical and industrial use of radioactive materials". (U)

[23] Sugihara, Wolfgang, and Libby, Review of Scientific Instruments *24* 511 (1953). (U)

[24] Pecher, C., University of California Publications in Pharmacology *2* #11 117–150 (1942). University of California Press. "Biological investigations with radioactive Ca and Sr", "Preliminary report on use of radioactive Sr in the treatment of metastatic bone cancer". (U)

[25] Pecher, C. and Pecher, J. Proc. Soc. Exp. Biol. *46* 91–94 (1941). "Radio-calcium and radio-strontium metabolism in pregnant mice". (U)

[26] Greenberg, D.M., J. Biol. Chem. *157* 99 (1945). "Tracer experiments with radioactive Ca and Sr on the mechanism of vitamin D action in rachitic rats". (U)

[27] Copp, D.H., Axelrod, D.J. and Hamilton, J.G., Am. J. Roent and Ra Therapy *58* 10 (1947). "The deposition of radioactive materials in bone as a potential health hazard". (U)

[28] Copp, et al, UCRL Quarterly Progress Report (April–June, 1948)

[29] Norris, W.P. and Kisieleski, W., Cold Spring Harbor Symposia on quantitative Biology *13* 164 (1948). "Comparative metabolism of Ra, Sr, and Ca". (U)

[30] Copp, D.H., Hamilton, J.G., Jones, D.C., Thomson D.M., and Cramer, C., UCRL-1464 (Sept. '51). "The effect of age and low P rickets on calcification and the deposition of certain radioactive metals in bone". (U)

[31] Comar, Personal communication.

[32] MacDonald, Nusbaum, Stearns, Ezmirlian, McArthur, and Spain, J. Biol. Chem. *188* 137 (1951). "The skeletal deposition of non-radioactive Sr". (U)

[33] Gross, W.J., Taylor, J.F., and Watson, J.C., UCLA 274 (Jan. 6, 1954). "Some factors influencing the metabolism of radio Sr by animals". (U)

[redacted]

DOE ARCHIVES

[redacted]

[34] MacDonald, Nusbaum, Ezmirlian, Barbara, Spain and Rounds, Arch. Biochem. Biophysics, *43* 118 (1953). "Mechanisms in skeletal accumulation of ions". (U)

[35] Kidman, Tutt, Vaughan, J. Pathology and Bacteriology *62* 209 (1950). "The retention and excretion of radioactive Sr. and Y (Sr^{89}, Sr^{90}, Y^{90}) in the healthy rabbit". (U)

[36] Comar, ORO–110 (1954) (U)

[37] Letter from J.G. Hamilton to Dunham dated Apr. 6, 1954. (S)

[38] Swift, M.N., and Prosser, C.L., MDDC 1388 (Aug. '47). "The excretion, retention, distribution and clinical effects of Sr^{89} in the dog". (U)

[39] Finkel and Clark, ANL 5200 (1954). (U)

[40] Brues, Notes for Sr toxicity conference at Washington, Jan. 1954. (U)

[41] Arnold, James, Personal communication. (U)

[42] Comar, Personal communication. (U)

[43] Edington, et al, Nature *172* 122 (1953). (U)

[44] Swift, M.N., Prosser, C.L., and Mika, E.S., AECU 108, (June '46). "Effects of Sr^{89} and x radiation on goats". (U)

[45] Rust, Colonel John., Personal communication. (U)

[46] Anthony, D., Lathrop, K., and Snyder, R., MDDC 1364 (Aug. '47). "Radiotoxicity of injected Sr^{89} for rats, mice, and rabbits. Part III. Lethal action and clinical changes". (U)

[47] Simmons, E.L., and Jacobson, L.O., MDDC 1387 (May '47). "Radiotoxicity of injected Sr^{89} for rats, mice, and rabbits. Part IV. The hematological effects of enterally and parenterally administered Sr^{89} in mammals". (U)

[48] Brues, A.M., Lisco, H., and Finkel, M.P., MDDC 145 (1946). "Carcinogenic action of some substances which may be a problem in certain future industries". (U)

[49] Brues, A.M., J. Clin. Investigation *28* 1286 (1949). "Biological hazards and toxicity of radioactive isotopes". (U)

[50] Cahan, Woodward, Higinbothan, Stewart, Coley, Cancer *1* 3 (1948). "Sarcoma arising in irradiated bone". (U)

[redacted]

DOE ARCHIVES